

Toughening Mechanisms in Particulate Dental Resin-Based Composites: A Short Review

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ABSTRACT

Resin-based composites represent a unique class of restorative dental materials; however, these composites have severe shortcomings such as low wear resistance which is mainly responsible for the short life-span of the materials. Composites characteristics such as strength, stiffness, resistance to abrasion, polymerisation shrinkage, thermal expansion coefficients and moisture absorption depend on the filler particles and coupling agents used. Fortunately, these composites have been the focus of attention for numerous researches in recent years which aim to improve the performance of the restorations in several ways. Using several types of coupling agents, and different particle sizes and types have gained the great interest of researchers. The latter plays a critical role in the toughening mechanisms in resin-based composites. Therefore, the purpose of this review is to discuss the literature regarding the toughening mechanisms in particulate dental resin composites since these mechanisms are also crucial factors for the improvement of mechanical properties. The four main types of toughening mechanisms discussed are: crack deflection, pinning, bridging and particle-matrix interface. The current review indicates an improvement in mechanical properties of particle-filled dental composites due to the presence of various toughening mechanisms. The dental resin composites' fracture toughness is mainly contributed by crack deflection, pinning and bridging that take place in micro- and nanocomposites, in addition to the hybrid composites. Filler-matrix interphase plays an important role in improving the mechanical properties, in addition to its positive effect on crack deflection and bridging. In reality, all these mechanisms could occur simultaneously at different intensities, respectively.

Keywords: Particulate dental composites; crack deflection; crack pinning; particle/matrix interactions; crack bridging; toughening mechanisms

INTRODUCTION

Resin-based composites are extensively utilised in the dental sector (Liu et al. 2014), for example, as cavity liners, inlays, onlays, core build-ups, crowns, provisional restorations, denture teeth, cementation of crowns, bridges and orthodontic brackets, structured scaffolds, root canal sealers and posts, plates and screws (Ferracane 2011; Muddugangadhar et al. 2011; Sakaguchi & Powers 2012; Khan et al. 2017). Resin composites are the most desired restorative materials because of their superior aesthetic, moderate cost, mechanical and biocompatibility properties, simplified clinical procedures, bonding ability, developed formulations, and decreased amalgam use due to mercury hazard and toxicity (Mota & Subramani 2018; Razali et al. 2018).

Basically, dental resin composites (DRCs) are composed of three main components: an organic polymer matrix (continuous phase), inorganic fillers (disperse phase) and coupling agent (coupling phase) (Rodriguez-Quiros &

Casanova-Yepes 2015). For the matrix, polymers that are often used include methacrylates, epoxy, and polyethylene (Ramakrishna et al. 2001; Lamichhane et al. 2014; Munhoz et al. 2017).

Several types of fillers such as glass or ceramic particles, fibres, nanoclusters, and nanotubes, which all have different compositions, sizes, and size distributions, are used to improve the matrix properties (Graciano et al. 2012; Habib et al. 2015). Coupling agents such as silane are used to provide strong bonding between the resin matrix and fillers, allowing the composite performances to improve. 3-methacryloxypropyltrimethoxysilane (γ -MPS) is the most common silane used in restorative dental composites. Filler amount, size, type, and shape are important factors that enhance the mechanical properties of DRCs (Mohseni et al. 2016). Although various types of fillers have been used to increase toughness, strength, and durability, several shortcomings still exist, such as poor mechanical properties, polymerisation shrinkage, water absorption, and low wear resistance of high occlusal loadings (Kim et al.

2006; Leprince et al. 2010; Liu et al. 2014). DRCs fail due to surface and/or bulk cracks, matrix and filler degradation, water absorption, and inadequate mechanical properties (Mohseni et al. 2016). The main cause of DRCs failure is degradation of the filler-resin bond after long time water absorption (Wang et al. 2014).

