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Effects of Nano-sized Particles on Mechanical Properties of Resin Dental Composites: A Review Article

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

Nanomaterials have unique physical and mechanical characteristics. The addition of nanoparticles advances the mechanical performance of polymers, because the ratio of surface area-to-volume is high. Nanoparticles are utilised to produce materials with different behaviour because of interfacial reactions. The current review compiles and discusses the available literature on the positive impact of nanoparticles on the mechanical behaviour of resin-based dental composites, in addition to the recent advancements. The effects of different types of fillers in nano-ranged size have been studied. It also presents appropriate filler quantity for the enhancement of some mechanical properties. Finally, the important role of energy dissipation mechanisms in dental composites has been emphasised.

Keywords: Dental composites; nanoparticles; nanocomposites; mechanical properties; energy dissipation mechanisms.

ABBREVIATIONS

1. INTRODUCTION

Composite restorative materials were evolved to overcome the shortages of silicate cement and methyl methacrylate-based resins [1]. Since the first application of dental composites in the early 1960s, numerous advances have been made [2]. Resin-based composites (RBCs) are extensively utilised in dentistry due to their satisfactory mechanical and biocompatibility properties, aesthetics and bonding ability [3]. The increased utilisation and popularity of these materials is attributed to the continuous improvements in strength, abrasion resistance, translucency, ease of manipulation and polishability [4,5]. RBCs are used as restorations, cavity liners, sealants, core buildups, inlays, onlays, crowns, cements, orthodontic devices, root canal sealers and posts, fixed and removable dentures, and implants [6,7]. Although the advancements of RBCs, there are multiple drawbacks continue to exist such as low mechanical properties. However, insufficient strength may lead to bulk fracture that is a common cause for short lifespan and failure of resin composite restorations [8-10].

In recent years, nanotechnology has been applied for the manufacturing of restorative materials (dental nanocomposites) [11], since the incorporation of nanoparticles (NPs) into polymers will produce new materials with advanced properties, such as hardness, diametral tensile strength, resistance to wear, fracture toughness, modulus of elasticity and flexural strength [12,13]. These improvements are owing to the large specific surface area and rich surface functional groups of NPs [14]. In addition, due to their high free surface energy, bonds between nanoparticles themselves and other particles are very strong. Agglomeration could be eliminated by functionalising NPs which produce same surface charge and hence they repel each other [15]. The main purpose of the present review is to highlight the research improvements made in determining the effects of the NPs on the mechanical properties of RBCs. Surface hardness, flexural strength and fracture toughness of dental nanocomposites are discussed. This review also addresses related factors (i.e., energy dissipation mechanisms) that affect the effectiveness of NPs to obtain the desired mechanical properties of the targeted composite materials.

2. COMPOSITION OF DENTAL RESIN COMPOSITES

Dental resin composites (DRCs) are typically consists of the following major ingredients: organic resin matrix (polymer), inorganic filler particles (such as glass, fused quartz, and silica), coupling agents, and the initiator–accelerator system. The components of the matrix include monomers, an initiator–activator system, stabilisers, and pigments. The fillers are interconnected with the matrix by a coupling agent. These basic components play a vital role in determining the performance of DRCs [16-18].

2.1 Resin Matrix

The resin is a monomer but is activated chemically and transformed to a hard polymer through a radical addition reaction [19]. The matrix in most DRCs contains a blend of dimethacrylate monomers such as bisphenol A glycidyl dimethacrylate (Bis-GMA) and urethane dimethacrylate (UDMA) [20]. Due to the high viscosity of Bis-GMA and to facilitate handling during manipulation, it is mixed with diluents such as triethylene glycol dimethacrylate (TEGDMA), UDMA and bisphenol A polyethylene glycol diether dimethacrylate (BisEMA) [21]. The addition of the diluents improves the degree of conversion, facilitates filler addition and mixing, and thus contributing to improvement of the mechanical properties [22].

