

Light curing of resin-based composites in the LED era

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ABSTRACT: This review thoroughly accumulated information regarding new technologies for state-of-the-art light curing of resin composite materials. Visible light cured resin-based composites allow the dentist to navigate the initiation of the polymerization step for each layer being applied. Curing technology was regularly subjected to changes during the last decades, but meanwhile the LED era is fully established. Today, four main polymerization types are available, *i.e.* halogen bulbs, plasma arc lamps, argon ion lasers, and light emitting diodes. Additionally, different curing protocols should help to improve photopolymerization in terms of less stress being generated. Conclusions were: (1) with high-power LED units of the latest generation, curing time of 2 mm thick increments of resin composite can be reduced to 20 seconds to obtain durable results; (2) curing depth is fundamentally dependent on the distance of the resin composite to the light source, but only decisive when exceeding 6 mm; (3) polymerization kinetics can be modified for better marginal adaptation by softstart polymerization; however, in the majority of cavities this may not be the case; (4) adhesives should be light-cured separately for at least 10 seconds when resin composite is applied directly; (5) photocuring through indirect restorations such as ceramics is still a problem, therefore, both dual-cured adhesives and dual-cured composites and resin coating in any way are recommended; and (6) heat generation with high-power photopolymerization units should not be underestimated as a biological problem for both gingival and pulpal tissues. (*Am J Dent* 2008;21:135-142).

CLINICAL SIGNIFICANCE: Light curing today is facilitated by the latest generation LED units providing light intensities of up to 2,000 mW/cm². Compared to earlier recommendations, curing time for 2 mm resin composite layers can be limited to 20 seconds, when the restorative is not more than 6 mm away from the light tip. However, severe heat generation always must be taken into account.

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Introduction

Visible light cured resin-based composites are the predominant restorative materials for both anterior and posterior restorations. In 2000, 94% of U.S. dentists used visible-light curing units.¹ Light-cured composites allow the dentist to actively initiate the polymerization step being a significant advantage compared to autocured composites.² Furthermore, a meticulous layering technique was employed to reduce polymerization shrinkage to be applicable even in larger stress-bearing cavities in re-dentistry.³ This enables the dentist to generate esthetic and durable restorations such as pit and fissure sealants, direct and indirect resin composite restorations, and luting of ceramic restorations. Even resin-modified glass ionomers rely on photopolymerization.⁴

The mode of curing has regularly changed during the last 30 years. By 2007, the era of light emitting diodes has been definitively established (Fig. 1). Today, four main types of polymerization sources are available: Halogen bulbs, plasma arc lamps, argon ion lasers, and light emitting diodes. Furthermore, different curing protocols were designed to improve photopolymerization, as there is soft-start, step-curing, or oscillating irradiation.⁵ Based on this background, the present review focused on questions arising from this change in technology:

1. Have clinical recommendations changed in terms of shorter polymerization intervals?
2. Are there differences in curing depth?
3. Are there differences in polymerization kinetics and shrinkage performance?

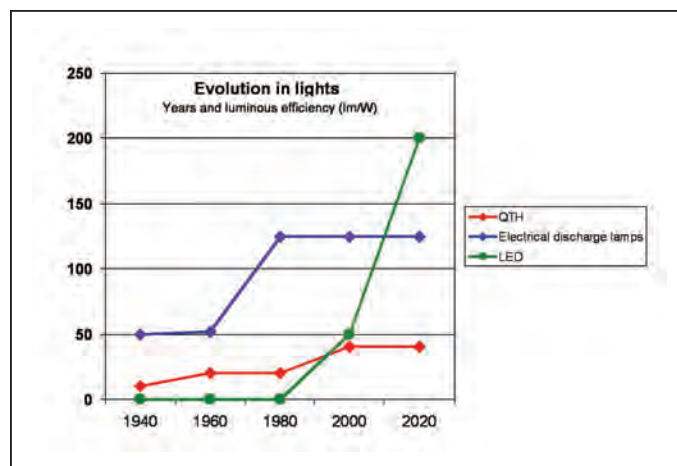


Fig. 1. Achievements in the area of lighting with different technologies.

