



The Transition from Conventional Methacrylate Based Composites to Ormocer Based Composites: A Review of Literature

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ABSTRACT

Resin composite materials have become well-accepted and widely used as direct restorative materials. Nowadays, they are replacing the traditional mercury-containing dental amalgams owing to their superior features. However, the conventionally-used dental composite resins possess several drawbacks, most noticeably polymerization shrinkage and its associated shrinkage stresses. These shortcomings have led to great evolution in the development of enhanced composite restorative materials. This review surveys composition, advantages and deficits of resin composites, then highlights recent developments aimed at enhancing the properties of composite restorative materials. Finally, a special attention is given to the development in the polymeric matrix formulation providing a new low-shrink composite known as Ormocer analyzing its composition, behavior, and properties.

Keywords: Resin composite; Polymerization shrinkage; Ormocer; NanoCeramic composite

Introduction

Dental caries is one of the most widely spread diseases all over the world. It has been estimated that around 44% of the world population suffer from untreated dental caries.⁸⁰ Despite the great progress in caries prevention and enhanced oral hygiene practices, Population-based studies showed that the prevalence of dental caries stays relatively high.²⁶ It still affected 2.3 billion people worldwide in 2017.⁶⁴ Based on this fact, operative dentistry plays a major role in oral healthcare by treating various carious lesions in its different forms.⁵⁰ Dentists recommend elimination of dental caries and restoring the resultant cavities directly with adequate filling materials.⁸⁵ The management of dental caries through the placement of direct restorations is the most significant component of the daily work of the operative dental team.²⁶ Direct restorations have been widely used for treating decayed teeth having relatively low cost, more sound tooth structure preservation, and acceptable clinical performance in comparison to indirect restorations.⁷ Direct filling materials have differed in their properties like durability, biocompatibility, and safety.⁴³ Some of them necessitated conventional designs for the prepared cavities, whereas others were designed to be used with adhesive dentistry.⁷⁹

Commonly, dental amalgam was used as a restorative material for posterior teeth.⁶⁰ However, its use has been decreased progressively due to the need of for esthetic restorations and the tendency for minimally invasive approach.⁶⁹ As well, The Minamata Convention on Mercury has stated a global reduction in the manufacturing and usage of mercury-containing products such as dental amalgam and so, the usage of resin composite increased as substitute for dental amalgam.⁵¹ At first, chemically cured resin composites were introduced, but their unacceptable clinical performance hindered their usage in posterior teeth. With further developments in dental resin based composites, light-cured resin composites were introduced to be used in restoring posterior teeth.⁸⁷

1. Advantages of Resin Composites

Resin composite restorative materials have gained popularity owing to their superior characteristics, such as non-toxic and antibacterial properties, availability of different shades which afford unlimited possibilities for this material to match the tooth shade,^{4,85} possibility of repair, adhesion to the tooth structure which obviates extra retention means and undesirable removal of sound tooth structure thus reinforcing the remaining tooth structure and achieving acceptable prognosis.^{44,84}

The presence of minimally invasive dentistry and the enhanced adhesion to tooth structures have allowed resin composites to show expected long-term success comparable to dental amalgam.^{8,50,72} Lynch et al.⁵⁰ reported that the Academy of Operative Dentistry European Section (AODES) has considered resin composite restorative materials of proper composition and characteristics to be the ‘‘material of choice’’ for replacing amalgam. Gaengler et al.³³ reported a successful 10 years performance of composite restorative materials in small to moderate-sized restorations. Rodolpho et al.⁷⁰ also reported acceptable overall success rate in a 22- year study of two light-cured composites restoring class I and II cavities. Moreover, Opdam et al.⁶¹ even reported better survival rate of composite restorations as compared to amalgam restorations after twelve years of observation.

