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Ceramic and glass-ceramic fillers in dental composites: A review

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Abstract

Dental composite fillings are most popular fillings wherein various fillers have been suitably dispersed in a polymerizable resin. Major challenges are to make choice of suitable filler and their distribution in the resin matrix in the development of the dental fillings. Apart from traditional fillers ceramic whiskers, glass fibres, branched fibres and nanoporous powders have also been investigated as fillers in dental composites. Recently, ceramic, and glass-ceramic fillers are attracting attention of the researchers due to their suitable combination of material properties. In the current paper, the prospect of ceramic and glass-ceramics as fillers in fabricating dental composites has been reviewed.

1. Introduction

Dental composites are consisted of a polymerizable resin base containing some amount of ceramic filler [1]. The filler materials are of inorganic composition and the matrix is of organic composition. Compared to dental amalgam, dental composites provide improved aesthetics and mechanical properties [2]. Thus, dental composites have been used as fillings for the last five decades. However, polymerization shrinkage and bacterial micro-leakage are the existing problems associated with the composite fillings causing de-mineralization of dentine and generation of disease [1-3].

Apart from traditional fillers ceramic and glass-ceramic fillers have also been studied as fillers in dental composites. The morphology and amount of the crystalline phases are major aspects that should be properly controlled in case of glass-ceramic filler reinforced dental composites. Further, the use of SiO2 based glass-ceramic particles may enhance their interaction with silane coupling agent in dental composites [3,4]. Glass-ceramics are proposed for bone repair and substitution on account of better mechanical properties as well as biocompatibility. They bond with the living bone through the formation of a hydroxyapatite layer [3]. 2% to 3% shrinkage is found for commonly used resin composite. Resin composite displays viscoelastic deformation during load application. Response of resin composite depends on matrix composition, amount of fillers and degree of conversion [5].

2. Resin based composites

The drawback of resin composite is polymerization shrinkage in spite of the aesthetic improvement relative to amalgam [3]. Three factors such as composition of resin composite, resin matrix conversion and volume fraction of filler contribute to the polymerisation shrinkage induced stress [6]. Ritzberger *et al.* [7] studied that leucite-type glass-ceramics demonstrate high translucency, desirable optical and mechanical properties for their suitable application as dental inlays, onlays and crowns. Thus, the lithium disilicate glass-ceramics are utilized as crowns and small dental bridges [7].

Recently, nanofilled or nano-hybrid resin based composite (RBC) are being used widely. The nanofilled RBC is consisted of nano-sized dispersed filler particles and nanoclusters [8]. Nanofilled RBC showed superior strength and high fracture resistance under critical situations. The interpenetrating phase composite (IPC) structure can retain enhanced mechanical properties as compared to other RBC's [9]. Nanofillers have greater tendency to multiple fractures than that of the traditional fillers. Recent technology has modified the filler size and chemical composition of filler for the formation of RBC. Silorane RBC shows the lowest polymerization shrinking stress [10]. Therefore, the silorane composites cause reduced stress generation by polymerization shrinkage and high reactivity. Therefore, these composites can be employed as an effective restorative material [11,12].

3. Ceramic/glass-ceramic fillers in dental composites

Mollazadeh *et al.* [13] investigated dental composites reinforced with fluoroapatite and mullite crystallite containing glass-ceramic fillers. The flexural strength measurement showed strong dependency on the filler composition. Kundie *et al.* [14] prepared dental composites by the addition of Al_2O_3 micro- and nano-particles to poly methyl methacrylate (PMMA). Al_2O_3 particles (2 and 0.5 wt. %) was surface treated using (3-methacryloxypropyl)trimethoxysilane (γ-MPS) added to methyl methacrylate (MMA) and mixed with PMMA

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powder. It was observed that the flexural modulus enhanced with the increase of filler content. On the contrary, the flexural strength decreased with increasing filler content [14]. Ekworapoj *et al.* [15] investigated mechanical strength of conventional dental composites. It was noticed that filler content was more effective in controlling diametral tensile strength (DTS) and Vickers hardness than monomer type and ratio. Moreover, the prepared dental composites indicated lower DTS and hardness values than those of the commercial composites [15].

