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ORIGINAL ARTICLE

Ultrasonic measurement of the effects of adhesive application and power density on the polymerization behavior of core build-up resins

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Abstract

Objective. To use ultrasonic measurements to monitor the effects of adhesive application and power density on the polymerization behavior of dual-cured core build-up resins. ***Methods.*** Ultrasonic measurements were carried out using a pulser–receiver, transducers and an oscilloscope. The core build-up resins were mixed, inserted into a transparent mold and then placed onto a sample stage with or without self-etch adhesive. Power densities of 0 (no light irradiation), 200 and 600 mW/cm^2 were used for curing. The transit time through the core build-up resin disk was divided by the specimen thickness to obtain the longitudinal sound velocity (V). ***Results.*** Light irradiation of the core build-up resins at a power density of 600 mW/cm^2 caused V values to rise to an initial plateau of 1550–1650 m/s, then to rise rapidly to a second plateau of 2800–3200 m/s. The rate of V increase was slower when the resin cements were light-irradiated and became faster when irradiated at a higher power density. There were no significant differences between the groups with or without adhesive. ***Conclusions.*** The polymerization behavior of the core build-up resins was affected by the power density of the curing unit. The influence of adhesive application differed among the core build-up resins tested.

Key Words: *adhesive, core build-up resin, polymerization, ultrasonic sonic velocity*

Introduction

Dual-cured resins, in which polymerization can be initiated by both chemical and light activation, are used in some clinical applications. Such materials should have an extended working life and be capable of reaching a high conversion ratio in areas with lower light penetration. However, dual-cured resins reportedly show unsatisfactory mechanical properties when the polymerization reaction is limited to the chemical-cure mode alone [1–3]. Light irradiation appears to be necessary for dual-cured resins to achieve optimal adhesion to tooth structures. The bond strength of dual-cured resins to dentin is expected to be influenced by the power density of the curing unit, which decreases as the depth of the cavity and/or the thickness of the restoration increases, because their polymerization behavior is dependent upon the energy of irradiated light [4]. Indeed, polymerization is mainly initiated by light activation in the superficial layers of the materials and by chemical activation in the deeper layers where there is insufficient light energy.

Some of the core build-up resins utilize self-etching adhesives for bonding to tooth structure. These adhesives form a continuous layer between the core build-up resin and the tooth surface, which undergoes demineralization by acidic functional monomers followed by resin-monomer penetration into the tooth substrate. However, self-etch adhesives and dual-cured composites have been reported to be incompatible [5]. Acidic resin monomers inhibit the polymerization reactions of dual-cured composites that are initiated via peroxide-amine binary redox catalysts. The adhesive resins are cured by a free-radical polymerization reaction that utilizes camphorquinone (CQ) as a visible-light-activated photoinitiator [6,7]. CQ requires a co-initiator in order for effective polymerization to occur and uses a tertiary amine photoreductant for this purpose. The tertiary amine interacts with activated CQ to produce reactive radicals for polymerization [8,9]. Tertiary amines in

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the core build-up resins can be neutralized by the acidic functional monomers in the self-etching adhesives, resulting in weak polymerization of adjacent core build-up resins [10]. Although several studies have investigated the utilization of core build-up resins, little is known about their polymerization behavior when combined with self-etching adhesives [11].

Ultrasonic imaging is a non-invasive technique that shows considerable diagnostic potential as well as being a valuable research tool. Ultrasonic devices can be used to detect carious lesions [12] and to measure the dentin thickness between the tooth surface and the pulp chamber [13]. As the longitudinal sound velocity (*V*) is sensitive to the viscoelastic properties of materials [14], ultrasonic devices can be used to monitor the setting processes of dual-cured resins and ceramics [15–17]. The *V* value is closely related to the elastic modulus of a material, such that the elastic coefficient changes as the *V* value changes. During the curing process, core build-up resins change from a viscous liquid state to a glassy solid state via the polymerization reaction. This process might result in measurable changes in the *V* value of core build-up resins, as reported in other dual-cured materials.

Here, we used an ultrasonic device to monitor the polymerization behavior of core build-up resins and investigated the effects of the application of self-etching adhesives and the power density of the curing unit. Our null hypothesis was that the polymerization processes of the core build-up resins were not affected by the application of self-etching adhesives or by the power density of the visible-light-curing unit.

