

Degradation and failure of dental composite materials

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6.1 Introduction

Over the past decade, there has been a drastic improvement in the clinical performances of the dental composite materials that may be possible with different modifications at every step of polymerization that might result in better color stability of the dental material. Incorporation of various binders, such as fillers, into the composite material will provide adequate strength and bearable wear resistance, and the required translucency of the composite material. After decades of development, dental composite material has found its application as a restorative material in the field of dentistry [1].

Dental composite materials are complex in nature. These are used as a colored material for filling. We need molecular particulates or fillers that can produce cross-linking in the polymer matrix during the polymerization process from demethacrylate resin as a monomer unit. The bonding of these reinforcements has gained worldwide acceptance as a restorative material for applications in dentistry. Fillers are used generally during the polymerization of dental restorative material in order to provide multiple properties to the dental restoration material such as strengthening [2], maximum stiffness, and dimensional change reduction when heated. The amount of additional filler is directly proportional to the enhancement of physical and mechanical properties of polymerized dental restorative material in dentistry. The initial phase of fillers used in dental composites includes crystalline or fused quartz, lithium alumina-silicate glasses, or borosilicate that were milled into particles of various sizes ranging from 0.1 to 100 μm added into monomer to make a paste that is hardened after drying, and is used as a dental restorative material. The fused quartz has the drawback of being not radiopaque, and it could be possible that these particles are not hard enough for the polymer matrix, which would result in degradation of the polymer matrix before the degradation of particulates. In the current scenario, the main reason of failure of dental composite material is the degradation of the polymer matrix before the particulate, in addition to the interface breakdown between the matrix and the fillers used in the dental restoration material. This chapter concludes with the impact of degradation of dental composite material on the development of dentistry [3, 4].

During the past 100 years, porcelain has been used in dentistry. The major advantage of it is aesthetics, while its weak point is its brittleness for load-bearing restorations. This was technically sensitive and innovative with respect to conventional powder build up processes. That's why "metal-ceramic restorations" are made by porcelain-fused-to-metal (PFM) restorations as the first preference for prostheses for aesthetic use, for their fit to the abutments, and their durability [5, 6].

6.2 Characteristics parameters

In dental science, polymeric dental restorative material is more popular than ceramic and metallic material [7]. This is due to easy fabrication, processing, and aesthetic aspects, in addition to being cost effective and easily available. The main topic of concern is the low resistance property of polymer-based composite material as a dental restorative. It's challenging to perform the assessment of dental material in terms of the erosion rate for different oral conditions [8, 9]. The main role for erosion in dental material is abrasion, which will vary in nature due to the different types of abrasion between the dental material, teeth, and the intake of different food particulates. The other parameters that will ultimately affect the abrasive erosion of the dental composite material include food and temperature, variation in food, and type of intake of food. There are two types of abrasive wear in dental science, two-body abrasive wear and three-body abrasive wear. The two-body abrasive mechanism includes the plastic deformation or fracture that occurs due to damage of the soft surface with the harder surface, while the three-body abrasives occur when the harder particulate comes in between the two soft surfaces [10, 11]. The example of two-body abrasive wear is investigated in dry conditions, and with distilled water cold drinks; whereas investigation of dental material with any food slurry is used in the example of three-body abrasive wear. Abrasion is the characteristic parameter of dental composite material, and it depends upon the physical and chemical properties, as well as on coefficient of friction between the surfaces, as reported previously [12]. The major components needed for composition of dental composite material include polymer matrix, filler particulates, a filler coupling agent, and an additive. Another characteristic parameter that should be taken into account is aesthetic beauty, or translucency of the dental restorative material. In order to have aesthetic beauty of the dental composite resin, the required parameters are the shape, shade, and contour characteristics, which should match well with the adjacent teeth [13, 14]. This occurs through idealization, which harmonizes the optical property of the dental restoration material with adjacent natural teeth. In order to have better development and enhancement of dental restorative resins, the dental material should be available in a wide range of restorative shades and opacities, so that the duplication with the natural teeth is possible, and the aesthetic beauty will be maintained. The optical properties of enamel and dentin are combined so that replication with the naturally existing teeth can be easily achieved, which will require translucency at various levels of polymerization. There is one more technique known as "layering" that is very popular, which enables the layering of more translucent

materials over a more opaque composite material in order to create a height of depth from within the restoration. It's helpful to measure the translucency of a particular composite resin in relation with the natural tooth structure [15, 16].