2. Composition of Resin Composites

Resin composites are composed of three distinct phases: the organic resin matrix phase, inorganic filler, and the filler-resin interface, each with its own impact on the resin composite properties.²¹ In addition, resin composites contain also smaller amounts of additional materials that influence the overall material features such as additives, stabilizers, and pigments.^{6,58} The organic matrix of resin composite is composed of a system of mono, di or trifunctional monomers, a free radical polymerization initiation system, which in photo-activated resin composite is mostly (camphorquinone) used in combination with a tertiary aliphatic amine reducing agent.⁴⁰ Upon light exposure, monomers are converted to a highly cross-linked three-dimensional network polymer.³¹ Light stimulates active centers formation, - typically radicals-, which motivate polymerization.²⁰

The inorganic fillers generally determine the mechanical properties of dental composites.²⁸ The type of incorporated fillers affect radiopacity, modulate translucency and enhance handling characteristics, particularly consistency and polishability.⁵⁵ Both fillers’ size and content influence light dispersion through the resin composite as the inorganic fillers are able to scatter light.⁸¹ As well, filler loading has a great impact on the material’s elastic modulus, strength, and wear resistance.⁴⁹ Finally, the filler-resin interface organosilane or coupling agent- links both organic and inorganic components together forming a strong bond.⁵ If the interface between the organic and inorganic phases broke down, the stresses generated under load would not

be evenly distributed through the restoration. As a result, the interface acts as a primary source of fracture leading to the failure of the restoration.⁴²

3. Drawbacks of Resin Composites

The inherent polymerization shrinkage was considered one of the serious drawbacks of resin composites, which occurs in the range of 2-5 vol%.¹³ Polymerization shrinkage occurs as the space between monomers decreases during the polymerization process. This happens due to the conversion of the weak Van der Waals forces which tie the monomers to covalent bonds.⁴⁵

The total polymerization shrinkage can be divided into two phases; the pre-gel and post-gel phases. Throughout the pre-gel phase, the composite can flow, thus relieving stresses within the structure.²⁵ The material's viscosity increases gradually during monomer conversion and the composite material converts from a viscous fluid state to an elastic gel "gel-point".⁷⁵ After gel-point, the elastic gel transforms to solid-state "Vitrification" which is characterized by elastic modulus development.³⁵ The composite flowability ceases and loses the ability to compensate for shrinkage stresses.²⁵ The accompaniment of volumetric curing contraction with the elastic modulus development and the restriction of the volumetric changes from surrounding tooth structure generate residual shrinkage stresses.¹¹

The magnitude of the generated shrinkage stress depends on many factors such as the compliance of the dental substance and the ratio between the unbounded to the bonded surfaces defined by C-factor.² The generated shrinkage stress is also largely influenced by the visco-elastic behavior of the resin composite, regarding its flow capacity in the initial stages of the polymerization, and the elastic modulus development during the formation of the polymer network. Lins et al.⁴⁸ stated that there was a strong correlation between polymerization stress and post-gel and a weaker correlation with total shrinkage suggesting that differences in polymerization kinetics and polymer composition affect their viscoelastic behavior and conversion rate.

Additionally, the generated stresses are affected by material composition, filler content, depth of cure, and degree of monomer conversion.⁹ Calheiros et al.¹⁷ suggested that the relationship between the contraction stress and degree of conversion is material dependent. Furthermore, the larger the cavity, the greater the shrinkage that will happen within the restoration as polymerization shrinkage is volume-dependent.³⁷ Braga et al.¹⁶ stated that the shrinkage stress in composite restorations is affected by both diameter and depth.

Shrinkage stresses generated at the tooth-restoration adhesive interface may cause loss of marginal integrity.⁵³ Subsequently resulting in interface fractures and micro-leakage.⁸² Papadogiannis et al.⁶³ reported that the setting shrinkage characteristics of resin composites influence their marginal adaptation with dentin. Moreover, Peutzfeldt and Asmussen⁶⁵ reported polymerization shrinkage and flow of the resin composites as important determinants of gap formation. Not only contraction stresses may immediately affect the marginal quality, but also exaggerated gap formation may occur on the long run as a response to generated stresses during mastication.¹⁰

In addition to polymerization shrinkage, resin composites had numerous drawbacks such as inadequate proximal contact, the difficulty of achieving optimum adaptation clinically,¹⁹ stickiness and subsequent handling problems,¹² and thermal expansion mismatch.⁸⁸ Additionally, low wear resistance - compared to metallic restorations-has been reported as a shortcoming of composite restorations. Ferracane²⁹ reported that the wear resistance of composite restorative materials could still be a concern especially for large restorations in direct occlusal contact, or for patients with abnormal masticatory behavior. Moreover, resin composites may be related to potential cytotoxic effects as a result of elution of unpolymerized monomers from

the composite.³⁹ Durner et al.²³ reported an inverse correlation between DC and elutable monomers in resin based composites demonstrating that the higher the amount of unpolymerized monomers indicates for the lower the degree of monomer conversion.