Nano-composites were recently introduced as polymer dental composites (Kumari et al. 2016). The addition of nanoparticle fillers enables the DRCs to have better optical, aesthetic and mechanical properties, such as resistance to fracture and tensile strength, as well as decreased polymerisation shrinkage (Sachdeva et al.). Moreover, nano-sized particles improve gloss retention, wear resistance and the fatigue properties of DRCs (Hosseinalipour et al. 2010). Reduced interparticle distance may lead to increased barriers to dislocation movements and decreased strain localisation (Kim et al. 2007). However, several factors influence the properties of nanocomposites; for example, a conversion degree that requires a higher degree of functionalisation due to the high surface area of nano-sized particles (Kim et al. 2007; Sideridou & Karabela 2009).

In addition to the aforementioned factors, toughening mechanisms play a significant role in improving the mechanical properties of DRCs (Calabrese et al. 2016). Fracture toughness highly relies on toughening mechanisms which increase crack propagation resistance. Thus, this short review focuses on the common toughening mechanisms observed in particulate dental resin composites. It elaborates the influences of toughening mechanisms on the improvements of mechanical properties of the particulate DRCs. In addition, it aims to summarise the attempts made so far to enhance the mechanical properties of particulate DRCs, discuss the reinforcing effects of these mechanisms, and improve the understanding of toughening mechanisms in these composites. However, no literature review paper on toughening mechanisms in particulate DRCs has been previously published.

TOUGHENING MECHANISMS

The strengthening and reinforcement of DRCs are achieved by the addition of particulate fillers. Several types, sizes, and volume fractions of these fillers are utilised in addition to filler surface treatment. The combination of these factors leads to the presence of toughening mechanisms and yields better mechanical properties. According to Ferracane et al. (2014), dental composites containing particulate fillers showed enhanced fracture toughness through toughening mechanisms. Crack bridging, crack pinning between fillers, and crack deflection or branching are the most active in these

composites (Lohbauer et al. 2013). According to Monfared and Bahrololoom (2016), as filler content increased, the fracture surface became rougher, indicating more ductile fracture. However, this takes place at optimum filler concentration and could be attributed to crack deflection and crack pinning. On the contrary, improvements in mechanical properties were obtained by a small addition of filler content; this is due to the effective energy dissipation mechanisms such as crack pinning and deflection (Nanda et al. 2019).

CRACK DEFLECTION

Crack deflection takes place due to the dominant interparticle/intercluster crack growth within the polymer matrix (Kumar et al. 2012) when the crack path is forced to move out from the initial direction to continue its propagation (Xavier et al. 2015). This results in an increment in fracture toughness due to the increased fracture surface roughness resulting from non-planar cracks (Dittanet 2011). It is a shielding mechanism where the fracture resistance increases by decreasing the stress intensity around the crack front (Kumar et al. 2012).

It has been reported that the fracture toughness of dental nanocomposites was enhanced by nanoparticles and surface treatment, which enhanced the interface toughness, crack bridging and deflection (Chen 2010). On the contrary, Ornaghi et al. (2014) reported that dental composites filled with 78 wt% glass microparticles (1.9 μm) had higher fracture toughness than composites filled with smaller particles. They attributed this increment to the mechanism of crack deflection. However, Monfared et al. (2014) studied the effects of different loadings (5, 7.5, and 10 wt%) of glass nanoparticles (140 nm) on urethane dimethacrylate composites. They showed that with increase in particle content up to 10 wt%, flexural strength and flexural modulus increased by 17.2% and 16.7%, respectively. They concluded that mechanical properties depend strongly on nanoparticles loading due to the crack propagation and crack deflection mechanisms, as shown in Figure 1, where the crack propagated and, at the same time, was deflected by filler grains (Shah et al. 2009). Hence, through this crack propagation and deflection, the toughness of the dental composite is increased due to a longer crack propagation path. Therefore, a suitable selection of filler type, amount and size could enhance the crack deflection mechanism in both dental micro- and nanocomposites. Furthermore, researchers should ensure the formation of optimum continued crack propagation and crack deflection to increase DRCs fracture toughness.