For the requirement of a smaller filling of restorative material, it is easy to achieve the required composite with an adequate level of opacity. However, in complex aesthetic conditions, one has to follow the "layered architecture" of the composite material [17]. There are a total of 39 dental restorative composites [18].

The calculation of the translucency parameter was evaluated by the difference in color of the specimen against the standard white background [18].

$$TP = \left[(L_w * -L_B *)^2 + (a_w * -a_B *)^2 + (b_w * -b_B *)^2 \right]^{1/2}$$

where W=value of the individual sample against the standard white background, B=value of the individual sample against the black background.

A higher value of T.P. is required for higher translucency. The analysis of data is done through Takey's tests and analysis of variance (ANOVA) ($P < .05$). In general, the conclusion of the study of translucency as the characteristic parameter reveals that the "opaque or dentin" composites have relatively low levels of translucency. The "universal or dentins" represent intermediate translucency. Enamel composites have the highest translucency.

6.3 History of dental composite material

For the past 60 years, composite material has been used for direct restorations in anterior and posterior teeth. Composite resin requires very little preparation, and can be used to preserve tooth structure and natural-looking teeth. It may also replace silver amalgam for direct restorations [19].

Silicate cement was the first developed material used for direct aesthetic restoration. Cement was formed from liquid phosphoric acid and alumina-silica glass, and was developed in the late 1800s. According to O'Brien, the silicate was degraded due to its solubility in oral fluids [20]. The German chemists developed self-curing acrylic resins in 1941. The methacrylate polymerization reactions were initiated by the use of benzoyl peroxide and tertiary amines. It led to the discovery of acrylic filling materials in 1948, for example, Sevriton. In this method, methyl methacrylate and polymethyl methacrylate were mixed together, and the major drawbacks were a high level of polymerization shrinkage, increased thermal expansion, absence of tooth structure adhesion, stiffness, and decreased color stability [21].

In the mid-20th century, acrylic resins replaced silicate cements as acrylics, which offered benefits over silicates, but bending was difficult. They first introduced Bunocore, an orthophosphoric-acid technique, which improved, but did not completely solve the adhesion problems. In 1962, Bis-GMA monomer developed by Bowen improved the physical properties of acrylics [22, 23]. In 1970, urethane dimethacrylate-based chemistry was introduced. Urethanes are a series of monomers

that incorporate a urethane group into the backbone of a dimethacrylate monomer molecule. In 1977, light-activated initiator methyl-p-toluene was introduced to eliminate the porosity in the paste mixing system, which gave the dentist better control over the setting and placement of the material. In 1990, in order to eliminate polymerization of methacrylate-based monomer materials, scientists used monomers that do not decrease in volume on polymerization. Nowadays, composites' physical properties and their clinical applications are increased by using filler technology and initiation systems [24–26].

In 1998, a comprehensive study was attempted to evaluate the clinical performance of two amalgams and three posterior resins. The results showed that the materials were degraded and were low on wear resistance. The conclusion showed that posterior composite material can be replaced, or it can be a good alternative amalgam. In fact, in 1999, about 26 million composite resins were introduced in the U.S., compared with the 71 million amalgam restorations. The reason for the gaining importance of the former is due to improvement in clinical performance, better aesthetic aspects, better tooth coloration compared with the naturally existing teeth [27]. After this, the use of amalgams became negligible, and was only substituted as dental composite material for class II cavities. The restoration of cavities by amalgams with class II was used for the shortest time (5 years), and class III restoration was used for 7 years [28].