4. Developments of Resin composites

Different Modifications in composition and placement techniques have been introduced to overcome the resin composite drawbacks.^{3,57} The initial tendency was to improve the filler technology for enhancing the mechanical and esthetic proprieties.⁷⁸ enhancing filler system has also reflected on reducing the resin fraction which in turn decreases polymerization shrinkage.²⁰ An initial effort has concentrated on reducing the size of the filler particles from 10 to 50 μ m, utilized in earlier “macrofilled” composites, to small-range size producing the microfilled resin composites aiming for improved polishability and wear resistance.³⁰

Since Nanotechnology has been incorporated in the dental industry, the production of functional filler particles in the range of 100 nm could be achieved.⁵² Hence, a markable modification in the composite properties has been achieved through the integration of nanoparticles with submicron particles producing nanohybrid composites.⁷¹ The nanohybrids systems permitted high packing of filler particles, which in turn improved workability and mechanical properties.⁷¹ Ilie et al.³⁸ reported higher mechanical properties for several tested nanohybrid resin based composites which exceed the average mechanical properties of microhybrid tested composites. Moreover, nanofilled composites, containing only nanoscale particles had been also produced.³⁰ Recently, the development of nanoclusters which combine nanoparticles and their aggregations have been introduced to combine high aesthetics and better clinical performance.⁸³ Pre-polymerized fillers (PPF), obtained using ground polymerized composite, were also introduced in an attempt to decrease the stress generated from polymerization and enhance polishability compared with earlier produced composites.⁶⁸

Generally, the resin composites based on nanotechnology have produced noteworthy improvements in wear resistance and remarkable evolution in mechanical properties.³² Although the mechanical properties of composite resins are essential for the longevity of the restorations, mechanics could not be the only factor to be considered. More attention had to be paid to the composite polymerization and the resin matrix to reduce the deleterious effects of polymerization shrinkage.⁶² Consequently, greater attention was focused on the modifications in the resin matrices formulations.³²

The resin matrix of conventional resin composites had been based initially on methacrylate chemistry. Since the development of resin composite in the late 1950s, Bis-GMA had been the commonly-used monomer in early composite formulations.⁷⁸ The high viscosity of Bis-GMA has negatively affected the conversion rate.³⁴ This was supported by Gonçalves et al.³⁶ who reported a reduction in conversion rate with BisGMA due to the high viscosity caused by the hydroxyl groups. Braga et al.¹⁵ stated that regarding the inherent volumetric shrinkage of BisGMA-based materials, the objective of enhancing degree of conversion and elastic modulus for better clinical performance may be achieved by maximizing viscous flow. Accordingly, It was substantial to add low molecular weight monomers in the matrix such as trimethylene glycol dimethacrylate (TEGDMA) and 2-hydroxyethyl methacrylate (HEMA) to obtain the appropriate viscosity.³⁴ As well, to enhance cross-linking, and to allow for more fillers incorporation.⁴⁷ However, these diluents added monomers increased water sorption of the composites and the polymerization shrinkage.⁷⁸ As a consequence, the search for new monomers was much needed.³²

5. Ormocer (organically modified ceramic)

A new packable restorative material called Ormocer -organically modified ceramic- has been introduced in 1998.⁴¹ Ormocer was developed by Fraunhofer Institute in cooperation with the dental industry.⁵⁹ Ormocer is an inorganic-organic copolymer.⁴¹ It consists of three main components organic polymers, inorganic components, and polysiloxanes.⁷⁶ Organic polymers have their impact on the polarity and the network formation.¹⁸ The inorganic components influence thermal expansion and chemical stability. The polysiloxanes are responsible for the elasticity and interface characteristics.⁷⁶ The synthesis of ormocer begins through a sol-gel process.⁵⁶ The hydrolysis and the polycondensation reactions result in a matrix of inorganic silica chain backbones with light- curable side chains.⁵⁹ The resultant oligomers can substitute the classic monomers in the resin composite and a network of three dimensions is constructed by these functional groups polymerization.⁵⁴ Different types of functionalized inorganic fillers, monomers, additives such as initiator and activator are incorporated to achieve the paste-like consistency of the ormocer based composite.⁵⁶ Metal alkoxides such as Al, Ti, or Zr alkoxides may be also condensed in addition to alkoxy silanes.²⁴