Materials and methods

The following dual-cured resin core build-up systems were employed in combination with the adhesives recommended by the manufacturers, as shown in Table I: Clearfil DC Core Automix (DC; Kuraray Medical Inc., Tokyo, Japan); and UniFil Core (UF; GC Corp., Tokyo, Japan). A visible-light-curing unit (Optilux 400; Demetron/Kerr, Danbury, CT) was connected to a variable-voltage transformer (Type S-130-10; Yamabishi Electric Co., Tokyo, Japan) in order to adjust the power density to 200 or 600 mW/cm^2; these values were determined using a dental curing radiometer (Model 100; Demetron/Kerr). The curing unit was positioned on the jig such that the distance between the light-tip end and the specimen surface remained unchanged (1 mm) during the experiments. All experiments were done in a temperature-controlled room under ambient conditions and wrote as suggested.

The ultrasonic equipment employed in this study comprised a pulser–receiver (5900PR; Panametrics, Waltham, MA), two different types of transducer (M203 and V112; Panametrics) and an oscilloscope (WaveRunnner LT584; LeCroy, Tokyo, Japan) [18,19]. The pulser–receiver, with a 200-MHz band width, was used to drive the transducer for transmitting and receiving ultrasonic signals at a pulse-repetition frequency of 500 Hz and an excitation energy of 16 µJ. This entailed sending a 16-µJ impulse to the transducer, which converted the electrical energy into a sound wave and propagated it through the specimen. The right-angle-type contact transducer had a tip contact diameter of 3.0 mm, a nominal center frequency of 10 MHz and a removable Plexiglas delay tip. Two transducers were orientated facing each other and the sample was placed between them. The transducers were connected to the pulser–receiver, operated in a through-transmission mode, and signals were obtained using the oscilloscope. No heat was generated by the transducers during measurements made at the pulse-repetition frequency employed in this study. The transducer equipment was calibrated at each use by employing a standard calibration procedure on calibration blocks.

The core build-up resins were mixed according to the instructions of the respective manufacturers and were inserted into a transparent mold (2-mm thickness) made of acrylic resin. Each specimen was then placed onto the sample stage, with or without a

Table I. Core build-up systems tested.

Core build-up resin (Lot No.)	Adhesive system (Lot No.)	Manufacturer (Code)
Clearfil DC Core Automix (000320A) Bis-GMA, TEGDMA, dimethacrylate, filler, photo/chemical initiator	Clearfil DC Bond A (00012A) HEMA, Bis-GMA, MDP, filler, photo/chemical initiator Clearfil DC Bond B (00008D) water, ethanol, chemical initiator	Kuraray Medical Inc., Tokyo, Japan (DC)
UniFil Core (0710241) UDMA, dimethacrylate, fluoroaluminosilicate glass, photo/chemical initiator	Self-Etching Bond A (0710241) 4-MET, ethanol, water, methacrylate esters Self-Etching Bond B (0711011) ethanol, initiators	GC Corp., Tokyo, Japan (UF)

HEMA, 2-hydroxyethyl methacrylate; Bis-GMA, 2,2bis[4-(2-hydroxy-3-methacryloyloxypropoxy)phenyl]propane; MDP, 10-methacryloxydecyl di-hydrogen phosphate; 4-MET, 4-methacryloyloxyethyl trimellitate; CQ, *dl*-camphorquinone; TEGDMA, triethylene glycol dimethacrylate; UDMA, urethane dimethacrylate; BPO, benzoyl peroxide.