Nanotechnology has been a boon to the field of material science. The properties of the material when implemented with nanoscience produce remarkable results. Nanocomposites are being used extensively in dental science. Fillers used in dental composites are given in table that follows [29]. The current dental composite material includes a blend of hard inorganic particles that are bound together with a soft resin matrix. Generally, it includes three main components.

1. (i) Resin matrix composed of 2 or more monomers; (ii) an initiator system that will enable free radical polymerization, and also a stabilizer that will eventually increase the storage stability of the unused resin composite and maintain the chemical stability of the used resin composite.
2. Inorganic fillers, for example, quartz, glass, fused silica, and so forth.
3. Coupling agent

The properties of dental composite materials depend upon the preceding three parameters. The parameter should be carefully considered, and implementation for the dental composite material should be done perfectly so that the required properties can be achieved [30].

In spite of various advantages, and the versatile nature of dental composite material, the restoration of dental adhesives is not resistant to failure. Through different clinical studies, it was found that there was decreased durability and an increased failure rate of dental adhesive restoration as compared with dental amalgams. The cause of failure is secondary caries and fracture restoration. The factors that aid in dental failure due to secondary caries are decreased antimicrobial activity, polymerization shrinkage, accumulation of plaque, micro-leakage, and gap formation. Recently it was concluded that oral biofilms play an important role in the development of secondary

caries. The adhesive interface is degraded in the presence of oral biofilms, along with sucrose. The degradation is least when there is a lack of sucrose.

6.4 Current scenario of dental composite material

The resin-based composite is now approaching a half century in clinical research, which makes it possible to conclude the “development cycles” of dental composite materials. In the mid-1990s to 2000, in order to reduce the postoperation crystal deflection, interfacial gap, and sensitivity, the research was directed at reducing the shrinkage of polymerization [31].

In this decade, the popular technique among the researchers in clinical areas is the use of a bulk-fill composite that will ultimately reduce the time required to insert the composite material into the prepared cavity. The use of restoration resin dental composite material is becoming more and more popular and ubiquitous, and results in a reduction of service time due to the development of new caries lesions at the interface level of tooth restoration (secondary caries), and it is also due to material fracture [32]. The material fracture is not always due to deficiency in material, but it can also be due to lack of skills of the professional, and it can also be due to unawareness of good dietary and oral hygiene habits by the patient. Therefore, steps have been taken to increase the levels of fracture toughness of material used and incorporation of protective mechanisms to have better longevity of the restorative dental material [33, 34]. The following explains the current scenario of dental restoration material technology.

- (1) Self-adhesive properties will eventually simplify the restorative procedure.
 - (2) Steps will eliminate fracture toughness of composite bulk materials.
 - (3) Defense from new caries lesions that are present at the tooth-restoration interface will be created using bacterial agents.
1. Self-adhesive composite (SAC): SAC was introduced in 2009. There is a difference between SACs and self-adhesive resin cements (SARCs); the former neither contains fluoride-releasing glass fillers, nor does it undergo acid-base neutralization reactions. SAC pertains to self-etched adhesive s/s because of the presence of acidic monomers, for example, carboxylic methacrylates or phosphate ethyl methacrylates (BMEP). In order to enhance the wettability of dental composite material on the dentin surface, hydroxyethyl methacrylate (HEMA) is added to the SAC [35].
 2. Fiber intercalated composite: the current value of fracture toughness (K_{Ic}) value ranges from 0.9 to 1.8 MPa, which is insufficient to render the bulk fracture. Inclusion of fibers such as fuse silica, glass, quartz, and so forth can enhance the level of strength of dental composite material [36, 37].
 3. Self-healing composites: the main drawback of any restoration material is the fracture of the dental composite material. There can be many reasons behind this, and the microcracks can also be propagated due to unawareness of healthy dietary and hygienic habits of patients. The crack protrudes into it, and will eventually damage the entire restoration of the cavity. Therefore, the propagation of the crack should be halted before its extended propagation. The self-healing mechanism includes the biomimetic models that can automatically repair the system within the living cells, for example, like the healing of a broken bone. Glass ionomer cement has been recently developed as a self-healing dental composite [38–40].