4. Which are the actual recommendations for curing through tooth-colored restorations?
5. Is heat generation a clinically relevant problem?

Polymerization of resin composites

Important for any polymerization is the resin matrix of composites, mainly di- or tri-esters of methacrylic acid. Those have proven ability to survive under intraoral conditions, since only methacrylates are found to be linked to different organic parts such as aliphatic chains, polyethers and, and aromatic ring structures. The most common molecule is the so-called Bowen-monomer BisGMA (2,2-bis [4-(2-hydroxy-3-methacryloxy-

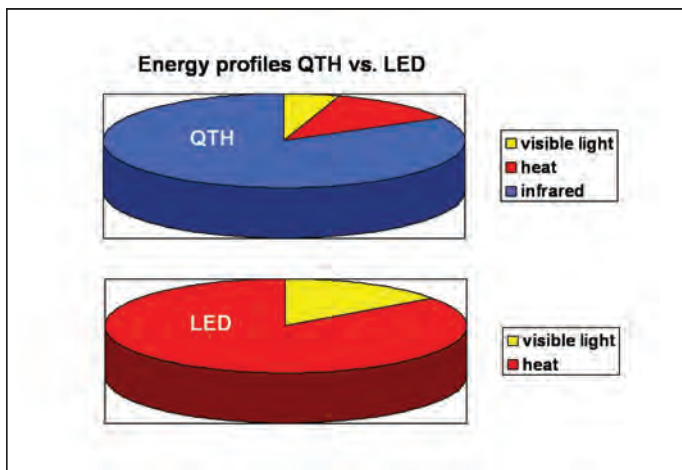


Fig. 2. Energy profiles of QTH and LED lamps.

propoxy)phenyl]propane). The main advantage is its considerably reduced polymerization shrinkage compared to pure methacrylates and high crosslinking ability. Another common monomer is the aromatic UDMA (urethane dimethacrylate). UDMA is characterized by its contribution to color stability, hydrophobicity, high viscosity, and good diametral tensile strength.⁶⁻⁸

Radical polymerization is either initiated by redox systems in autopolymerizing resins being always delivered in two components or by visible light at a wavelength of 468 nm. Classic initiator systems are di-benzoylperoxide for self-curing systems and camphorquinone for photopolymerization with blue light, or in former times butyl hydroxytoluol or lucirine for ultraviolet light curing (< 390 nm).⁶

In addition to the resin matrix, camphorquinone still serves as photoinitiator in almost all commercially available composite materials. Its absorption range was found to be between 370-500 nm with a peak at 468 nm.⁹ This light spectra is responsible for effective light curing; however, sufficient intensity of the light source is a fundamental requirement to achieve acceptable material properties for intraoral use even in stress-bearing cavities and to prevent discoloration and premature degradation.¹⁰ The depth of cure is dependent on different co-factors such as filler particle size and distribution, color and optical translucency of the composite, and refractive index ratio of the single components being used.¹¹⁻¹⁵ Therefore, a minimal intensity at the most efficient wavelength is needed over a defined irradiation period. At a given depth, curing will not occur without the inhibitor being consumed by the generated radicals in this particular region.

Initiators divide C=C double bonds thus leading to crosslinking and build-up of a three-dimensional methacrylic network. The phase of growing chains is determined when monomer molecules are consumed or when two radicals react. In order to obtain good incorporation of fillers and to reduce setting stress, smaller and highly mobile co-monomers are added to the matrix, such as TEGDMA (triethyleneglycol dimethacrylate) or Bis-DMA (bisphenol dimethacrylate). These co-monomers inhibit quick setting after the polymerization is initiated. A high amount of these dimethacrylates guarantees high conversion rates; however, polymerization shrinkage and

hygroscopic expansion are increased.¹⁶

Polymerization of resin-based composites leads to a highly crosslinked structure, but steric hindrance causes residual unsaturation by pendant methacrylate groups. The degree of conversion is defined as the percentage of reacted C=C double bonds. It affects several important parameters such as flexural strength, fatigue, solubility, discoloration, and biocompatibility.¹⁷⁻¹⁹ It has been reported that the same degree of conversion is produced by a fixed amount of energy density, leading to the recommendation of an energy density of 21-24 J/cm² for proper polymerization of a 2 mm portion of resin-based composites.²⁰⁻²²