Dental resin based composites mixed with hydroxyapatite (HA, $Ca_5(PO_4)_3OH$) micro-particles were observed to produce enhanced mechanical properties [16-19]. The dental RBCs reinforced with high-aspect-ratio HA filler showed improved mechanical properties than those of the spheroidal HA reinforced ones [20-23]. Novel urchin-like hydroxyapatite (UHA) prepared by microwave irradiation had been applied as a dental filler. The silanized UHA remarkably improved the mechanical properties than unfilled resin matrix. UHA with silica nanoparticles increased the mechanical properties significantly [24]. However, the mismatch of refractive index of HA with resin matrix could result in reduced monomer conversion and cure depth [25]. Despite this fact HA filler containing dental RBCs are promising bioactive restoration materials. Silicon dioxide is the most widely used filler. Apart from this filler many other ceramics have also been studied for use as fillers [16, 26-29]. It has been seen that zirconia filler and hybrid zirconia-silica fillers are significantly used in commercial dental composites due to high hardness of zirconia [30, 31].

Aljabo *et al.* [32] developed light-curable, high strength reactive filler reinforced dental composites. During immersion in simulated body fluid (SBF) the developed composite released calcium phosphate and chlorhexidine (CHX) and thereafter co-precipitation of hydroxyapatite/CHX occurred on the surface. Glass fillers containing CHX and different amount of reactive mono-and tricalcium phosphate (CaP) was mixed with urethane dimethacrylate based liquid. The tooth restoration failure can be avoided by the hydroxyapatite precipitation and surface antibacterial accumulation. Figure 1 shows the SEM images for 0% CaP, 10% CaP, 20% CaP and 40% CaP containing composites immersed for 1 week in SBF. Any CaP containing sample was covered with HA spheres when stored in SBF (Figure 1 (b) to 1 (d). From SEM it was clear that with increasing CaP content the average size of the HA spheres and the percentage of the surface covered by HA was increased. Figure 1e and f shows that the spongy spheres had pores with 100 nm diameter and 10 nm thick pore walls. Figure 2 shows the XRD patterns for the 10, 20 and 40% CaP containing composite surfaces after immersion for one month in SBF indicating clearly the formation of hydroxyapatite.

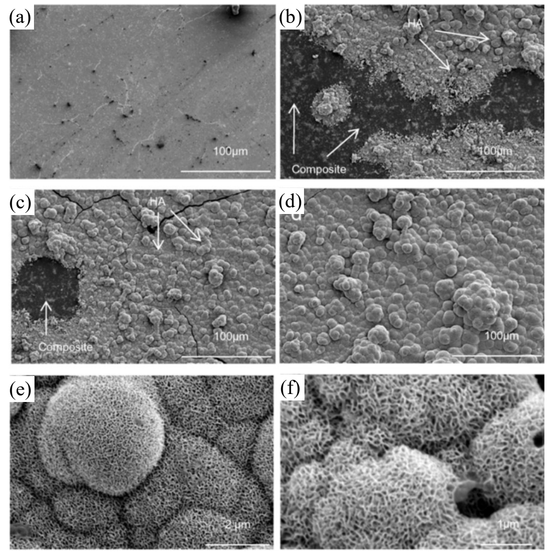

Figure 1. SEM images of the composites after immersion for 1 week in SBF with (a) 0% CaP, (b) 10% CaP, (c, e, and f) 20% CaP and (d) 40% CaP [32].

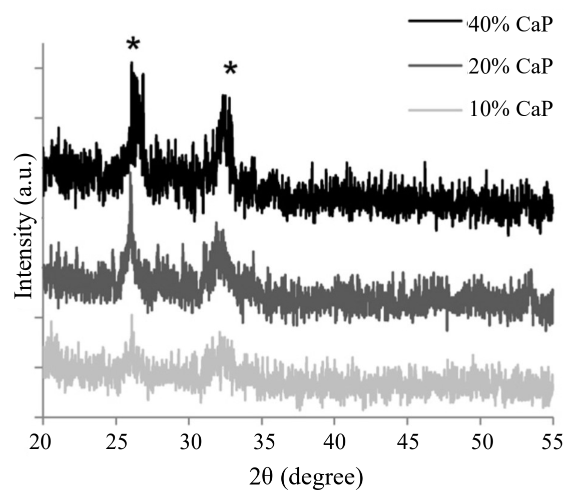

Figure 2. XRD patterns of the 10, 20 and 40% CaP containing composite surfaces after immersion for one month in SBF solution. Stars (*) indicate HA peaks [32].