6.5 Stability of dental composite materials

Composites were introduced in 1960. Efforts have been made to enhance the durability of composite restorations. Hence, there is a need to improve the optical properties of these types of materials [41]. There are very important properties in aesthetics, including color and surface roughness [42]. The composite structure and characteristics of the inorganic fillers have a direct impact on composite resin surface smoothness [43] and susceptibility to extrinsic staining [44].

There are some factors that are responsible for the color change of composites, which include; (1) intrinsic discoloration, which involves changes in the chemical stability of the resin matrix and matrix/particles interface, and the other is (2) extrinsic staining of the material. Extrinsic factors are related to absorption of staining solutions from exogenous sources that are associated to hygiene habits, smoking, and food [45].

In some studies it has been shown that composite resins are susceptible to color alteration when exposed to staining solutions, especially red wine, tea, whiskey, coffee, and cola [46, 47], and also depend on both water absorption of the composite and its hydrophilic resin matrix, which permit the permeation of staining agents, thus resulting in greater color changes [46].

At present, almost all people using mouth rinse solutions for antimicrobial control reduce halitosis and experience a cooling sensation [48]. This solution contains many components, such as detergents, emulsifiers, organic acids, dyes, and alcohol. Composite resins exposed to ethanol [49] are able to reduce bonding between the resin matrix and inorganic fillers, which might decrease erosion resistance and cause staining of the resin matrix. According to Sarrett et al. [50], alcohol acts as a plasticizer of the polymeric matrix, making the material more ductile [49, 50].

6.6 Repair techniques

Dentists used dental amalgams for more than a century, but due to several drawbacks, such as continuous corrosion, they are not able to fulfill the demand for cosmetic purposes, and also they do not adhere to the structure of the tooth properly. As there is advancement in the composite resin-based technology, the restorative treatment for amalgam restoration is usually the primary choice for various dental practices [51–54]. It gained recognition because of relatively low costs, good clinical performance, and cost effectiveness [51]. The fracture and secondary caries are widespread, and linked to the restoration of the amalgam, and identified with amalgam rebuilding efforts, and are the principle behind the substitution of deficient amalgam reclamations. Different reasons for substitution include deficient peripheral trustworthiness and insufficient interproximal contact [53–56]. Total rebuilding substitution is the most well-known treatment for imperfect amalgam reclamation [53, 55, 56]. However, this approach repudiates the present pattern toward negligible interventional systems to diminish the risk of pulpal wounds and spare tooth structures [52].

Repair is preferably substituted for the cure of restoration of the amalgam, with limited imperfections. It includes the evacuation of part of the rebuilding and any damaged tissue adjoining and subjacent to the imperfect area, and the remodeling of the primed site [57]. This method takes into account the safeguarding of sound tooth structure [58]. The minor repair of amalgam remodeling efforts plays an imperative role in clinical practice [53, 59, 60]. The utilization of amalgam holding operators has turned out to be noteworthy in the reclamation of posterior teeth, and exhibits numerous potential points of interest, including tooth fortification, improved peripheral adjustment, reduced postoperative sensitivity, reduced microleakage, decrease dental caries, and a more moderate preparation [61–65].

To date, there have been significant advancements in therapeutic materials, especially in tooth color materials. For the development of dental composites, nanotechnology plays an important role in the production of dental nanocomposites [66], endodontic sealers, glass-ionomer bonds, and tooth recovery.