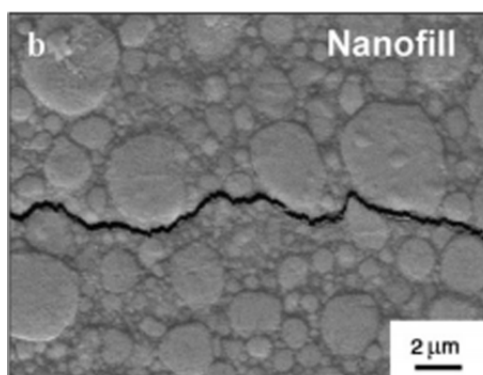


FIGURE 1. Crack propagation together with crack deflection observed in nanofill dental composite (Shah et al. 2009)

CRACK PINNING/BOWING

Another toughening mechanism occurs when a crack-tip is being forced to bow out around the particles and remains pinned at the particles (Albdiry et al. 2013). It is a considerable mechanism, especially in rigid particle-filled brittle polymers (Zhang et al. 2006). The occurrence of crack pinning in micro- and nanocomposites has been reported in the literature (Sun et al. 2009; Albdiry et al. 2013). It easily happens at a nanoscale level due to the reduced inter-particle distance for nanofillers even at higher loadings (Zhang et al. 2006). According to the fractography results obtained by Medina et al. (2008), the incorporation of nanoparticles induced crack pinning in the experimental composite. The

presence of river-like lines in nanocomposites could be a result of the crack pinning mechanism and blocking effects of nanoparticles (Zhang et al. 2006). Besides this, hybrid dental composites showed increased fracture toughness due to crack pinning/bowing and crack deflection mechanisms (Moezizadeh & Mokhtari 2011). The crack initiation sites and arrest lines on the fracture surface indicating the occurrence of crack pinning are shown in Figure 2 (Karci et al. 2019). Therefore, this crack pinning mechanism for DRCs can be optimised through the correct selection of very hard, rigid and inert filler particles such as ceramic materials i.e. viz. alumina and zirconia. At the same time, the number and distribution of the pinning effect can be further increased when the filler particles are in nanosize.

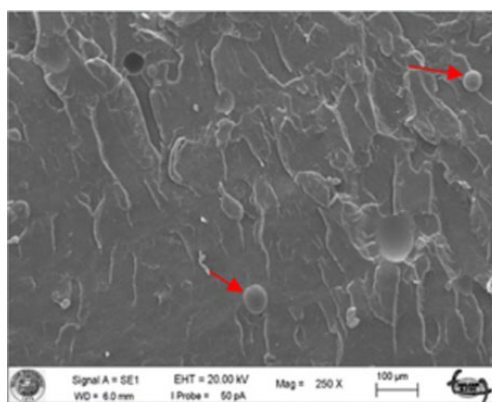


FIGURE 2. Fracture surface of PMMA/TiO₂ nanocomposites with crack initiation sites and arrest lines, which are evidences of crack pinning (Karci et al. 2019)

Crack bridging is identified as the main mechanism for improving crack resistance (Lohbauer et al. 2013). It occurs due to the interparticle/intercluster crack growth in the matrix (Kumar et al. 2012) when a particle connecting the two sides of the crack at the crack tip wake (De Souza et al. 2011; Xavier et al. 2015). Such bridges particles sustain part of the applied load, which would be experienced at the crack front, effectively increasing fracture resistance by minimising the stress intensity at the crack front, thus working as an extrinsic toughening mechanism (Shah et al. 2009).

As the load is further raised, a microcrack is initiated very close to the tip of the main crack, and a bridge connects the two cracks. The initiated microcrack propagates toward both directions while the main crack propagation is stopped. Finally, due to the microcrack extension, both cracks will link with each other. Toughening ceases at this point.