Ormocer-based composites are generally characterized by reduced shrinkage stress.⁴⁶ Yap and Soh⁸⁶ reported low polymerization shrinkage values for ormocers. The ceramic polysiloxane matrix account for lower shrinkage in comparison to the classic monomer matrix seen in conventional composites.⁶⁶ The polymerizable side chains linked to the polysiloxane account for the lower volumetric shrinkage and decreased microleakage.⁷⁶ Further Incorporation of filler particles changed volumetric shrinkage from 2–8% to 1–3% after fillers incorporation. The availability of abundant polymerization opportunities of ormocer allows curing without leaving residual monomer which in turn has greater biocompatibility.⁴¹ Polydorou et al.⁶⁷ stated that ormocer based materials have shown a significantly lower elution of monomers as compared to traditional composites.

Ormocer, when compared to conventional methacrylate-based composites, has shown certain advantages such as higher wear resistance, lower water sorption, better manipulation characteristics, and superior esthetics with comparable clinical performance.²² Bottenberg et al.¹⁴ reported in a three year study that the ormocer-based composites performed comparably to the conventional BisGMA-based composite in occlusal stress-bearing cavities. In 2015, a resin composite for direct restoration has been launched in the market, supposedly free of any classic methacrylate-based monomer, completely based on the Ormocer technology, and marketed as Admira Fusion (Voco GmbH, Cuxhaven, Germany).⁴⁶

Ormocer resin has also been used in Ceram X (Dentsply Sirona GmbH, Konstanz, Germany). By combining both ormocer technology and nanotechnology, this organically modified nanoceramic composite was developed.⁵² Ormocer, in the mentioned composite type, was used in combination with other dimethylacrylates in an attempt to decrease the viscosity of the composite material.⁴⁶ Introduction of this ceramic-based composite to the market was considered an advancement in the field of nanohybrid composites with pre-polymerized fillers.²⁷

In the manufacturing process, the fillers were covered in a layer of pre-polymerized resin matrix and then applied to the resin mixture with additional nanofiller particles.²⁷ This composite material has organically modified spherical ceramic nanoparticles and nanofillers combined with conventional glass fillers and merges hybrid composite filler technology with advanced nanotechnology resulting in nanoceramic technology.⁷⁴ The organically modified resin matrix and the spherical pre-polymerized nanofillers led to improved mechanical strength and low contraction stress during the polymerization process.²⁷ Also, it exhibited superior esthetics with superb handling properties.⁷⁴

Al-Qahatani et al.¹ evaluated the degree of conversion (DC) of nanoceramic (Ceram X, Dentsply DeTrey GmbH, Konstanz, Germany) and micro-hybrid composites (Spectrum TPH, Dentsply DeTrey, GmbH, Konstanz, Germany) activated by different polymerization modes at different intervals. In this *in vitro* study, 80 cylindrical samples were performed using (5 × 2 mm) Teflon molds and photopolymerized using a light-emitting diode (LED). The degree of conversion of the top surfaces was measured by a Fourier-transform infrared spectrophotometer (FTIR) using a direct technique; attenuated total reflectance (ATR). The study showed that the degree of conversion for the tested samples ranged from 44% to 55% for Ceram X and from 42% to 45% for Spectrum TPH. The author stated that the nanoceramic composite has shown higher DC in comparison to the micro-hybrid one at different polymerization modes.

Fanoni et al.²⁷ evaluated the degree of conversion (DC) of three nanohybrid composites (Ceram. X, Dentsply DeTrey GmbH, Konstanz, Germany), two nanohybrid composites (Tetric EvoCeram, Ivoclar Vivadent AG, Schaan Liechtenstein), and two nanofilled composites (Filtek TM Supreme, 3M ESPE, St Paul, MN, USA) with equivalent dentin and enamel shades. In this *in vitro* study, DC was assessed by infrared spectroscopy. The study showed that only Ceram X showed DC higher than 60% in comparison to other composites of corresponding shades. The author concluded that regarding DC, Ceram X composites showed the best performance.