Previously, the repair technique was dependent upon lab methods that exhibited impressive interfacial bond strength among “new” and “old” composite tar materials [67–70]. The *in vivo* repair technique, which involves long-term studies, has proved to be viable for total restoration replacement and is also a nondestructive technique [28, 71–73]. The method of reasoning for repair strategies is fortified by the confirmation that auxiliary caries are typically another sort of caries adjoining a rebuilding, as opposed to caries created in the tooth: the reclamation interface. The usage of suitable repair and renovation strategies builds the longevity of the influenced restoration, while constraining the degree of any agent mediation. Along these lines, the use of repair procedures builds the possibility of a reestablished tooth being one of the patient’s teeth forever.

6.7 Analysis of degradation products

The resin polymer and filler glass particles of dental composites have an elegantly look and hold fast to dentin and dental enamel [74–76]. The resin is generally made out of methacrylate polymers, for example, bisphenol A, diglycidyl methacrylate (BisGMA), and tetra ethylene glycol dimethacrylate. The coupling substances bind the filler particles and the polymer resin covalently, to confer better mechanical properties to the composite [55, 74, 77]. 3-(Trimethoxysilyl) propyl methacrylate (MPS) is a typical coupling machinist used to fasten the filler molecule to the methacrylate end of BisGMA [77, 78]. Both physical and synthetic procedures influence the dental composites’ degradation in the oral cavity [74, 79, 80]. Synthetic debasement of dental composites can be caused by hydrolysis, and additionally compounded by catalysis from spit and proteins in the oral cavity. The dental composites are degraded chemically by the process of hydrolysis or by the enzymes present in oral cavity or enzyme catalysis from saliva.

The degraded composite materials are accumulated in the body on their release, which could be of medical concern. Dental specialists have studied, and have concluded, that bisphenol A may have a significant risk to human health [81–84]. In dental materials, bisphenol A is not used directly [79], yet BisGMA and different parts of dental

composites are derivative of bisphenol A chemically, and are converted into related species with conceivably suspect wellbeing on its degradation. In the oral cavity, siloxane bond hydrolysis accelerates the dental composite degradation [85–88]. In this pathway of degradation, ester bond hydrolysis also takes place. The studies for most of the resins are eluted in a water medium to mimic the same environment as the oral cavity [89]. The composite material diffuses with water and is collected on reacting with the silane coupler on the surface between the filler and the resin, and the product produced due to its degradation gets released into solution [85, 90]. The diffusion of water may also produce the resin monomers, which are unreacted into the solution [52, 53, 74, 91].

6.8 Potential toxicity of degradation material

The conception of the biocompatibility of dental composite materials has been significantly enhanced, based on the results of in vitro tests utilizing a various assortment of test parameters. Acute toxicity influences cell culture studies related to mammalian cells, as proven by various types of preparation of samples, including sample aging, and conditions for curing and extract preparations [92–94]. The resinous samples that were noncured were much more cytotoxic compared with cured ones, and the impact of test samples on aging varied with respect to cytotoxicity [95].

The toxicity may be due to the residual monomers being produced, due to partial material polymerization or degradation from the dental composites [94]. Nearly 30 or more various compounds are being processed and extracted from polymerized dental composites, and the detected reaction products were found to be monomers, co-monomers, and additives [96, 97]. It has been demonstrated that through the elimination of leachable parts from polymerized composites by the utilization of natural solvents, the cytotoxicity was decreased completely [98]. The cytotoxic compounds include monomers, co-monomers, and other substances [99].

