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Biodegradation of resin composites and adhesives by oral bacteria and saliva: A rationale for new material designs that consider the clinical environment and treatment challenges

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ABSTRACT

Objective. To survey the recent literature from the late 1980s to recent years in order to assess the relationship between resin degradation, catalyzed by biological factors, and clinical failure outcomes such as marginal breakdown.

Methods. The literature shows that degradation occurs in many manufacturers' products despite varied vinyl acrylate compositions. The authors examine salivary enzyme activity and their ability to degrade the polymeric matrix of resin composites and adhesives, as well as oral microorganisms that can promote demineralization of the tooth surface at the marginal interface. A survey of recent research relating matrix metalloproteinase (MMPs) to the degradation of the exposed collagen at the dentin adhesive interface is also discussed in the context of marginal breakdown.

Results. The literature provides strong support that together, the above factors can breakdown the marginal interface and limit the longevity of resin composite restorations. The authors have found that the field's current understanding of resin biodegradation in the oral cavity is just beginning to grasp the role of bacteria and enzymes in the failure of resin-based restorations.

Significance. Knowledge of these biodegradation processes is pertinent to areas where innovative strategies in the chemistry of restorative materials are anticipated to enhance the longevity of resin composites.

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1. Introduction

Resin composite is the most widely used dental restorative material in practice today due to its superior esthetics and

ease of handling [1]. Despite the extensive use of polymeric composites that contain vinyl resins, the resultant restorations lack the durability of amalgam fillings in terms of both inherent mechanical properties and inherent chemical stability, thereby limiting their lifetime *in vivo* [2–4]. The shorter

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longevity has been attributed in-part to material degradation, compromised adhesion by clinical factors, fracture, polymerization shrinkage, and secondary caries [1,2]. Secondary caries is the recurrence of caries at the tooth–restoration interface [5]. Secondary caries is often cited as the most frequent reason for restoration replacement, accounting for approximately 50% of the reported replacements regardless of the restorative material [6–17]. The latter studies reporting on over 18,229 restorations worldwide and spanning 20 years, provide data explaining the reasons for their replacement, ranking secondary caries as the predominant cause [18]. Upon the degradation of margin interfaces, acid-producing bacteria such as *Streptococcus mutans* can infiltrate the margins and contribute to the progression of recurrent caries [5]. It is believed that the accumulation of such microorganisms in the marginal gap can promote the demineralization of the tooth interface, thereby contributing to the progression of caries [5,18].

Over the years, many groups have studied the degradation of resin composites in the oral cavity. In the earlier years of this research most of the work focused on material loss as a result of wear and mechanical function [19–21]. However, beginning in the early 1990s, the focus of the studies shifted toward the chemical breakdown of these restorative materials because it was suggested that enzymes in the oral cavity may contribute to the degradation of resin composites [22,23]. Since then, a number of studies have investigated the degradation of resin composites in the presence of salivary-like enzymes [24–27], and the subsequent biological effects of the by-products on the surrounding bacteria and mammalian cells [1,28–33]. These biological processes that render commercial resins and adhesives vulnerable to premature failure are currently beyond the control of the clinicians.

The article will first report on the inherent vulnerability of modern day restorative resin chemistry with respect to degradation and then survey the literature of the biological factors present in the oral cavity, which challenge this chemistry. Lastly, strategies that are being pursued to address these processes will be reported on.

2. Dominant factors influencing bio-degradation

2.1. Chemistry of resin composites

A composite by definition is formulated from two or more components with inter-atomic interactions, producing a product that has superior properties to those of the individual components alone [34]. Dental resin composites are composed of four major components: a polymeric matrix that is usually methacrylate based (several examples of commonly used monomers are illustrated in Fig. 1), filler particles (commonly glass, quartz, or ceramic oxides), coupling agents between the filler and the matrix such as silanes, and an initiator/inhibitor polymerization system. The major constituent by weight and volume are the filler particles, usually inorganic, which provide the composite with improved mechanical properties such as compressive strength and modulus of elasticity, and reduced polymerization shrinkage, water absorption, and thermal expansion coefficient [2].

A very commonly used monomer for the polymeric matrix is 2,2-bis [4(2-hydroxy-3-methacryloxypropoxy)phenyl]propane, also known as bisphenol A glycidyl methacrylate (BisGMA), which was first introduced in late 50's by Bowen [36]. This hybrid molecule was initially synthesized by the reaction of glycidyl methacrylate and bisphenol A [36]; however, it was later produced by the coupling of methacrylic acid and diglycidyl ether of bisphenol A via an ester linkage [37]. In comparison to methyl methacrylate that was originally used in the early 1930s, BisGMA has superior mechanical properties, undergoes less polymerization shrinkage, and hardens more rapidly under oral conditions [36]. BisGMA contains hydrophobic aromatic rings in the backbone that provide the resin with low chain mobility and less deformation upon mechanical loading relative to linear non-aromatic monomers. The pendent hydroxyl groups in the alkyl chain further enhance the mechanical properties by participating in hydrogen bonding with the carbonyl groups on the methacrylate moiety. As a result of the aforementioned hydrogen bonding and the pi-pi interactions between aromatic rings of BisGMA molecules, this high molecular weight monomer has a viscosity that is 10^5 – 10^6 orders of magnitude greater than water at room temperature [38,39]. The high viscosity prevents the addition of high amounts of filler and reduces the degree of conversion (polymerization) when this monomer is used on its own. Therefore, diluent monomers are used in conjunction with BisGMA to enhance resin mobility for ease of handling and operation [40]. Among diluent monomers, triethylene glycol dimethacrylate (TEGDMA) is the most extensively used in current resin restorations. TEGDMA is a low molecular weight di-vinyl monomer that enhances the efficiency of polymerization by reducing the overall viscosity. This allows for better mixing and blending of the different constituents within resin composites. However, there is a limit to the amount of diluent that can be added to resin composites, as it greatly increases water sorption due to the triethylene oxide spacers in TEGDMA [41]. Furthermore, with increasing TEGDMA content, the overall resin composite experiences a greater volumetric shrinkage upon polymerization [41], which results in the potential for a greater marginal gap upon curing.

The majority of dental composites undergo solidification via free radical chain polymerization of di-vinyl oligomers, which enables the formation of a cross-linked network. The reaction is commonly initiated by predominantly photochemical means, but also by chemical means [42]. As monomers polymerize, chain motion becomes increasingly restricted, and ultimately the free volume occupied by the molecules decreases. The increase in cross-linking density increases the hardness and stiffness of the composite, therefore enhancing the overall modulus and strength. However, with further cross-linking and the additive effect of van der Waals interactions, the resin material experiences a volumetric contraction upon polymerization [2]. It is believed that the internal stresses generated by this process can ultimately lead to adhesive or cohesive failure at the tooth/resin interface [43]. Volumetric contraction can further result in tooth distortion and/or gap formation between the restoration and tooth structure, which allows for microleakage of salivary fluids,

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