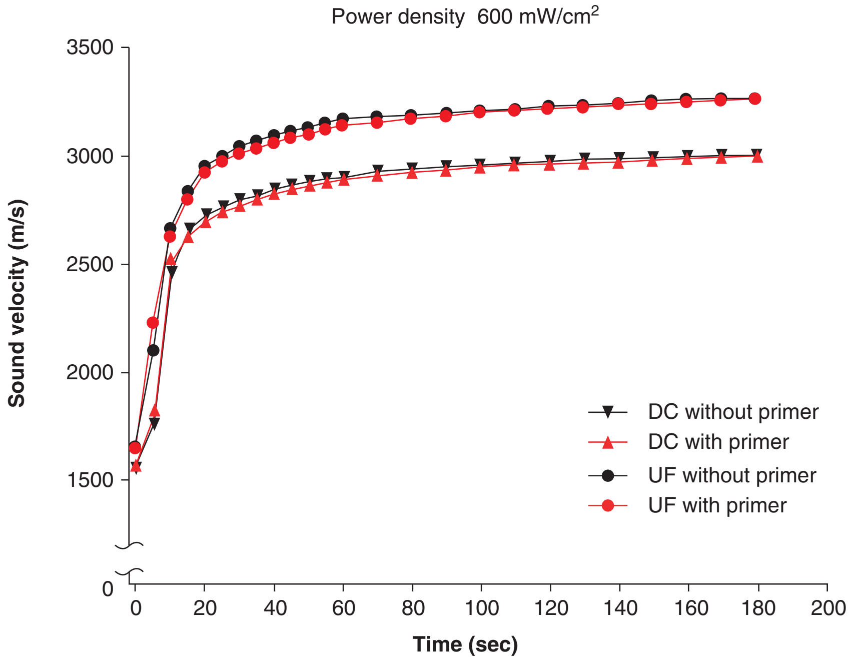

Figure 1. Changes in V values in core build-up resins as a function of time when irradiated with 600 mW/cm^2 power density. As the core build-up resins hardened, *V* values increased until they reached a plateau.

self-etch adhesive, and subjected to air blowing. Light irradiation was carried out from the lateral side of the specimen through the transparent mold for 30 s at power densities of 0 (no irradiation), 200 and 600 mW/cm^2. *V* values were determined by measuring the time taken for an ultrasonic pulse to traverse each specimen. Two transducers mounted on a calliper (the sample stage) were pressed onto the specimen and the light irradiation and ultrasonic measurement were performed simultaneously. Ultrasonic longitudinal waves of 10 MHz were propagated from the transducer to the specimen and the time taken for the pulse to travel from one transducer through the specimen to the other transducer was determined from the waveform. The *V* value within the specimen was calculated as L/t, where t was the traverse time of the ultrasonic pulse through the specimen and L was the measured path length (the specimen thickness). Six specimens were tested for each irradiation condition.

For the purposes of statistical analysis, each type of core build-up resin was treated as an independent experiment, in order to compare the results obtained under different experimental conditions. The mean and standard deviation for each type of core build-up resin were subjected to a one-way analysis of variance (ANOVA) for the six groups for each type of material. Multiple comparisons were then conducted using Tukey's Honestly Significant Difference (HSD) test at a significance level of 0.05. All statistical analyses were performed using the Sigma Stat Version 3.1 software package (SPSS Inc., Chicago, IL).

Results

V value changes in the core build-up resins were plotted as a function of time, starting 1 min after the paste-mixing commenced (Figures 1,2,3). The changes in *V* transmitted through the core build-up resins at the end of each measurement period (that is, at 180 s for power densities of 200 and 600 W/cm^2 and at 900 s for 0 W/cm^2) are summarized in Table II.

When the core build-up resins were light-irradiated with a power density of 600 mW/cm^2, *V* values increased to an initial plateau of 1600–1650 m/s then rapidly increased to a second plateau of 2800–3200 m/s, regardless of the application of adhesives. When the core build-up resins were light-irradiated with a power density of 200 mW/cm^2, the rates of increase of *V* were comparatively slower. Light irradiation caused no significant differences in the *V* values of the groups with or without adhesives for the core build-up resins tested in this study.

In the absence of light irradiation, the *V* values of the core build-up resins rose to an initial plateau of 1550–1650 m/s, then gradually rose over the next 200–400 s for UF and over the next 600–800 s for DC (Figure 3). Although sonic echoes could be detected from the start of the measurements, the rates of increase of sonic velocity were relatively slow. For DC, the *V* values at the end of the measurement period (at 900 s) were significantly greater with adhesive application than without. For UF, no significant difference was seen between the groups with or without adhesive application.