2.2 Filler Systems

Different types of fillers have been used such as silica $(SiO₂)$, glass, carbon nanotubes $(CNTs)$, alumina (Al₂O₃), zirconia (ZrO₂) and titania (TiO₂) [23]. The inorganic phase in current dental composites composed of silica, quartz, borosilicate, and aluminum silicates. The filler loading usually varies between 35–70% volume percent (vol.%) or 50–85% weight percent (wt.%) [24]. The presence of filler has a positive impact on various properties, such as hardness, wear resistance, translucency, strength, coefficient of thermal expansion, overall curing shrinkage, fluids absorption, handling properties and aesthetic [25-27].

In order to improve the mechanical properties of restorative composites, changes on filler particles size and size distribution have been made [28]. Over the years, the particle size of fillers used in dental restorations has decreased from the conventional composites to the nanocomposite materials [29]. Based on the filler particle size, DRCs are classified as macrofilled (10–50 µm), microfilled (40–50 nm) and hybrid (10–50 µm and 10–50 nm) [30]. Hybrid composites are classified as microhybrids and nanohybrids. Microhybrids are then divided into two types, midfill (1–10 μ m to 40 nm) and minifill (0.6–1 μ m to 40 nm) [31,32]. Nanohybrids contain NPs and microparticles (0.1 to 2 μ m and \leq 100 nm). Finally, nanofilled composites were introduced. These composites contain NPs with more even distribution (<100 nm) [33,34]. Fig. 1 shows a flowchart of the classification of DRCs according to filler particle size.

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2.3 Coupling Agents

Due to the high surface energy of NPs, they exhibit higher tendency to agglomeration and inhomogeneous distribution [15,35]. However, the filler dispersion and adhesion at interface have a strong impact on the mechanical properties [36]. Therefore, the treatment of filler surface with coupling agents is essential to improve the adhesion between filler and matrix [37], giving better stress distribution from matrix to fillers [38], thus improving the mechanical performance [39]. 3methacryloxypropyltrimethoxysilane (γ-MPS) is the commonly applied silane coupling agent (Fig. 2) in the DRCs [40].

During silanation, the γ-MPS molecules bond chemically with the surface of filler particles via direct condensation between –OCH3 groups and the surface hydroxyl groups on the particles surface to form covalent bonds [41]. Another condensation occurs between the $-OCH₃$ groups on neighbouring molecule to give a siloxanecoated particles [42]. Silanation must involve the whole surface of inorganic particles to promote improved mechanical properties. However, reduced mechanical performance has been reported due to incomplete coating of silanated fillers, which leads to agglomeration. In clinical practice, the interfacial bonding among the composite's phases is susceptible to degradation in the humid mouth environment [43].

Fig. 1. Classification of DRCs according to filler particle size

Fig. 2. Silane coupling agent

3. EFFECTS OF NPS ON MECHANICAL PROPERTIES OF DENTAL COMPOSITES

3.1 Surface Hardness

Hardness is the surface resistance to deformation caused by external load [44]. It is indication of the material's ability to resist scratching, and its ease of finishing [45]. The achievement of esthetic is based on the proper finishing and polishing. However, the presence of scratches acts to decrease the fatigue life and cause failure of restoration [46]. The hardness of resin composites is determined by their ability to resist abrasion and stay stable in the mouth fluids [47].

In general, the addition of filler particles enhances the mechanical properties of resin composites [48] due to their high rigidity and stiffness as compared to polymer matrices [27, 49]. However, the integration of a high filler content leads to particles aggregation that reduces the crosslinks between the polymer chains, and causes plastic flow in the polymer matrix, which decreases the mechanical properties [50]. Several factors could influence the hardness such as size of filler particles, weight fraction of fillers [51], type and volume of silane, method of surface treatment, degree of dispersion, and hardness of fillers, because the addition of harder particles can significantly enhance the hardness of composites [35].