4. Ceramic/Glass/Glass-ceramic fillers in dental nanocompistes

During the last decades oral health has attracted considerable attention to the researchers. Therefore, dental restorative materials have received specific attention. Several researchers have studied the physical properties of nanocomposites [33]. Extensive research has revealed that nanocomposites can further improve the mechanical properties e.g. compressive strength, flexural strengths and wear resistance. Further, they can provide high esthetics. Many nanocomposites composed of nanomaterials and traditional matrix materials such as metals, ceramics, resin etc. have been widely used in prosthodontics [34]. Effect of particle size and surface pretreatment of ceramic fillers on the properties of dental resin composites was reported by some researchers elsewhere [35]. The type of the filler and surface treatment significantly improves the properties of dental resin composites. Figure 3 shows the microstructure of the microfiller containing composites with and without silanation.

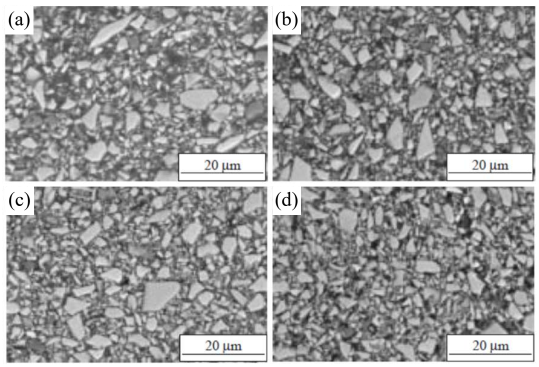

Figure 3. Microstructure of microfiller "K3M" containing composite: (a) without silanation; after silanation with (b) 0.15 wt. % silane, (c) 1.5 wt. % silane, (d) 3 wt. % silane [35].

Khurshid *et al.* [36] reported that the quality of dental biomaterials has been improved by using nanotechnology. Moraes *et al.* [37] has demonstrated that the nanohybrid resins generally present inferior properties than those of the nanofilled composite and almost comparable properties with respect to the microhybrid material. SiO_2 microsphere reinforced dental nanocomposites with low shrinkage were developed by Miao *et al.* [38]. They prepared various resin mixtures having similar filler content (71 wt%). The composite offered low volume shrinkage by changing composition of the resin. The study showed that best compressive strength, depth of cure and the lowest volume shrinkage were influenced by the resin mixture [38].

Boumezgane *et al.* [39] developed bioactive nanocomposites by reactive suspension method for dental application. They prepared hydroxyapatite (HA) filled polymer (PMMA/PHEMA) blends by using reactive suspension method. The decrease in the water absorption values was observed by the presence of HA in the polymer blends. The nanocomposites showed good bioactive behaviour during immersion in simulated body fluid. Dental nanocomposites with nanosilica fillers were investigated by some researchers and reported elsewhere [40]. It was established that the synthesized nanosilica could be used as suitable filler material for the fabrication of dental nanocomposites for tooth-filling applications. Karimzadeh *et al.* [41] studied the effect of thermo-cycling on the properties of commercial silica and zirconia nanofiller containing dental nanocomposite using nano-indentation method. Some samples were stored in distilled water at room temperature while other samples were kept in the ambient condition Another samples were thermo-cycled for 1000 cycles between 5°C and 55°C in distilled water. The samples stored in distilled water without thermo-cycling showed the highest elastic modulus. Further, they noticed that the thermo-cycled specimens in distilled water had the maximum hardness values.

Mohseni *et al.* [42] studied that addition of glassy flakes into the spherical nanosilica particle containing dental composites might improve their mechanical properties. Monfared *et al.* [43] synthesized new dental nanocomposite with glass nanoparticles using amorphous borosilicate glass as nanofillers. Higher volume of glass nanoparticles improved the composite's properties. Liu *et al.* [44] studied mica-apatite porous glass-ceramic filler reinforced dental resin composites. It was observed that the porosity of particles, chemical composition and the porous structure had significant effects on the mechanical properties of dental resin composites. It was shown by the researchers that radiopaque colorless glass-ceramics and a polymerizable synthetic resin composite were useful for dental fillings in order to replace the conventional dental fillings [45]. The compositions of the dental fillings were formulated based on rapidly polymerizable synthetic resins such as acrylate or methacrylate and a pulverulent inorganic material (e.g. glass) embedded in the synthetic resin.