6.9 Strategies for the development of improved materials

Physicist Richard Feynman first developed the concept of nanotechnology in 1959, and introduced it in a paper titled “There’s Plenty of Room at the Bottom.” Dr. Nori Taniguchi, a Japanese scientist coined the term “nanotechnology” in 1974, and he defined nanotechnology as “The processing of separation, consolidation, and deformation of materials by one atom or one molecule.” He represented this concept in the meeting of the American Physical Society at the California Institute of Technology in 1959, where he described all the novel processes to manipulate individual components at the atomic/molecular levels, but he did not use the term “nanotechnology” or “nanosciences.” Dr. Drexler published a book titled “Engines of Creation – The Coming Era of Nanotechnology” in 1980, and expanded the concept of nanotechnology. In 1991, Sumio Iijima represented the idea of nanotubes by her publication “Helical Microtubules of Graphitic Carbon” [100]. Dental materials must restore the

dental structure and function of the affected dental surfaces due to caries or trauma; however, the desired effect is not usually accomplished. No synthetic materials are effective enough on external stimuli [101]. To overcome this problem, nanotechnology techniques have been applied in dentistry to improve the quality of dental biomaterials. Dental materials developed within the field of nanosciences offer improved antimicrobial properties and better safer for the oral cavity. In the field of dentistry, nanotechnology has covered a broad range of applications. Physical, chemical, and biological properties of materials have become different at the nano-scale. There are many ways to produce effective biomaterials that act as nature-made tissues, such as:

1. Preparing a synthetic material that resembles with the nature-made tissues in physiology and properties.
2. Producing biomaterials that have similar properties as those replaced by lost dental tissues.
3. Replacing lost dental tissues following regeneration principles and applying regenerative medicine and tissue engineering.

But all these approaches are not successful without the involvement of nanosciences [100]. Nanocomposites are one of the most widely used restoring dental biomaterials used for dental cavities [102]. Good translucency, countering, and surface finish are the beneficial qualities of nanocomposites [100]; but their use can create shrinkage stress and limited toughness. Nanocomposites are divided into two types: nanofills and nanohybrids. Nanofills include particles ranging from 1 to 100 nm size and nanohybrids include particles ranging from 0.4 to 5 μm in size [103].

In the 1970s, Wilson and Kent introduced glass ionomers as dental filling materials. The major function of glass ionomers is to retain the restorations on prepared teeth for a long time. Compomers, hybrid ionomers, zinc phosphate, zinc polycarboxylates, or resin-based cement are also used as strong cement [100]. Nanoparticles such as zinc oxide, silver, calcium phosphate, calcium fluoride, quaternary ammonium polyethylenimine and nanohydroxy apatite and nanofluoro hydroxyl apatite are mainly used in dentistry. The main advantage of these materials is that in smaller sizes, they can easily penetrate cell membranes and can affect host cells, which results in higher antimicrobial activity [102]. Silver nanoparticles (NAg) show bactericidal activity by reacting with the peptidoglycan cell wall and the plasma membrane, and also prevent DNA replication in bacteria. It shows a broad spectrum of bactericidal activity against the *Streptococcus mutans* and *Lactobacillus* species. NAg used in the range of concentration 0.5%–1% shows antimicrobial activity. Zinc oxide nanoparticles (NZn) are similar to NAg in their antimicrobial activity, and show a broad spectrum of antibiotic activity against several types of bacterial spp., including *S. mutans*. Zinc oxide nanoparticles (NZn) are more selective for bacterial cells, so it's a safer drug for human cells. Quaternary ammonium polyethylenimine nanoparticles (QAS) show antibiotic activity due to its positive charge and lipophilic nature [101, 102].

6.10 Future prospects

Today, there is a high demand for dental biomaterials, but there are no dental materials with ideal properties for any dental applications. Advances in nanotechnology

offer hope that nanomaterials will be helpful in developing new materials or providing significant improvements in the properties of existing dental materials. The potential use of nanomaterials in dentistry offers promise for the future, and will be helpful in boosting the quality of life of dental patients. It can be expected that in the future, the science of dental materials may change significantly with better understanding and the introduction of new nano-biomaterials. For example, nanorobots or dentirobots will fight against bacteria and quay them within the oral flora, and the diagnosis and treatment time period will be reduced from months to days, or even days to hours, thanks to scientific research in nano-dentistry. This is an area of very active research all around the globe that currently receives a great deal of research funding.

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