A previous study evaluated the hardness of micro and nanodental composites. The results displayed that the hardness of nanocomposites filled with 40 and 50 wt% nanoparticles was higher than the microcomposites, and the highest value was recorded for the group containing 50 wt% nanoparticles [52]. Others compared the hardness of three dental composites, i.e., Filtek Z350 XT (Nanofilled, aggregated ZrO_2/SiO_2 cluster and $SiO₂$ with particle size 5–20 and 20 nm, respectively, at filler loading of 78.5 wt%), Grandio (Nanohybrid, ceramic glass and spherical $SiO₂$ with particle size 1 km and 20–60 nm, respectively, at filler loading of 87.0 wt%), and Esthet X (Nanohybrid, barium boron fluoralumino silicate glass and $SiO₂$ with particle size 0.6–0.8 μm and 40 nm, respectively, at filler loading of 77.0 wt%). The findings showed that Grandio exhibited the highest value of hardness due to the filler particle size and filler loading, in addition to the high contact surface that observed

between nanofillers and organic phase [53]. Another comparison has been performed to estimate the hardness of four dental composites, i.e., Feeling Lux (Microhybrid), Amelogen Plus (Microhybrid), Filtek Z350 (Nanohybrid) and Te-Econom Plus (Hybrid). Filtek Z350 composites presented the highest mean values of hardness due to their smallest filler particles size (5–20 nm) [54]. In another work, the hardness of nanohybrid and microhybrid composites, light cured nanoionomer, and conventional glass ionomer materials has been examined. The hardness value of nanocomposite was higher than the others owing to NPs and increased filler content [55]. These outcomes are in agreement with others who found that nanocomposites exhibited higher hardness value compared to microcomposites due to the addition of high surface area particles at high filler content [56-58].

Furthermore, the effects of surface modification of nanofillers on hardness of acrylic dental resin have been investigated. Modified and unmodified nano-ZrO₂ (100 nm) at different percentages (1, 3 and 5 wt%) were added to poly(methyl methacrylate) (PMMA). Surface functionalisation has been done by two types of silanes, i. e., Vinyltris(2-methoxyethoxy)silane and 3- Methacryloxypropyltrimethoxysilane. The modified particles were efficiently dispersed with an increment in cross-linking, and thus hardness increased by 46.3% and 50.7%, respectively, while the addition of unmodified particles improved hardness by 24% [59].

In a recent study, treated Al_2O_3 (20 nm) and marble dust (70 nm) have been added to dental composites at concentrations of 5, 10, 15, and 20 wt%. The hardness elevated by 86% and 61% upon the inclusion of 20 wt% treated Al_2O_3 and marble dust, respectively. This enhancement is due to the high percentage of hard filler, which stiffened the matrix elasticity and improved the indentation resistance. More hardness of the nano- Al_2O_3 composite was attributed to the stronger bonding between nano- Al_2O_3 and silane as compared to the bonding between marble and silane. Moreover, the strong filler-matrix interfacial adhesion is attributed to the improved dispersion, which resulted from the reduced particle size [60]. Zidan et al. [61] attained similar results when they added $ZrO₂$ nanoparticles (30 and 60 nm) at different percentages (1.5, 3, 5, 7 and 10 wt%) to PMMA denture base. The highest value was obtained at 10 wt%. Table 1 displays the effect of nanoparticles on hardness of DRCs.

Material	Filler	Particle size (nm)	Filler loadings (wt%)	Optimum loading	Reference
Dental composite	SiO ₂	$20 - 50$	20, 30, 40 & 50	40 & 50	(Hosseinalipour et al. [52])
	$\mathsf{Al}_2\mathsf{O}_3$	20	5, 10, 15 & 20	20	(Meena et al. [60])
PMMA	ZrO ₂	20	0.5, 1, 3 & 8 & 5	5	(Algahtani [57])
		100	1.3 & 8.5	5	(Elmadani et al. [59]
		30 & 60	1.5, 3, 5, 7 & 10	10	(Zidan et al. [61])

Table 1. The improvements in hardness of nanoparticles filled dental composites

3.2 Flexural Strength

The flexural strength can be enhanced by incorporating ceramic fillers into the polymer matrix [62]. Increasing the inclusion content resulted in improving flexural strength [63]. In general, the mechanical characteristics of polymer composites could be affected by the filler particle size [64]. Other factors may play a role in promoting the flexural strength such as filler/matrix bonding, filler ratio [65] and conversion degree of the polymer matrix [66,67]. Although the stiffness and strength are increased in traditional dental composites, the dimensional changes are decreased [68]. Salination can also promote filler/polymer bonding and deviate the crack propagation which leads to better flexural strength [65].