Fluoroapatite-mullite based glass-ceramic filler particles were synthesized by Mollazadeh *et al.* [13]. Strong dependence of the flexural strength on the filler particle composition was noted in this study. On the contrary, filler composition had no effect on the tensile strength and microhardness properties. Guo *et al.* [46] fabricated novel zirconia-silica ceramic nanofiber reinforced dental composites. Reinforcement of dental composites with zirconia-silica ceramic nanofibers could significantly improve the properties of these composites. Efforts to emphasize the mechanical performances of nanofiber and nanotube containing dental composites are demonstrated in a review paper [47]. Nanofiber or nanotube filler reinforced composites showed distinct performances. Ateyah *et al.* [48] showed stable elastic and mechanical behaviors of nano-filled hybrid composite kept under water. The flexural strength of the nano-hybrid materials was reduced due to aging under water whereas all materials showed increased Young's modules values. The morphology and mechanical properties of the nano-SiO_2 filler containing nanocomposite resin matrix were investigated by the researchers [49]. They showed that nano-SiO_2 inclusion significantly enhanced the mechanical properties of resin matrix.

Sachdeva *et al.* [50] had written an excellent review article on nanocomposite dental resin. According to them, nanocomposite may be an ideal material for anterior and posterior applications due to improved physical properties in combination with superior esthetics. Another review paper was written by Govindankutty [51] on the application of nanotechnology in orthodontics. Use of nanotechnology in dentistry was written as a review paper by some researchers [52]. Some researchers have reported about the recent advances and modifications of dental restorative materials [53]. Amdjadi *et al.* [54] described the essential role of filler/matrix interface in case of dental composites. The performance of dental composites depends on polymerization shrinkage stresses as well as degradation of the filler-matrix interface. Use of long chain hydrophobic polymers improves the hydrolytic stability and uniform filler dispersion. Additionally, it reduces the stress evolved from polymerization shrinkage. Further, surface treatments of composite filler particles are very beneficial for restorative composites.

The surface roughness of different restorative composites has been evaluated by the researchers using atomic force microscopy (AFM) [55]. They showed that all the posterior composites revealed surface roughness after polishing as indicated by AFM analysis. Effect of zirconia nano-clusters on the compressive strength of dental composite has been reported by some researchers [56]. They showed that the compressive strength of the dental composites increased with increasing volume percentage of zirconia. Jain and Wadkar [57] investigated the role of nanofillers on the surface properties and the abrasion resistance of the nanocomposites.

5. Properties of ceramic/glass-ceramic filler reinforced dental resin composites

Incorporation of filler into resin matrix improves certain properties such as increased strength, hardness, workability and reduced polymerization shrinkage, thermal expansion and contraction, softening and water absorption. Processing parameters such as monomer and filler composition, temperature and shear rate greatly affect the properties of the composite. It has been observed that increment in filler particle size leads to increase of stress concentration and decrease of flexural strength. Both filler loading and morphology influence the hardness, flexural strength, modulus and fracture toughness of the composites [58]. On the other hand, filler particle size has lesser influence on the properties of resin composite. However, nano-filler particles show superior performance in the composites than micro-particles [59,60]. Tanimoto *et al.* [61] studied the bending properties of the dental composites using finite-element method (FEM), which indicated that the bending properties increased with increasing the amount of filler. Considerable increment in the fracture toughness, flexural and compressive strength, hardness and modulus values was achieved by the addition of apatite and titanium nanotubes to resin based cements without changing biocompatibility [62,63].

Decrease of filler particle size (<1 micrometer) and lower filler loading help in clinical development and maintenance of smooth surface with microfilled composites than conventional composites [64]. Wear resistance of composite resins can be enhanced by using higher volume functional silane treated microfiller particles. Further, the filler size and content controls the abrasion resistance of the composites [65-67]. Increased filler content in composite resins guarantees enhanced strength, wear resistance, postoperative sensitivity and durability [68].