Energy densities (J/cm² or mWs/cm²), i.e. the product of light intensity (mW/cm²) and irradiation time(s), have been suggested to account for variations in irradiation intensity, time and mode. The same degree of conversion is produced by a fixed amount of energy (energy density: J/cm²), independent of variations in light irradiance.²⁰

Lamp technology

Quartz tungsten halogen (QTH)

QTH lamps have been the standard curing units for several years, despite a remarkably low efficiency compared to heat generation.⁵ Since QTH lamps emit a rather wide range of wavelengths, band-pass filters are required to limit the wavelength between 370 and 550 nm in order to fit the peak absorption of camphorquinone.²³ QTH lamps have a limited lifespan of 100 hours with consecutive degradation of bulb, reflector, and filter caused by high operating temperatures and considerable quantity of heat being produced during operating cycles.²⁴ This implicates a reduction of curing efficiency over time by aging of the components. Many QTH lamps used in dental offices operate beneath the minimum power output specified by the manufacturers.²⁵ This may even deteriorate over time due to insufficient maintenance of the light sources and especially the light tips. With QTH lamps, 5% of the total energy is visible light, 12% heat, and 80% light emitted in the infrared spectrum (Fig. 2).^{26,27}

Plasma arc curing (PAC)

Plasma arc curing lamps emit at higher intensities²⁸ and were primarily designed to save irradiation time as an economic factor. PAC lamps emit light from glowing plasma being composed of a gaseous mixture of ionized molecules such as xenon molecules and electrons.

PAC units are characterized by high intensities in a narrow range of wavelengths around 470 nm. Due to the described high energy output of plasma arc systems, the manufacturers of these lamps repeatedly claimed that 3 seconds of PAC irradiation would achieve similar material properties compared to 40 seconds curing with QTH lamps. However, this claim has been fully rejected.^{27,29-32} Today, recommendations for PAC lights are based on 3 x 3 seconds.³³

Argon-ion lasers (AL)

Argon lasers emit blue-green light of activated argon ions in selected wavelengths (between 450 and 500 nm) and are therefore suitable for light-curing of resin-based composites.³⁴ Argon-ion lasers operating with 250 ± 50 mW/cm² for 10 seconds achieve improved curing of light-activated restorative

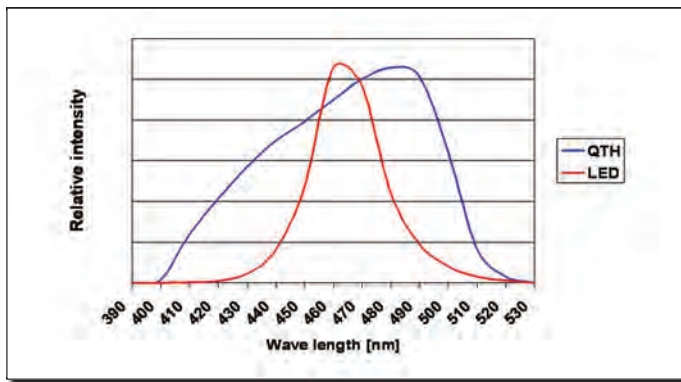


Fig. 3. Characteristic spectra of QTH and LED lamps.