Moreover, the inclusion of nanoparticles in dental composite materials exhibited a remarkable improvement in flexural strength [69]. Previous work was performed to study the flexural strength of three dental composite materials (Filtek Z350 XT, Grandio, and Esthet X) [53]. The highest value was achieved by Filtek Z350 XT. The results suggest that the flexural strength is

positively affected by the filler particle size and concentration. Another study was conducted to measure the flexural strength of UDMA dental composite materials mixed with glass (140 nm) at loadings of 5, 7.5 and 10 wt%. The flexural strength increased by 17.2% at filler loading of 10 wt% [70]. Nano-ZrO₂ particles were also added to the dental composite at different percentages (1, 2 and 3 wt%). The maximum filler level (3 wt%) displayed higher flexural strength [71]. Alqahtani [57] investigated the effect of adding various ratios (0.5, 1, 3 and 5 wt%) of hexagonal boron nitride particles (70 and 800 nm) on flexural strength of self-cured PMMA. An amount of 550% increment was recorded for PMMA composite filled with 800 nm hexagonal boron nitride at 5 wt%. Table 2 shows the improvements in flexural strength of dental polymer composites filled with various ratios of nanoparticles. Overall, low NPs loadings could enhance the flexural strength of dental polymer composite. Selection of filler particle size should be done according to the characteristics influenced by the filler content e.g., curing contraction and viscosity. Polymer structure turns out to be another factor that has a marked impact on flexural strength.

Material	Filler	Particle size (nm)	Filler loadings (wt%)	Optimum loading	Reference
Dental composite	Al_2O_3	$25 - 40$	10, 20 & 30	30	(Foroutan et al. [69])
	Glass	140	5, 7.5 & 10	10	(Monfared & Bahrololoom [70])
	ZrO ₂	\overline{a}	1, 283	3	(Kumar et al. [71])
PMMA	Hexagonal boron nitride	800	0.5, 1, 3 & 5	5	(Alqahtani [57])

Table 2. Improvements in flexural strength with increasing nanoparticles loading

However, reduced flexural strength with the further rise in filler ratio has been reported. Hosseinalipour et al. [52] compared the flexural strength of dental composites mixed with nanosized $SiO₂$ (20–50 nm) at different concentrations (20, 30, 40 and 50 wt%) and dental composite mixed with micro-sized $SiO₂$ (10–40 µm) at a concentration of 60 wt%. They inferred that the nanocomposites showed greater flexural strength than the microcomposite. However, the highest value was obtained at 40 wt%, then decreased at 50 wt%. In another study, Liu et al. [72] added nano-SiO $_2$ to dental composite at concentrations of 0.5, 1, 1.5 and 2 wt%. The highest flexural strength was obtained via 1 wt% nanofiller. When a higher amount of nanoparticles were impregnated, the flexural strength was reduced. This behaviour was also observed by Kundie et al. [73], who added Al_2O_3 nanoparticles (7 nm) at different loadings (0.13, 0.25 and 0.5 wt%) to the PMMA denture base. The maximum value was reached at 0.13 wt% and then dropped. Another study also stated that the incorporation of 1 wt% TiO² nanoparticles into PMMA improved the flexural strength, and then decreased at a filler ratio of 2 wt% [74]. The decreases in flexural strength of dental composites beyond optimum nanoparticles loading are summarised in Table 3. Inferior flexural strength with high filler content can be ascribed to aggregation of NPs which leads to stress concentration and alterations in the elastic modulus of the matrix and accelerating the crack pathway due to partial embedding of the fillers in the polymer [74,75].

3.3 Fracture Toughness

Fracture toughness (K_{IC}) is an essential characteristic of a material to anticipate the strength in the existence of a crack [76]. It is assigned to be one of the most crucial mechanical attributes of any dental material [77]. It can be defined as the energy absorbed by the material during deformation before fracture due to crack propagation [78]. In another word, it is a

measure of the energy required to propagate a crack from an existing defect [79]. However, it differs from strength, in that it is an inherent property of a material, and therefore its value should be independent of testing modality or specimen geometry [80].

The inclusion of fillers in the polymer matrix improves tensile strength, modulus of elasticity and fracture toughness [81]. The influence of filler on the K_{IC} probably happens due to the cracks propagation around filler particles through the polymer matrix. A higher filler ratio encourages larger compression of the polymer, leading to a decrease in the force applying on filler particles thereby inhibiting the crack propagation [63]. Accordingly, greater K_{IC} could be attained by increasing the filler content [79].