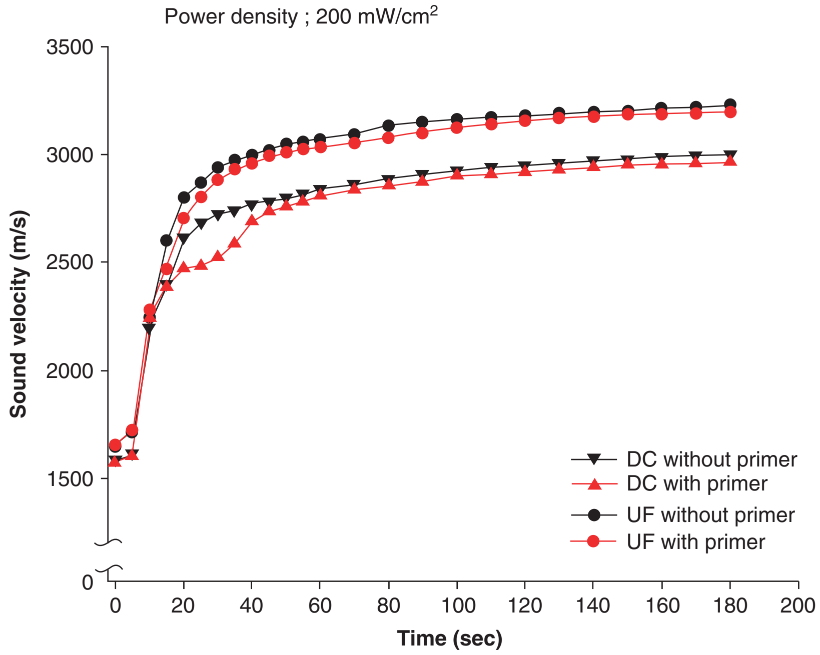

Figure 2. Changes in V values in core build-up resins as a function of time when irradiated with 200 mW/cm^2 power density. The *V* values increased, but were slower to reach a plateau than those irradiated with a power density of 600 mW/cm^2.

Discussion

Knowledge of the mechanical properties of core build-up resins is important for understanding their polymerization behavior. However, it is particularly difficult to quantify changes in the polymerization processes of core build-up resins at any given time point because they have more than one setting reaction. Here, we used non-destructive ultrasonic measurements to monitor the polymerization processes of core build-up resins, based on the relationship between the *V* value and the elastic constant described by a micromechanical model [20]. The ultrasonic approach does not define the degree of conversion as the time point at which the core build-up resins reach a specific strength; rather, it

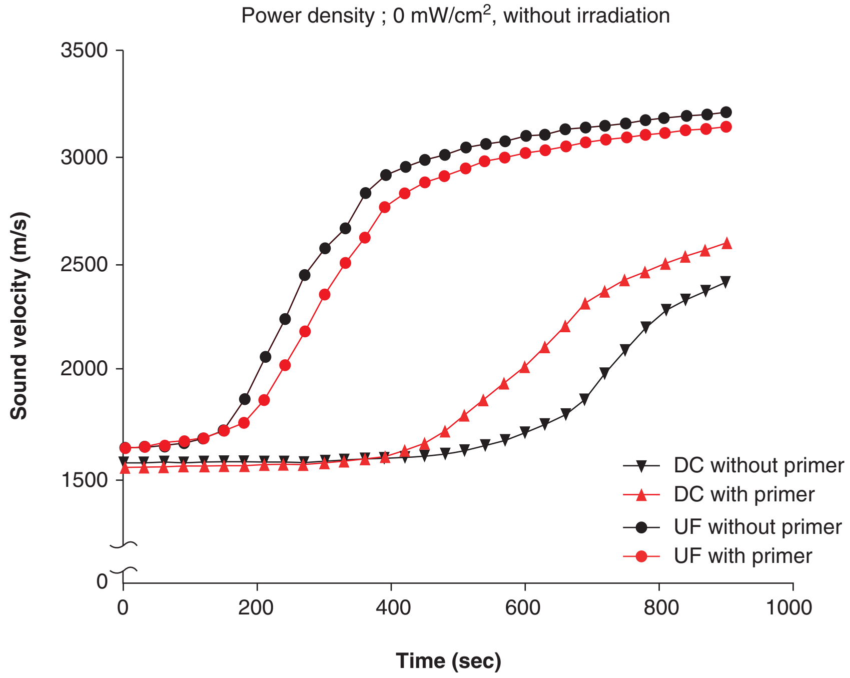

Figure 3. Changes in V values in core build-up resins as a function of time without irradiation. The rate of *V* value increase was slower than in the presence of light irradiation.

Table II. Changes in ultrasonic velocity transmitted through core build-up resins.

	Power density (W/cm^2)					
	0 Adhesive		200 Adhesive		600 Adhesive	
Material code	–	+	–	+	–	+
DC	2418 (56)	2601 (50)	2999 (32)*	2968 (50)*	3002 (63)*	2989 (57)*
UF	3212 (47)*	3143 (37)*	3230 (46)*	3200 (39)*	3260 (41)*	3255 (47)*

Values in parentheses indicate standard deviations.
For the same material, values with asterisks indicate no significant difference ($p > 0.05$).

monitors the viscoelastic properties of the materials [21].