materials in a shorter period of time resulting in equal or even superior physical properties as compared to the conventional QTH systems on the market.³⁵ On the other hand, heat generation during polymerization combined with considerably high initial shrinkage stresses have been reported to be problematic.^{36,37} Compared to QTH, argon-ion lasers obtain higher conversion rates³⁴ and polymerization depths.³⁸ In general, the literature in the field reflects a strong divergence of opinions covering many aspects of the efficiency of laser curing compared to conventional light curing.³⁸

Light emitting diodes (LED)

To solve the previously described problems being connected with conventional QTH technology, solid-state LEDs were introduced to the market.³⁹ Whereas halogen bulbs operate with a hot filament, LEDs use junctions of doped semiconductors (p-n junctions) for the generation of light. In gallium nitride LEDs under forward biased conditions, electrons and holes recombine at the LEDs p-n junction leading to the generation of blue light. A small polymer lens in front of the p-n junction partially collimates the light.⁴⁰

The spectral emittance of gallium nitride blue LEDs cover the absorption spectrum of camphorquinone so that no filters are required in LED light curing units.⁴¹ Recent reports revealed that blue LED lamps offer the highest photo polymerization efficiency.²³ LEDs are less energy-consuming compared to QTHs and do not require external cooling in the majority of products on the market. Moreover, LED lamps have a lifetime of several thousands of hours without a significant intensity loss. LEDs emit approximately 15% visible light and 85% heat (Fig. 2). In the direction of the curing tip, LEDs are mainly not emitting heat; however, 85% heat is produced in a backward direction.²⁶

LEDs were subjected to dramatic changes in technology over the last 10 years (Fig. 3). The development of recent generations of high power LEDs is comparable to advances in high tech computer technology. Not so long ago, the power density of early LED generations was very low which forced the manufacturers to build complicated arrangements of 10 to 15 diodes into one lamp (Fig. 4). This was the reason why the first generation of LED curing units could not compete with conventional QTH units.^{27,42} The initial expectations towards a possible reduction of curing times by use of first generation LEDs could not be confirmed.⁴³

Today, LED technology has considerably changed towards

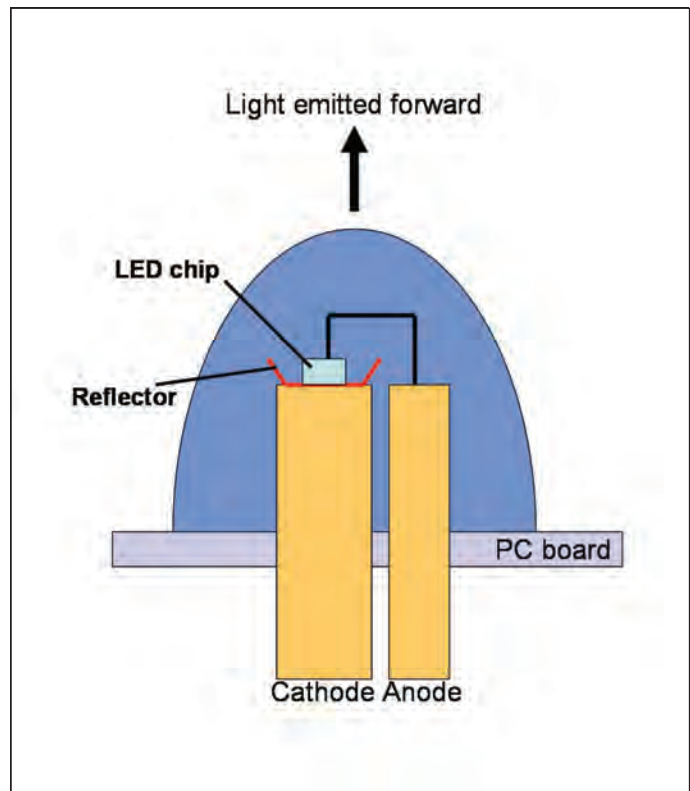


Fig. 4. Diagram of old-styled conventional LED lamps.

