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Degradation of dental resin composites during intra-oral wear

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Summary

Summary

Background

Dental resin composites are used worldwide to replace missing tooth structures, to enhance aesthetics and restore function. The oral cavity is a complex, hostile and aggressive environment. When placed in the oral cavity over a longer period of time, resin composites undergo changes in shape, dimensions and color as a result of functional wear, secondary caries due to inadequate oral hygiene and material degradation.

Over the years many changes with respect to the chemical composition of composite resins have been made and proposed to improve handling characteristics and durability. An in depth and detailed understanding of the etiology of (f)ailing restorations is of great importance, both from a dental health and from an economical perspective. It is a prerequisite for further improvements.

Degradation during intraoral wear is the resulting effect of several chemical, mechanical and physical factors, with time as an overruling factor for all. How they work together is not well understood. *In vitro* aging models employed for the evaluation of resin composites merely study single factors, and therewith lack the synergy of factors operative in the oral cavity. Moreover, little is known about the complex interplay of different factors during wear and necessary oral hygiene procedures, such as tooth brushing, affects the properties of dental resin composites.

Therefore, the aim of this PhD study was to investigate the effect of oral factors on the degradation of a selection of direct and indirect dental resin composites by observing parameters related to its color stability, the biofilm formation, and physicochemical deterioration in an *in vivo* study model.

Finally, and based on the results, some light is shed on the multifactorial etiology of degradation of dental resin composites. Recommendations are made that may aid others when designing future degradation studies and clinical considerations with respect to the selection and maintenance of resin composites are proposed in order to achieve longer lasting performance to the benefit of patients.

SUMMARY

The clinical study, results, conclusions and recommendations

The backbone of this PhD project is formed by a clinical study in which 16 volunteers wore removable acrylic palatal appliances with integrated disks of direct (Beautifil II, Filtek Z350 XT) and indirect resin composites (Lava Ultimate CAD/CAM, Estenia C&B,) during periods of 30 days. Three regimes of manual brushing were applied during this period: brushing with a fluoridated toothpaste, brushing with water and no brushing in order to evaluate the cumulative effect of exposure to the human oral cavity, including wear and brushing on various parameters.

The acrylic appliances were supplied with a thermosensitive chip (TheraMon®) to monitor actual wearing time. This resulted in the exclusion of one volunteer because of lack of compliance. Hence a complete data set for 15 volunteers was considered valid and available for the different analyses. Depending on the purpose and focus of the study, a selection of direct and indirect resin composites was included in **Chapters 2**, **3** and **4**.

The rationale for the total PhD project and specific aims are laid out in **Chapter 1** and based on the observation in **Chapters 2**, **3** and **4**, specific recommendation for several parties (i.e. manufacturers, researchers and clinicians) are brought into context in the **General discussion** chapter.

Chapter 2 deals with the color stability of three resin composites (Beautifil II, Filtek Z350 XT and Estenia C&B) which was related to the bulk filler content, surface filler exposure and chemical surface composition. Color values (ΔE) were evaluated according to the *L, a, b*, color coordinate system (International Commission on Illumination).

Significant discoloration occurred in all composites and was caused by a slightly darker, more reddish and yellowish appearance of the composite, that was in general not impacted by the absence or presence of brushing. The discoloration ΔE related with bulk filler content, but not with filler particle exposure at the surface and was always accompanied by a doubling or tripling of the %N exposed at the surface of the composite. Major increases in %N observed after intra-oral wear yield the conclusion that amines (i.e. photo-co-initiator) diffuse from the bulk to the composite surface to attract discolorants.

Hence, it was concluded that in the absence of significant and systematic effects of brushing or no brushing, the causative discolorant is presumably the photoinitiator camphorquinone, known to produce a yellowish hue. This suggests that color stability of dental resin composites might be improved by changing the two-component photo-initiator/co-initiator system.

The focus of **Chapter 3** lies on the biofilm composition related to composite degradation during intra-oral wear. As already mentioned degradation of resin composite restorations during intra-oral wear limits their functional and aesthetic life-time and is attributed to the aggressive oral environment, including biofilm formation on composite restorations. Since biofilm forms on all surfaces exposed to the oral cavity, while biofilm inhabitants may produce different amounts and types of degradative enzymes, we hypothesize that composite degradation relates with biofilm composition. This hypothesis is tested in **Chapter 3** using samples of distinctly different resin composites (Beautifil II and Lava Ultimate CAD/CAM). Beautifil II is a nano-hybrid Bis-GMA/TEGDMA composite with glass ionomer particles (83 wt%), while Lava Ultimate CAD/CAM Restorative is a CAD/CAM milled, nano-ceramic Bis-GMA, UDMA, TEGDMA, PEGDMA, bis-EMA resin (80 wt% filled). They differ in their amount of ester-linkages, as determined using X-ray Photoelectron Spectroscopy (XPS).