The current study examined the effects of light intensities of 0 (no irradiation), 200 and 600 mW/cm^2. The intensity of the incident radiation in the appropriate wavelength range and the period of irradiation have crucial effects on polymerization processes and determine the development of the physical and mechanical properties of light-cured materials. Although some recommendations have been made for the minimally acceptable light intensity for a given irradiation time at a given material thickness, there is still no consensus on the length of exposure or energy density required to produce adequate polymerization. An exposure time of 60 s was previously reported to provide optimal polymerization when the light intensity was greater than 280 mW/cm^2, the composite increment was 2 mm or less and the light tip was held within 6 mm of the surface [22]. We therefore employed a light intensity of 200 mW/cm^2 as the lower level in the present study.

At the beginning of the ultrasonic measurement period, the *V* values in the core build-up resin mixes ranged from 1550–1650 m/s, which were similar to those for an ultrasonic pulse-transversing liquid [23]. During the initial stages of the pre-gel, the *V* value characteristics might be attributable to the attenuating properties of the core build-up resins. The length of the dough phase might be related to the power density of the curing unit, with a longer time period for a lower power density (200 mW/cm^2). Core build-up resins can undergo both a free-radical photo-polymerization process and a chemical polymerization reaction. When core build-up resins are not light-irradiated, chemical polymerization alone initiates cross-linking of the polymer chains. Upon light irradiation, photo-initiated polymerization occurs after an induction period that is associated with the consumption of polymerization inhibitor and dissolved oxygen by initiator-derived radicals. The conversion of methacrylate-functionalized dental restorative materials via photoinitiated polymerization is dependent upon several parameters and monomer formulation has been shown to have an impact on the conversion of resin-based composites [24].

When core build-up resins were irradiated with a power density of 600 mW/cm^2 in the present study, a rapid increase in the *V* value was observed after a short induction period. Light of the appropriate wavelength is absorbed as a visible-light-activated photoinitiator by a photosensitizer such as CQ [25]. The rate of polymerization is reduced at lower power densities, which results in longer induction times than at higher power densities. Even with the most reactive monomers, the fraction of reacted functional groups is significantly less than unity due to the highly cross-linked structure of the developing polymer. As monomer conversion is affected by the radiant intensity absorbed by the photoinitiator, the power density of the curing unit and its spectral distribution become critical variables [26]. In the current study, although there were no significant differences between the *V* values with different power densities (200 and 600 mW/cm^2), longer induction periods were detected for the 200 mW/cm^2-irradiated groups. In clinical situations, debonding might occur soon after the core build-up resin is applied if it is subjected to stresses caused by the restorative procedure, contraction shrinkage of the material or normal oral functions such as mastication. Sufficient mechanical strength shortly after placement is one of the determinant factors contributing to the clinical success of dental restorations [27].

When the core build-up resins were not light-irradiated, the *V* values remained similar after the start of mixing, then gradually increased after 200 s for UF and after 600 s for DC. This implied that the materials could not withstand the occlusal loading or even the preparation processes in the early stages of placement. When the self-etching adhesive was applied, no significant change was seen in any of the materials tested at the start of the measurement period. At the end of this period, significant increases in the *V* values were observed in DC samples with adhesive. Adhesive application might increase the amount of amine, thereby enhancing the polymerization reaction and increasing the *V* value. Our null hypothesis that the application of self-etch adhesives did not affect the polymerization reaction was therefore partially rejected. The use of adhesives might be useful for enhancing the polymerization reaction of core build-up resins and could potentially increase the hardness in regions of the resin–dentin interface where there is limited access to light [28]. In contrast, no significant difference in the *V* values was observed with or without adhesive for UF. In comparison to DC, UF appeared to have a more efficient chemical setting reaction, which could lead to a stronger polymerization reaction even in the absence of light attenuation (Figure 3).

The polymerization behavior of core build-up resins depends on the monomer composition, the concentration of the initiator system and the types of filler used [29,30]. The present *in vitro* findings suggest that light irradiation with a sufficient power density is necessary to achieve optimal mechanical properties, even in materials with a dual-cured setting reaction. Adequate polymerization is a pre-requisite for the overall success, longevity and biocompatibility of restorations. Recent advances in light-curing techniques have attempted to improve the dentin-bonding capability. The use of self-etch adhesives in combination with dual-cured core build-up resins appears to be beneficial from the view point of the polymerization reaction of the materials. Further studies are needed to establish how core build-up resin systems including self-etch adhesives bond to tooth structures in clinical situations.

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