Dental composites filled with salinated nanoparticles have higher fracture toughness through a combination of silanisation, presence of a strong interface, crack deflection

and bridging (Chen 2010). In case of reinforcing filler, the bridge formation depends to a large extent upon the strength of particle–matrix interface. As the crack propagates, the presence of weak interface allows particles to debond, which results in a friction between the two phases, before the particles are being pulled out. In case of a strong interface, the propagating crack can be arrested at a particle, which leads to a stress concentration in front of the main crack, resulting in a secondary crack nucleation (Khvostenko 2014). Figure 4 shows an example of crack bridging in bioactive glass dental composite (Khvostenko et al. 2013). The way the crack bridging occurred can be clearly seen: the crack propagation was arrested and deflected by the filler particles. As a result, crack bridges created in the crack wake due to the particles connected the crack front, or new microcracks forming ahead of the crack tip. In summary, crack bridging plays a crucial role on the toughness of particle-filled dental composites. It depends primarily on the existing of a strong particle–matrix interface which allows an effective crack arrest process.

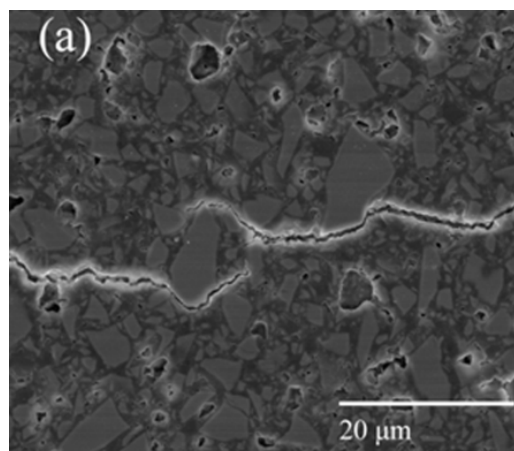


FIGURE 3. The particles in bioactive glass dental composite connected the crack faces at a wake near the crack front and resulted in crack bridging (Khvostenko et al. 2013)

PARTICLE–MATRIX INTERACTIONS

The particle–matrix interface is a region in the vicinity of the particle surface, where polymer properties are changed as compared to the bulk (Gu et al. 2012). The presence of thick and weak interface layer results in crack propagation and crack blunting within the interphase. However, a thin and strong interface layer facilitates efficient stress transfer and gives rise to crack deflection and propagation within the matrix (Wang et al. 2008). The interfacial interactions between the polymer matrix and filler give a positive effect on stress transfer between the two constituents, thus becoming the key driving force for the toughening and strengthening effects (Calabrese et al. 2016). The greater interface between the filler and the matrix and the stronger interfacial interaction results in greater reinforcement effect (Joseph et al. 2019). In addition, high amounts of energy are consumed at the filler–matrix interphase due to the perfect

bonding conditions (Du & Zheng 2007; Sadeghipour et al. 2013). The mechanical properties of composites are strongly affected by the interface quality. According to the literature, bond integrity at the particle–matrix interface plays a crucial role on the strength and toughness of nano- and micro-particle filled composites through an increased stress transfer from matrix to filler (Fu et al. 2008; Nassar & Nassar 2013; Sadeghipour et al. 2013). Hua et al. (2015) reported that the interphase modulus and quality of interfacial bonding have a significant effect in increasing the stiffness of nanocomposites. A study by Nayyer et al. (2018) on two types of dental composites reported that the hardness of nanocomposites was higher than that of microhybrid composites. They attributed this increase to the higher nanoparticles content and higher surface area, thereby allowing stronger interfacial adhesion between nanofillers and matrix. This is in accordance with Aminoroaya et al. (2021) who reported that nanoclusters display good filler-

matrix interface properties owing to their high surface area in comparison with micron-size filler particles. Meena et al. (2019) stated that the improvement of hardness was due to the uniform dispersion of smaller particles within the matrix which increased the interfacial bonding between the two phases.

According to Boussès et al. (2019), the interface was expected to improve due to the silanisation, and it was confirmed by SEM micrograph (Figure 3). The divided particles are seen as two halves on both sides of the fracture

surface, indicating that the crack propagation through the filler particle requires minimum energy compared to bypassing along particles interface with the matrix. The researchers concluded that energy is well transferred to the filler particles by the interface. To summarise, silanisation is the key factor in achieving a high quality interface. Thus, an efficient stress transfer between filler and matrix considerably improves the mechanical properties of micro- and nanocomposites.