In addition, the size of the filler particles has a distinctive influence on the mechanical behaviour of polymer composite materials [64]. Fu et al. [82] stated that particle size considerably influenced the K_{IC} of polymer composite materials. The incorporation of nanofillers into RDCs leads to substantial changes in mechanical properties [83]. Nanofillers can be uniformly distributed in the polymer matrix, which enhances K_{IC} in comparison with microcomposites [84]. As stated by Yeli et al. [85], dental nanocomposites displayed improved K_{IC} . This is in agreement with Zhou et al. [32], who revealed that nanocomposites exhibit remarkably greater fracture toughness than microcomposites. Another work was carried out to compare the K_{IC} of nanohybrid and microhybrid composites, light-cured nanoionomer, and conventional glass ionomer materials. The result was 1.75, 1.52, 1.27 and 0.83 MPa.m^{1/2}, respectively. The K_{IC} of the nanocomposites was greater than other dental composites. The fracture toughness of nanohybrid is improved because of the higher filler level and nanoparticle constituents [55].

Material	Filler	Particle size (nm)	Filler loadings (wt%)	Optimum loading	Reference
Dental composite	SiO ₂	$20 - 50$	20, 30, 40 & 50	40	(Hosseinalipour et al. [52])
		12	70	70	(Atai et al. [88])
		12 & 40	$(15, 20, 25 \& 30)$ and (25, 35, 45, 50, 8, 53)	25 & 50	(Barghamadi et al. [89])
PMMA	SiO ₂	12	0.25, 0.50, 0.75, 8, 1	0.25	(Topouzi et al. [12]
	Al_2O_3		0.13, 0.25, 0.5, 1, 2 & 5	0.5	(Kundie et al. [73])

Table 4. The improvements in fracture toughness of nanoparticles filled dental composites

Salination and NPs increase the K_{IC} of dental composite materials by enhancing the bonding between the NPs and organic matrix through a greater surface area and superior particle strength [86]. In addition to the crack extension that occurs via several possible toughening mechanisms, like, crack deflection and crack bridging [84,86,87]. An earlier study by Hosseinalipour et al. [52] evaluated the fracture toughness of dental polymer composites containing silanated $SiO₂$ particles (20–50 nm) at different loadings (20, 30, 40 and 50 wt%). The control group was containing 60 wt% $SiO₂$ particles with sizes ranging from 10–40 μm. The fracture toughness values increased gradually as filler loading increased, then decreased when reached 50 wt%. The maximum fracture toughness value was 1.43 MPa.m^{1/2} that obtained at 40 wt% filler content, whereas the control group achieved 1.07 MPa.m $1/2$. Another study indicated that the impregnation of 70 wt% of silanated porous $SiO₂$ (12 nm) to dental polymer composite, substantially enhanced the fracture toughness [88]. One study found that adding silanated $SiO₂$ particles with the average size of 12 nm, at different percentages (0.25, 0.50, 0.75 and 1 wt%) considerably improved the K_{IC} of PMMA for fixed interim restorations. The fracture toughness was reduced steadily by raising the nano-SiO₂ ratio. The highest value was recorded at a filler content of 0.25 wt% [12]. Similarly, other researchers found that adding silanated nano-SiO₂ particles (12 and 40 nm) with loadings of 15, 20, 25, 30 wt% and 25, 35, 45, 50, 53 wt%, respectively, improved the K_{IC} of dental polymer composites. The findings revealed the K_{IC} is improved due to a higher filler level. The optimal increase is reached by both groups, i.e., 12 and 40 nm (25 and 50 wt%, respectively), and then decreased with an increase in nanofiller content up to 30 and 53 wt%, respectively. In general, the fracture toughness of the composite group filled with 12 nm $SiO₂$ was higher than the composite group

filled with 40 nm $SiO₂$. They concluded that the K_{IC} of the dental composite could be affected by the filler size. Rougher materials can be attained by using a smaller filler size [89]. A similar occurrence was observed when nano- Al_2O_3 particles (7 nm) mixed at different loadings (0.13, 0.25, 0.5, 1, 2 and 5 wt%) with a PMMA denture base. The fracture toughness improved with a higher filler level to a maximum and then dropped. The composite loaded with 0.5 wt% filler exhibited the highest fracture toughness [73]. The improvements in K_{IC} of dental nanocomposite are shown in Table 4. The K_{IC} of nanocomposites depends on the filler levels and the particle size. Composites with lower aspect ratio NPs have higher fracture toughness than that of higher aspect ratio NPs.