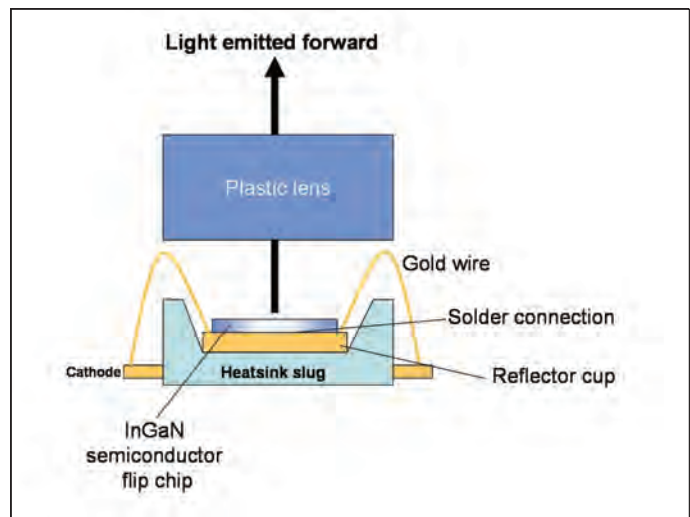


Fig. 5. Diagram of recent high-power LEDs.

high power LEDs being capable of delivering a rather high output with one single diode inside the curing unit (Fig. 5).¹ On the other hand, heat generation became a clinical concern for gingival and pulpal tissues using power LEDs. This is caused by the so-called photodynamic effect. Facing maximum light intensities of up to 2,000 mW/cm², the problem of heat generation should be seriously taken into account. Power LEDs definitely rely on external cooling.

Curing time

For the incremental technique for layering of resin composites, the maximum thickness of each individual com-

posite layer was advocated to be < 2 mm with a required curing time of 40 seconds for each layer.⁴⁴⁻⁴⁶ In order to achieve a maximum conversion rate, some authors recommended curing at lower intensities (< 500 mW/cm²) within extended polymerization intervals.⁴⁷ More recently, this paradigm was questioned more often when facing high output curing lights. Koran & Kürschner¹⁰ evaluated the variables hardness, adhesion, shrinkage, viscosity and degree of polymerization at different light intensities and different polymerization times with QTH. At energy densities > 17,000 mW/cm², no further improvement of mechanical properties was achievable. This leads to the conclusion that with latest generation LED units providing output levels consistently between 1,500-2,000 mW/cm², polymerization time can be reduced to 20 seconds.⁴⁸

Experiences with plasma arc curing demonstrated that 3 x 3 seconds light curing with constant high energy densities are sufficient for appropriate polymerization of hybrid resin composites.³³ Nevertheless, in the majority of surveys dealing with curing light intensity and curing time in private dental offices, curing units often lack maintenance and thus provide weak performance, combined with curing times often being limited to 20 seconds.^{2,49} Therefore, compensation of these practically relevant problems by higher energy output may be the most important point in recent photopolymerization technology. Derived from these observations, and based on the surveys published, most of the resin composite restorations in dental offices may not be sufficiently cured with all consequent disadvantages such as higher abrasion and less biocompatibility. This situation could be changed in favor of more durable and biocompatible resin composite restorations in the future.

Curing depth

Caughman *et al*¹¹ postulated clinical guidelines for photocuring in 1995, indicating secure polymerization of resin composites for layers < 2 mm at 280 mW/cm². For QTH light units and 3 mm layers, even at 800 mW/cm² and 80 seconds exposure time (energy density of 64,000 mW/cm²) no adequate polymerization was achieved.⁵⁰ These results question the recommended bulk curing of packable resin composites.⁵¹

A recent study⁵² demonstrated a linear relationship between light intensity of both QTH and LED lamps and curing depth. Interestingly, even prolonged curing times did not guarantee higher curing depths.^{52,53} If the light tip is placed at a distance of more than 6 mm from the resin composite surface, polymerization depth is affected.⁵³ At a distance of 12 mm from the light tip, no appropriate curing of resin composites was achieved, being independent of the type of light (QTH vs. PAC) and the curing mode (soft start vs. standard).⁵⁴