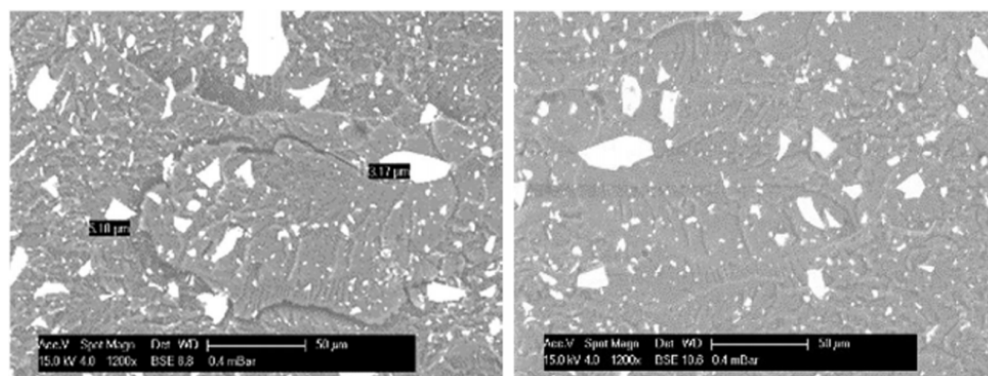


FIGURE 4. Particle–matrix interactions in dental composites are confirmed by the presence of two halves of the broken particles on each side of the fracture surface (Boussès et al. 2019)

MIXED TOUGHENING MECHANISMS

In reality, when the particulate DRCs are subjected to stress, it is not necessarily dependent only one single crack mechanism but is most likely contributed from mixed mechanisms which could occur simultaneously. Crack deflection and crack bridging often work in harmony, given that crack deflection usually leads to crack bridging (Shah et al. 2009; Kumar et al. 2012). A study by Pałka et al. (2020) observed the presence of crack deflection and crack bridging in experimental DRCs containing a mixture of ceramic fillers. However, the dominant fracture mechanisms in experimental dental nanocomposites are crack deflection and particle-matrix interface (Chan et al. 2007). In composites composed of 180/500 nm irregular SiO₂-BaO glass particles, active crack pinning/deflection were observed (Belli et al. 2014). To sum up, different toughening mechanisms could occur when load is applied to the DRCs. Usually, crack deflection is combined with crack bridging and takes place before. Additionally, both crack deflection and bridging are affected by the strength of particle–matrix interface.

CONCLUSIONS

The properties of particulate dental composites are dependent upon a multitude of factors, including the loading and quality of the reinforcing filler particles, the polymer matrix, and the coupling agent which facilitates stress transfer between these phases. Toughening mechanisms (i.e., crack deflection, crack pinning/bowing, particle–matrix interactions, and

crack bridging) play the key role in enhancing fracture toughness. Crack deflection leads to an increment in fracture surface roughness, thus to increased fracture toughness. Crack pinning/bowing occurs due to the reduced inter-particle distance at higher nanofillers concentrations. It effectively increased the fracture toughness of hybrid dental composites. The interface quality gives a positive effect on stress transfer between the two constituents. The increased stress transfer from matrix to filler increases the strength and toughness of dental composites. The good bonding results in higher energy consumption at the filler–matrix interphase. Thus, particle–matrix interactions play a very important role in improving the mechanical properties. Crack bridging increases fracture resistance by decreasing the stress intensity at the crack tip. It effectively increases fracture toughness through a combination of silanisation and presence of a strong bonding at the particle–matrix interface. Finally, further review is needed to determine: 1) the role of each mechanism, in-depth, and explore if other toughening mechanisms might exist; 2) toughening mechanisms in fibre-reinforced dental composites and hybrid dental composites; 3) whether the particle shape is more effective in toughening and why; 4) the existence of these mechanisms at very dense particle packing; and 5) the main mechanisms for micro- and nano-composites.

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DECLARATION OF COMPETING INTEREST

None

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