Viscoelastic behavior of dental restorative composites during setting
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Citation for published version (APA):
GENERAL INTRODUCTION

George Washington, the first president of the United States of America, used to wear an artificial denture made of ivory and bovine teeth (Fig. 1.1). Too vain to appear in public without it, but unable to speak when wearing it, he resorted to work from home - the White House - rather than from his office in the Congress building. American presidents have made a tradition of this behavior since.

Fortunately, a basic change in the nature of dentistry has occurred since George Washington's time, most noticeably over the past few decades. Dental care has become available to the industrialized world and a growing portion of the population now retain all or part of their natural teeth well into old age. Thanks to regular check-ups, disease can be discovered and treated in an early stage. As a rule, only relatively small portions of a tooth have to be removed and replaced by fillings.

Figure 1.1 One of the six artificial dentures of George Washington (1732-1799). The elements used were made of bovine teeth, human teeth, and ivory in a lead base, with springs that allowed the first president of the USA to open and close his mouth. The artificial dentures, made by Dr. John Greenwood (1790), fitted poorly and distorted the shape of his mouth. Reprinted from Mount Vernon, Virginia, USA, with permission.)
The crown of a tooth consists of dentin covered with a layer of enamel about one-millimeter thick. Tooth enamel contains much more hydroxyapatite than dentin, which gives it a hardness comparable to that of semi-precious stones. Due to our diet and the presence of certain bacteria in saliva, a process called caries is initiated. During this process, the acidic products of the metabolism of the bacteria (plaque) soften the enamel and eventually the dentine by dissolving hydroxyapatite, resulting in the formation of cavities in the teeth.

![Figure 1.2 Effects of shrinkage stresses in restoration.](image)

It will be clear that the materials used for the restoration of cavities should be able to be brought in the appropriate anatomical shape, and that they should retain that shape throughout the life of the tooth. The use of metals such as gold alloys and amalgam has for many decades provided satisfactory results with respect to the preservation of tooth anatomy. Nowadays, conventional glass ionomer cements, compomers, and resin composites have gained a permanent position on the dental market as direct restorative material. Their superior esthetics and consecutive preparation requirements (less destructive than amalgam) have been instrumental in this commercial success.

The ideal restoration has a perfect seal with the remaining tooth structure, since otherwise bacteria and the toxins they produce can invade and populate in the gap formed, resulting in pulp irritation and even secondary caries (Fig. 1.2). This perfect seal must be obtained during setting (solidification process) and then maintained during thermal and mechanical cycling for the lifetime of the restoration or the patient. Unfortunately, the present generation of esthetic restorative
material does not yet guarantee a tight seal. Most of this shortcoming is related to bulk shrinkage of the restorative material during the setting process. Due to the adhesion to rigid tooth tissue, this shrinkage is constrained, and this, in combination with the increasing stiffness of the restorative material, inevitably leads to the development of mechanical stresses in and around the restoration.

These stresses are a major problem, since they have a negative influence on the durability of the restoration. While loss of adhesion can occur at any time, the most likely moment is when the magnitude of the shrinkage stress exceeds the strength of the developing restoration-tooth bond. Since bulk shrinkage takes place largely within 15 minutes of setting [1], adhesive failure starts early, occasionally even before the patient has left the dentist’s chair [2]. Although the restoration will probably not fall out of the preparation, it has to be replaced to prevent adverse biological reactions. Even if adhesion survives the mechanical stresses, there may be cusp movement, postoperative sensitivity, cohesive fracture, or tooth fracture.

Thus far the literature has given considerable attention to factors such as bulk shrinkage, restoration configuration, water absorption, porosity, and the effect of the kinetics of the setting mechanism on shrinkage stress [3]. The outcome of these studies has resulted in time-consuming restorative techniques for the practitioner (preparation design, special filling techniques, linings, variable light intensity, etc.) designed to obtain restorations with a tight seal and low ultimate internal stresses. And yet there is still no clear understanding of why the adhesive bond fails in some situations but not in others.

To gain more insight into the problem of shrinkage stresses, research has focused on the viscoelastic behavior of dental restorative materials during setting. This mechanical behavior during setting - when the material passes from a liquid to a solid state - is an important factor in the relation between shrinkage of the restorative material and stress development in the restoration. Once the viscous flow and solid property of the restorative material during setting can be quantified, then the research into shrinkage stress development can be enriched with numerical analyses and simulation techniques.

Finite Element Analysis (FEA) is widely used to calculate material stresses [4]. In a finite analysis, a complex structure - such as a tooth - is subdivided into a number of small, simply shaped elements, for which individual stresses can be more easily calculated than for the structure as a whole. By solving the stresses of all the small elements simultane-
ously, the total stress of the whole structure can be approximated. The first attempt to study the stress build-up caused by the shrinkage of the restorative composite by means of FEA appears promising [5]. However, for proper simulative shrinkage stress studies, a reliable viscoelastic model is required, one whose material parameters for the setting of the restorative material are known.