Light-curing through ceramic restorations is still a considerable problem. For ceramic inlays, dual-cured and solely light-cured resin composites are described.⁵⁵ The polymerization depth of solely light-curing resin composites depends on the thickness of the ceramic itself, and from the shade and material of the inlay (ceramic or composite).⁵⁶ Safe polymerization beneath ceramic inlays is possible up to approximately 3 mm distance from the polymerization tip.^{57,58} However, with darker inlay shades, curing of luting composites

is reduced already with ceramic thicknesses of more than 2 mm.⁵⁹ The same is true for LED units of the first generation exhibiting weak curing potential through a ceramic layer thickness exceeding 2 mm.¹³ Compared with light-curing resin composites, dual-cured materials exhibit improved curing through ceramic discs.⁶⁰ QTH units have been reported to be more efficient compared to PAC units.⁶⁰

Degree of conversion

The degree of conversion of a methacrylic resin composite is defined as the percentage of reacted C=C bonds. This ratio substantially affects many properties including mechanical properties, solubility, dimensional stability, color change and biocompatibility of the resin composite.^{21,61,62} Thus, the degree of conversion plays an important role in determining the ultimate success of a light activated direct restoration.^{32,63}

Degree of conversion is commonly measured by Fourier transform infrared reflectance spectroscopy (FTIR). This method has been reported to produce highly reliable results.⁶⁴ Calculation is based on the measurement of the net peak absorbance area of the C=C bonds and the aromatic C-C bonds as reference. The net absorbance peak area ratio of cured to uncured material provides the percentage of converted double bonds. Based on that method, a variety of correlations could be proved. It has generally been observed that the higher the conversion in resin composites the higher the polymerization shrinkage will be.⁶¹ However, by applying increased light intensities, composites restrain this stress relief much more by not allowing enough flow to reduce internal stress.⁶⁵ Applied moderate light intensities, in contrast, activate a reduced concentration of initiator molecules to form the network more slowly, thus allowing the material to flow during the early stages of curing.^{10,61} Within a narrow range, the same degree of conversion is produced by a fixed energy density, independent from variations of light irradiance and exposure time.²⁰ Polymerization at extremely high light intensities was found not to result in adequate curing, due to inferior measured flexural moduli and inferior depths of polymerization.³²

Recent work²⁰ has shown a close correlation between energy density and degree of conversion. Also, increased energy densities lead to superior physical and mechanical properties,⁶² such as fracture strength and the surface degree of conversion, but worse bulk properties like reduced flexural fatigue limits and in-depth degree of conversion. Bulk properties were found to be improved by applying moderate light intensities.⁶⁶

An important effect on the final mechanical behavior is derived from the applied energy densities and thus from the induction of internal stresses.¹⁰ It is conceivable that the different irradiation protocols and thus the build-up of internal stresses will lead to different polymer structures, even though the degree of conversion is the same. A reduced intensity polymerization is probably associated with relatively few centers of polymer growth which may result in a relatively low crosslinked structure.⁶⁷ High light intensity in the initial phase of the irradiation period will, in contrast, initiate a multitude of growth centers, resulting in a highly crosslinked polymer. Even with a high degree of conversion, a resin composite based on a

polymer with few crosslinks may be more sensitive to crack initiation or visco-elastic degradation.⁶⁷ Furthermore, cross-linking differences might be derived from the use of different monomers. A significantly increased degree of conversion was found for UDMA compared to BisGMA monomers.²¹

For clinically successful restorative dentistry, a minimum degree of conversion has not yet been precisely established. Nevertheless, a negative correlation of *in vivo* abrasive wear depth with dual conversion has been established for dual conversion values in the range between 55 to 65%.⁶¹

Polymerization of adhesives

In order to achieve durable bonds between tooth dental hard tissues and directly applied resin composites, a separate polymerization of the adhesive is routinely performed. This is proven to be beneficial especially for dentin aspects.^{68,69} However, differences relating to the outcome of polymerization of adhesives with different curing units are scarcely reported, with some advantages for QTH lights.⁷⁰ Regarding the duration of the separate light-curing step, manufacturers normally recommend a 10-second period. An unpublished study from our laboratory confirmed this; for Heliobond,^a part of the Syntac^a adhesive system, no separate curing resulted in significantly more gaps in adhesive Class V restorations after thermo-mechanical loading.