4. ENERGY DISSIPATION MECHANISMS

Some energy dissipation mechanisms have a remarkable effect on crack suppression [90]. The possibility of a particle being hit by a crack in a favourable direction for energy dissipation rises with a greater level of the filler [91]. The fracture toughness increases due to the good filler/matrix interfacial strength and high mechanisms of energy dissipation like the deflection of the crack and filler/matrix detachment [92]. The composite material becomes tougher as the crack grows, and resists the catastrophic failure [80]. The inclusion of micro- and nanofillers enhanced fracture toughness. The particles of the filler offer toughening via improved energy dissipation through crack propagation like, crack bridging, pinning and deflection [93].

4.1 Crack Bridging

The fracture toughness of polymer nanocomposites can be enhanced by one of the principal mechanisms such as crack bridging [94]. When nanofiller link both sides of the fracture, it might either fracture or pull out from one of the surfaces. Such criteria depend on the interfacial strength, embedded length, flexibility of nanofiller and angle to fracture surface. Therefore, the bridging mechanism is very crucial in promoting the K_{IC} of polymer composite materials. Lohbauer et al. [91] stated that crack bridging is one of the toughening methods that active in dental composite materials.

4.2 Crack Pinning

The stress at the crack tip can be less localised due to the existence of adjoining particles in polymer composites. Energy dissipation can happen by a crack pinning leading to a greater fracture resistance [95]. Alternatively, a propagating crack can be pinned by the particles which results in the crack front to "bow" among the filler particles, thus enhancing the K_{IC} [96]. Yadav and Gangwar [97] reported that the increase in the K_{IC} is related to the crack pinning and crack deflection effect of NPs. The energy dissipation methods advanced through crack propagation provides developed fracture toughness.

4.3 Crack Deflection and Branching

Deflection of crack at particle was recommended to have a prominent effect in the toughening. According to the theory proposed by Faber and Evans [98], when a crack approaches an obstacle, it is tilted and twisted out of the initial plane [99]. This increases the roughness of the fracture surface due to non-planar cracks, thus resulting in increased fracture toughness [100]. For fracture toughness and initial fracture strength, dental composite materials reinforced with greater particle size proved superior products because of the deflection of the crack [101]. According to Lohbauer et al. [91], the underlying energy-dissipating mechanisms in dental resin composites are crack branching and crack deflection.

4.4 Particle–Matrix Interface

The interface has a critical effect on deformation behaviour so that under the strong bonding conditions high amounts of energy will be consumed at the interface of the particle and matrix [70]. The strong interfacial allows load transfer leading to the improved mechanical performance of the dental polymer composite.

The impact property and crack propagation resistance are affected by the interface adhesion strength. These failure mechanisms led to high energy absorption as a result of dissipation during the propagation of the crack [102].

5. CONCLUSIONS

The effects of the size of filler particles on the mechanical behaviour of dental composite materials are quite challenging because most studies were performed to determine the influence of varying one parameter. Though from the cited literature, it could be concluded that the addition of nanofillers produces improved mechanical characteristics for DRCs. Enhancing the bond strength between filler particles and the matrix is the main point for promoting mechanical performance, which could be attained with the functionalisation of the filler surface by a coupling agent. Energy dissipation mechanisms can take place in DRCs. Filler particles offer toughening via improved energy dissipation mechanisms through the propagation of the crack. These toughening mechanisms ensure better mechanical properties of dental composites.

DISCLAIMER

The products used for this research are commonly and predominantly use products in our area of research and country. There is absolutely no conflict of interest between the authors and producers of the products because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge. Also, the research was not funded by the producing company rather it was funded by personal efforts of the authors.

CONSENT AND ETHICAL APPROVAL

It is not applicable.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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