**Aim of this research project**

The aim of this research project was to use modeling to obtain more information on the viscoelastic behavior of resin composites during the setting process. Part of the research focused on finding a mechanical model capable of predicting the viscoelastic behavior of dental composites during setting. With a suitable model, the viscoelastic parameters viscosity ($\eta$) – an inverse related measure of viscous flow - and elastic modulus ($E$) – a measure of stiffness - can be quantified on the basis of experimental data. Although different classes of bulk restorative materials exist, this research project focused on two-paste and light-activated resin composites.

The other part of the research dealt with the use of mechanical models to study the effect of resin formulation, the initiator system, configuration (C-factor), and temperature on the mechanical behavior of composites during and after setting. For this purpose, conventional (dimethacrylate) and experimental (oxirane-based) composites were studied. The quantification of the viscoelastic parameters will lead to a better understanding of the relation between bulk shrinkage and stress development within the composite.

**Scope of this thesis**

This thesis represents the findings of the research project on modeling the viscoelastic behavior of dental composites during setting. It deals first with the improvements in the quality of stress-strain data obtained by the dynamic testing method. Next, the modeling of resin composites is described, starting with chemically activated (two-paste) composites. And finally, the effect of dimethacrylate composition in the resin and the use of low-shrinking oxiranes as resin, are examined. Characterization studies such as infrared spectroscopy, wear, and scanning electron microscopy, were performed to provide additional information pertinent to the discussion of the effect of resin formulation.
The results described in the present thesis are of importance for future numerical analysis. They can serve as input for FEA studies, which make it possible to simulate the stress distribution in and around the restored tooth. Practitioners will then have a better understanding of how and where stress develops in composite restorations, and can then refine their techniques to minimize the deleterious effects of shrinkage forces. Although this investigation focused on dental resin composites, the numerical investigation described here is also applicable to other materials used in dentistry and medicine.

Chapter 2 reviews the underlying causes of shrinkage in polymeric restorative materials, and the various factors that are of influence. Some factors affecting stress development are beyond the control of the clinician (e.g., the formation of the composite); however, the methods used for placement and light-curing are aspects which he can control directly. This review stresses the importance of knowing the relation between these manipulative factors and the development of shrinkage stresses. Special attention is given to the polymerization reaction, the composite structure, and its relation to viscoelasticity.

The choice of a mechanical model requires a thorough knowledge of the viscoelastic properties of the resin composites. Dynamic tests are necessary in order to determine the major characteristics of the composites. Moreover, these tests must provide reliable stress-strain data, which are important for the modeling of the viscoelastic behavior of dental composites during setting. Chapter 3 describes the experimental details of the present research, including the preparation of specimens, the dynamic test system, and test protocols. On the basis of the recorded stress-strain data, the limitations of the dynamic test system for shrinking dental restoratives are discussed.

Chapter 4 introduces the modeling of axial stress-strain data. It is intended for readers who are not familiar with mechanical models and have no experience in data modeling. A number of possible models for the description of the viscoelastic behavior of dental composites are presented, and a modeling procedure capable of calculating material parameters from a set of experimental data is described step-by-step. A schema of the validated procedure for parameter identification is given, and the influence of noise on the identification procedure is discussed.

 Chapters 5, 7, and 8 deal with the modeling of the viscoelastic behavior of two-paste dimethacrylate composites, light-activated dimethacrylate composites, and light-activated oxirane composites respectively. On the basis of the modeling results, a suitable mechanical model has been
selected, which can predict the viscoelastic behavior of the composites during setting.

The effect of bisGMA-TEGDMA on the mechanical properties of two-paste composites is described in Chapter 6. Special attention is given to the question of whether flowable composites undergo a prolonged viscous flow state, which may ultimately lead to less shrinkage stress in the material. The setting process of several experimental bisGMA-TEGDMA composites has been monitored and characterized by dynamic tests, dilatometry, infrared spectroscopy, and mathematical modeling. In addition, the tensile strength of the composites after one hour of setting, and the wear process over a period of one year were evaluated.

Chapter 9 describes a preliminary study focusing on the potential of a low-shrinkage composite for use in restorative dentistry. The shrinkage strain, stiffness development, and tensile strength at different configurations (C-factor) of an experimental oxirane composite have been measured and analyzed at room temperature and oral temperature.

Lastly, the summary describes what has been accomplished with this project and a number of conclusions are given. In addition, suggestions for future work related to the viscoelastic behavior and shrinkage stress relief of dental restoratives are suggested.

References

3. See chapter 2 of this thesis.