Kalra et al.⁴¹ compared the marginal sealing ability of ormocer with a hybrid composite. In this *in vitro* study, 44 class I occlusal cavities of 1.5 mm depth were performed in extracted premolars and were restored by randomly distributed adhesive and restorative materials. The results of the study revealed that ormocer based restorative material has shown the least marginal leakage in comparison to hybrid composite.

Poggio et al.⁶⁶ evaluated the impact of immersion in an acidic drink on the microhardness of one nanohybrid ormocer-based composite (Admira Fusion, Voco GmbH, Cuxhaven, Germany), one nanoceramic composite (Ceram X Universal, Dentsply DeTrey GmbH, Konstanz, Germany), one nanofilled composite (Filtek Supreme XTE, 3M ESPE, St Paul, MN, USA), and one microfilled-hybrid composite (Gradia Direct, GC Corporation, Tokyo, Japan). In this *in vitro* study, 30 specimens of each composite were performed and divided into 3 groups; the control group, after 1-day immersion, and after 1-week immersion. Vickers microhardness measurements were obtained using a microhardness tester. The study showed that the nanohybrid ormocer-based composite and the nanofilled composite showed the best behavior. The Ceram X Universal (nanoceramic composite) although reached lower hardness values than the previous materials, it showed well resistance to the one-week immersion in acidic drink

Tagtekin et al.⁷⁷ evaluated hardness, surface roughness, and wear resistance of an ormocer based composite (Admira, Voco GmbH, Cuxhaven, Germany) and a micro-hybrid composite (Amelogen, Ultradent, Utah, USA). In this *in vitro* study, cylindrical blocks of both materials were prepared in 3 mm in diameter, and 2 and 5 mm thickness. Blocks were cured by two different polymerization systems. With a load of 600 mN, Vickers microhardness was measured. The study showed that ormocer based composite showed the highest hardness values in all polymerization types at the top surface. The author concluded that ormocer had higher microhardness and wear resistance when compared to a hybrid composite.

Cavalcante et al.¹⁸ evaluated the surface integrity of solvent-challenged ormocer-based composites through surface-hardness measurements. In this *in vitro* study, Disk specimens (4mm×2mm) of a 100% pure ormocer-based composite (experimental ormocer, Voco GmbH, Cuxhaven, Germany), a mixed dimethacrylate-ormocer-based composite (Admira, Voco GmbH, Cuxhaven, Germany), and two commercial

dimethacrylate-based matrix composites (Grandio, Voco GmbH, Cuxhaven, Germany) and (Premise, Kerr Corporation, USA) were prepared, photoactivated by different light exposure modes and measured at different periods. The resultant data revealed that the pure ormocer-based material had the lowest percentage of hardness change and showed more resistance to solvent degradation than the other composite types, regardless of the light exposure method, thus, better surface integrity.

Sahoo et al.⁷³ evaluated the surface hardness of a Nanohybrid ormocer-based composite (Admira Fusion, Voco GmbH, Cuxhaven, Germany), a microfilled hybrid composite (Gradia Direct, GC Corporation, Tokyo, Japan), and Nanofilled composite (Filtek Supreme XTE, 3M ESPE, St Paul, MN, USA). In this in vitro study 30 cylindrical specimens were performed, photopolymerized, stored in a dark container, and kept dry at room temperature for 24 h before testing. Vickers hardness measurements were assessed with a microhardness tester and the resultant data was statistically evaluated. The author concluded that nanohybrid ormocer-based composite has the maximum Vickers hardness number (VHN) in comparison to tested materials representing the maximum degree of conversion and improved clinical performance.

Conclusion

Currently-used composite restorative materials provide an alternative to conventional mercury-containing dental amalgams. However, they still suffer from a variety of drawbacks, particularly the polymerization shrinkage and its associated stresses. Recent improvements have significantly focused upon this issue through a number of advanced strategies, such as modifying the resinous matrix of composite materials. Ormocer has been introduced and employed either combined with conventional methacrylates or solely. In reviewing advantages and properties of Ormocer and according to the literature, it showed promising extent of success and appeared to be an acceptable alternative to the Bis-GMA based composite materials.

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Disclosure

The author reports no conflicts of interest in this work.

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