A completely different situation is found when adhesives placed under ceramic or composite inlays are polymerized. Hikita *et al*⁷¹ named critical factors related to adhesive luting of indirect tooth-colored adhesives. When no separate light-curing of a solely light-curing adhesive is carried out prior to cementation, adhesive performance may be poor. A previous study⁷² also demonstrated the separate light-curing to be beneficial prior to the application of luting resins; however, light-curing of the bonding resin prior to the insertion of the luting composite produced unacceptable large diameters of the luting spaces.^{72,78}

One possible solution of this polymerization problem beneath tooth-colored inlays may be the introduction of truly dual-polymerizing adhesive/composite combinations.⁷⁴ However, after 4 years of clinical service no difference was found compared to a not separately cured light-curing adhesive for bonding of a dual-cured resin composite.

Recent results⁷⁵ indicated that the separate light-curing of the adhesive prior to adhesive cementation may be an over-estimated phenomenon. Due to the fact that the dentist desperately tries to thoroughly air-thin the adhesive to avoid pooling, it may not polymerize any more due to oxygen inhibition.⁷⁵ The most promising way to effectively seal the dentin may therefore be an adhesive lining or build-up, which is referred to as immediate dentin sealing or resin coating technique. Both techniques guarantee contamination-free reliable curing of the adhesive and therefore better sealing as represented by high dentin bond strengths.⁷⁵⁻⁷⁸

Polymerization kinetics, strain, stress

Major shortcomings of resin-based composites are inferior conversion and its intrinsic polymerization shrinkage. However, from the clinical point of view, these properties are al-

ways in conflict with each other. Increased conversion enhances resistance to wear and flexural fatigue, while an increased polymerization shrinkage and thus a higher stress level in bonded resin-based composite restorations is expected.¹⁹

During polymerization, dental resin composites transform from plastic viscous through a rubbery visco-elastic into an elastic glassy stage. Initially, the composite remains in its viscous stage and is then able to flow prior to reaching the glassy stage. After passing the gel-point, steric hindrance becomes prominent and with that elastic properties are measurable.⁷⁹ The elastic modulus increases with growing conversion reaching its final level at the glassy stage. Therefore the degree of conversion has a substantial effect on finally obtained mechanical properties and wear resistance being independent of the cure method.⁸⁰ In order to investigate changes in modulus and visco-elastic properties of resin-based composites, dynamic mechanical thermal analysis is routinely used. Different polymeric transitions can be identified under changing testing conditions.¹⁹

Curing protocols

Different light curing protocols are available such as soft start, step curing, or oscillating irradiation. These special curing modes have been considered to increase the degree of conversion for better material properties, and to decrease internal stress to achieve better marginal quality in bonded resin composite restorations.^{42,65}

The introduction of step curing may be interpreted as the first attempt to reduce initial shrinking stress by delaying the gel phase.^{81,82} During the pre-gel phase, the resin composite flows, and the stresses with the structures are relieved.⁷⁹ After gelation, flow ceases and cannot compensate for shrinkage stresses.⁸³ Using step curing, the polymerization process is started for 10 seconds on a low level of intensity (100 mW/cm²). Consecutively, the light unit automatically increases the power output to 700 mW/cm². First results were promising and indicated less polymerization stress compared to conventional QTH curing,⁸¹ however, after 10 years, no unanimous proof for improved marginal adaptation could be found.⁸⁴