PCR-Denaturing Gradient Gel Electrophoresis patterns of biofilms on both composites after intra-oral wear showed a higher prevalence of *Streptococcus mutans* on the high ester-linkage composite than on the low ester-linkage composite, especially in the absence of brushing. Composite degradation after wear was expressed as the percentage surface exposure of filler particles, calculated from measured advancing, receding and equilibrium water contact angles using the Cassie and Baxter equation. Degradation after intra-oral wear was significantly stronger on the high ester-linkage than on the low ester-linkage composite, especially in absence of brushing, while no significant increases in filler exposure of the low ester-linkage composite, indicative of degradation, were observed.

These results indicate that bacterial esterases in biofilms containing *S. mutans* are demonstrably responsible for the *in vivo* degradation of composites containing a high number of ester-linkages. Since the development of a composite-degradative microbiome can be slowed down by brushing, these results emphasize the need for proper oral hygiene of composite restorations and point out a developmental pathway for composites less amenable to degradation during intra-oral wear.

SUMMARY

Identification of sensitive methods to detect early degradation of composite resins, either after *in vitro* experiments or intra-oral wear is important as the use of more sensitive methods will allow shorter experimentation times to reach statistically significant and relevant conclusions. The aim of the study described in **Chapter 4** was to compare the results of the different conventional methods and more surface-sensitive techniques as applied in the previous chapters to investigate the early degradation of dental resin composites.

Data from all four resin composites (Beautifil II, Filtek Z350 XT, Lava Ultimate CAD/CAM and Estenia C&B) were included. Early composite degradation was evaluated using two conventional methods (hardness and surface roughness measurements) and two less conventional surface-sensitive techniques to calculate filler exposure (measurement of equilibrium, advancing and receding contact angles and XPS analyses).

The dynamic range over which the results of the different methods and techniques ranged was largest for the surface roughness measurements, followed by contact angle measurements and XPS analyses. Moreover, filler exposure calculated from measured contact angles correlated with surface roughness data.

This led to the conclusion that early degradation is best studied by surface roughness measurements or analysis of equilibrium, advancing and receding water contact angles as they most sensitively reflect changes at a surface and their results for composite degradation confirm each other. XPS is also suitable for surface analysis of early composite degradation, but its results partially reflect the bulk matrix making it slightly less suitable. Hardness, though suitable to provide information on bulk matrix degradation, is unsuitable for measuring early composite degradation that is still confined to the surface.

In the **General discussion**, an overview regarding the state of the art in dental resin composites is provided. The introduction of resin composites to dentistry and the pathway to improvement are discussed. A better understanding of pathways that are relevant to the process of (resistance to) degradation of dental materials and their interaction with the oral environment may build on current knowledge and lead to improvements on one of more qualities of presently used dental materials.

The ideal restorative material should have excellent optical qualities, should have physical properties matching those of the tooth structure that it is replacing, should be as wear resistant as the enamel surface of its sound

antagonist or as the restorative material opposing it and be able to resist fatigue. It must be possible to achieve and maintain an excellent bond between the composite material and enamel and especially dentin, be tasteless, repairable and biocompatible, hence not be cytotoxic. In addition, the material should be easy to handle and polishable, cheap and if it were antimicrobial as well, it would truly be the 'holy grail' of restorative dentistry. Clinical reality is that we are not there yet.

In the **General discussion**, the findings are brought into perspective. Schematic figures are provided that illustrate and summarize the present understanding from literature and our findings from the various studies that are presented in this thesis. They involve the tentatively presumed parameters in the process of degradation of dental resin composites.

It is clear that the material-factor dependency is an important key-indicator to predict the onset of composite degradation. Polyurethane based monomer with the highest nanofiller content (%weight) exhibited the steadiest performance on three assessed parameters, i.e. higher hardness and lower surface-roughness as shown in **Chapter 4** and better color stability as shown in **Chapter 2**.

The molecular basis for the color change phenomena is presumably related to the filler content, indicating that composites having the highest filler content will be more stable. This is the result of superior mechanical and chemical wear resistance properties. Our data underscore such presumption since it is in accordance with our surface roughness measurements. It indicates that the change of physical properties on the outermost-surface affects its color stability and roughness. Hence, the %filler determines the stability of these two surface-degradation parameters under intra-oral exposure.

As also demonstrated in other studies, bacteria in the oral cavity can potentially cause degradation of resin composite. We showed that oral degradative-microbiome rich in *S. mutans* was associated with the degradation of matrix resin composite (**Chapter 3**).

General recommendations for future material development include the development of an alternative polymeric monomer and resin composite formulations with enhanced biochemical stability and a less cariogenic microbiome adherence as this will enhance the resistance of composite resin materials to degradation in the oral environment.