It has been observed that filler content has certain effects on optical properties of the resin. There is a direct relation between filler content of resin composite and optical scattering. The effectiveness of optical scattering is related to filler size and shape [69]. The volume fraction of the filler and matrix in composite resins affect their color. Presence of TiO_2 nanoparticles yields human enamel like look [70, 71]. Juhasz *et al.* [72] prepared composites consisting of apatite-wollastonite glass-ceramic based filler and polymer matrix as implant materials. Increment in the filler content resulted in enhanced mechanical strength. The mechanical properties were found to be slightly decreased with increasing the filler particle size. Xu *et al.* [73] studied dental resin composites wherein dental resin was reinforced with high strength, elongated ceramic single-crystalline whiskers. Silicon nitride whiskers were coated by silica particles. The coated whiskers were silanized and mixed with resins. Significant increase in strength, toughness and enhanced resistance to micro-cracking and contact damage was achieved.

It was observed by some researchers that the mechanical properties and fracture toughness can be improved by addition of ceramic nano-fibers in dental composites and thereby, extending their service life [74]. They characterized the dental composites reinforced with different fillers such as in various amounts. Reinforcement of dental composites with zirconia-silica (ZS) or zirconia-yttria-silica (ZYS) ceramic nanofibers can significantly improve the mechanical properties of the dental composites. All ZS reinforced composites showed significantly higher fracture toughness [74]. Novel dental restorative composites have been produced using hydroxyapatite (HA) whiskers by Hongquan and Darvell [75] and reported elsewhere. They established that the efficiency of reinforcement depends on filler morphology. It had been observed that hydroxyapatite whiskers has good wettability with polymer leading to toughening as compared to that of the HA nano-scale powder.

Xavier *et al.* [76] investigated the effect of silane concentration, filler size and distribution on the chemical-mechanical properties of the composites. Although 3% silane content enhanced the initial strength but degradation occurred after water storage. The filler distribution did not affect the biaxial flexural strength. However, it was noted that that increase in the filler size improved the fracture toughness significantly [76]. It was found that the failure of composites was materialized on account of failure of the resin matrix and or the filler-matrix interface [77]. Incorporation of 5 wt% of nano-fillers into conventional composite resins did not result in any changes in their flexural and tensile strengths. On the contrary, the tensile and flexural strengths increased when the content of nano-filler increased to 10% [78].

Many types of nanofillers have been used in dental composites. The mechanical properties of dental nano-composites are superior to microfilled dental composites. Effective stress transfer from resin to nanofiller results in good flexural strength whereas increased filler volume fraction leads to high hardness. The composite strength depends on the load transfer between filler and matrix while stiffness is dependent on particle loading. The filler particle shape also has a remarkable effect on some mechanical properties of the composites [79]. Sakr *et al.* [80] studied the nanosized zirconia resin composite showed high wear resistance. SiO_2 nanofiber-containing composite resins were promising material with long-term durability [81]. SiO_2 nanofibers improved the overall performance of composite resins compared to the SiO_2 nanoparticles. Some researchers [82] investigated the impact of preheating of TetricEvoCeram (TEC), FiltekSupremeXT (FSXT), and Venus (V) on mechanical properties.

In vitro tests were performed to compare the wear properties of indirect dental composites and all ceramic materials [83]. Indirect dental composite is relatively more superior than all ceramic restoration. Therefore, indirect composites are promising for long life dental restorations. Esteban-Tejeda *et al.* [84] fabricated two types of antimicrobial ZnO and CaO containing glass fibers by a laser spinning technique. ZnO glass fiber composite is non-cytotoxic having requisite antimicrobial property after immersion in human saliva. Thus, these composites with antimicrobial properties have great potential in the field of dental applications. Zha *et al.* [85] showed that the inclusion of the nano- SiO_2 particles in the dental rein composites can apparently increase the hardness and elastic modulus. Based on the experimental results, it was observed that the dental resin composites having 4 wt% nano-SiO_2 particles had the best elastic recovery behavior. The improved mechanical properties were ascribed to the role of the nano-SiO_2 particles as the nucleating agents during crystallization of the dental composite matrix and thereby, triggering the enhanced crystallinity.

6. Conclusions

Current literature reveals that ceramic/glass-ceramic fillers has bright prospect for the dental applications. Researchers are trying to explore new types of reinforcements for dental resin composites. There is a trend to use small filler particles to achieve the requisite properties. Recently, fillers are being developed having bioactivity and antibacterial property. More thrust is needed to optimize interactions between the resin matrix and the filler to achieve desired properties, which will lead to evolve new generation safe, strong dental fillers and composites. However, further research is more anticipated to improve the performance of the commercially used fillers.

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