Soft start polymerization means also starting at a lower level (100 mW/cm²); however, the increase to the final power density (800 mW/cm²) takes an exponential curve. This special curing protocol is offered with different QTH and LED models. Nevertheless, the effectiveness of soft start curing is not unanimously clarified. On one hand, a certain reduction of polymerization stresses was shown.^{31,85} On the other hand, a true beneficial effect on marginal quality of resin composite restorations was not proven.⁸⁶⁻⁸⁸ Only in Class V cavities, some positive effects have been reported.⁸⁹

The pulse-activated polymerization uses short impulses of high intensity (*e.g.* e-light^b: 10 pulses for 2 seconds each 750 mW/cm²). To date, no enhanced polymerization kinetics have been found.⁸⁵

The so-called pulse-delay technique was repeatedly investigated *in vitro*. With this technique, the restoration is initially irradiated with short pulses of light energy (prepolymerization at low light, *e.g.* 3 seconds or 20 seconds with 100 mW/cm²). After a short waiting period of 3 minutes, the final polymerization is carried out for 30 seconds at high intensi-

ties.⁹⁰ Also, a significant advantage could not be found.⁹¹ Finally this method is controversial due to the low degree of polymerization at the cavity floor.⁹¹

Heat generation as a biological concern

The most significant temperature rise during the application of direct resin composite restorations is found during light-curing.⁹² The heat generated by photopolymerization can theoretically damage pulpal and gingival tissues.⁹³ Dentin is reported to behave as a good isolating substrate, however, in deep cavities a thin remaining dentin thickness may be problematic.⁹⁴ With less than 1 mm dentin thickness remaining, a critical temperature rise of 5.6°C inside the pulp chamber has to be taken into account.⁹⁵ When LEDs have been introduced to the market, lower heat generation was expected involving less risk of tissue damage.⁹⁶

However, even in modern power LEDs, up to 93% of the total energy amount is still heat.²⁶ Facing the fact that recent generations of high power LEDs reach output intensities of up to 2,000 mW/cm², the problem in deep cavities and those close to the gingiva may be even more serious compared to QTH curing units.⁹⁶ Therefore, some manufacturers already included special curing modes for adhesives and first layers for cavity floor dentin in deep areas being close to the pulp. Inside the resin composite, temperature was found to rise up to 10°C, therefore in deep cavities LEDs and QTHs are recommended with lower energy.^{97,98} Darker shades also promote heat generation within the resin composite.⁹⁹

Polymerization from the clinical view

Dealing with photopolymerization of resin-based composites, it must be taken into account that clinical circumstances may often differ considerably from laboratory conditions. There is a difference between resin-based composites light cured in the laboratory under perfect conditions with 100% access to cavity and materials cured in the oral cavity. Also in the oral cavity, a shallow Class V in an upper first incisor is easier to irradiate than a deep Class II in a second molar. Therefore, clinical recommendations always have to be adjusted to the individual clinical situation.

Due to the fact that in the majority of cases the dental assistant is guiding the photopolymerization steps, a certain amount of knowledge is fundamental for clinical success. How many assistants know that the distance from the light tip may be one of the most crucial factors in adhesive dentistry?

Nevertheless, the recent achievements make the older guidelines questionable. In 1995, an exposure time of 60 seconds at 280 mW/cm² was strictly recommended.¹¹ Today, with the latest generation of LED units, curing time of 2 mm thick increments of resin composite can be reduced to 20 seconds to obtain durable results.

Curing depth is fundamentally dependent on the distance of the resin composite to the light source, but only decisive when exceeding 6 mm. The polymerization kinetics can be modified for better marginal adaptation by soft start polymerization, however, in the majority of cavities this may not be the case. It is still not proven whether modified polymerization protocols improve clinical long-term success. Adhesives should be light-

cured separately for at least 10 seconds when the resin composite is directly applied. This also affects heat generation inside the pulp chamber, so finally heat generation with high-power photopolymerization units should not be underestimated as a biological problem for both gingival and pulpal tissues. The photocuring through indirect restorations such as ceramics is still a problem; therefore both dual-cured adhesives and dual-cured composites and resin coating are recommended.

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