Direction of shrinkage of light-curing resin composites

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It is occasionally stated in the dental literature that light-curing resin composites shrink towards the light source. As light travels at the speed of light, this dictum is not obvious. It was the purpose of the present study to investigate the direction of shrinkage of a light-curing resin composite in relation to the attachment and the thickness of the material. The resin composite was applied in cylindrical brass molds in such a manner that a flash, serving as attachment, was produced at one side of the specimens, while the material was flush with the mold at the other side. The specimens were now irradiated from either the flash or the flush side, and the convexity or the concavity of the specimens was measured. At a material thickness of 3 mm, the shrinkage was towards the light source, irrespective of the position of the flash. At 4 and 5 mm thickness of the molds, the direction of shrinkage could be directed towards or away from the light source, depending on the position of the flash. The number of light quanta emitted from the light source and passing through the material was compared with the number of molecules of camphorquinone present in a resin composite of 3, 4, or 5 mm thickness. It was concluded that under the conditions of the present study, the direction of shrinkage was the result of an interplay between the direction of the light, the attachment of the material, and the thickness of the material. □ *Blue light; light quanta; polymerization kinetics*

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It is occasionally stated in the dental literature that light-curing resin composites shrink towards the light source, while so-called chemically-curing resins shrink towards the center of mass (1–3). There is very little evidence to support the statement as regards the light-curing materials; this has been considered as self-evident. The concept has been the basis of recommendations for certain procedures in clinical practice, procedures according to which the direction the light is shone from is thought to be of significance in minimizing gap formation and maximizing bond strength to dentin and enamel (1, 2, 4, 5). However, a recent theoretical study questions this concept, concluding that the direction of shrinkage of a light-curing composite is determined predominantly by cavity shape and bond quality (6).

As light (obviously) travels at the speed of light, in a very short time the curing resin composite should be saturated by the curing light, and the direction the light is supplied from would seem to be of no importance. The resin material would not "know" in which direction to shrink. However, as light intensity decreases through depth, the state of saturation would depend on the number of light quanta emitted from the light source, and on the number of initiator molecules per volume traversed by the light. This would imply that at small thickness of material, the shrinkage of the resin composite would have no relation to the direction of the light, and would be directed towards the site of greatest attachment. Only at a material thickness above a certain level would the material shrink towards the light source. In general, the depth of penetration of light into a resin composite depends on type and amount of filler, as well as on particle size and distribution (7). Besides these factors, the critical thickness of material would depend on the intensity of the light source and on the number of initiator molecules per volume of resin composite.

It was the purpose of the present study to investigate the direction of shrinkage of a light-curing resin composite in relation to the attachment and the thickness of the material. It was hypothesized that the attachment would play a role and, superposed on this influence, a tendency would be present that at small thickness of material the direction of shrinkage would be independent of the direction of the light, and only at a large thickness would the shrinkage be towards the source of the light.

Materials and methods

The light-curing resin composite was Z100 of shade A3 (3M Company, St. Paul, MN, USA). A ring-shaped brass mold (outer diameter = 20 mm, inner diameter = 6 mm) was placed on a transparent matrix band and filled with excess of the resin composite. The top surface was covered with another strip of matrix band and the sample compressed between the plane metal plates of a paralleling device for about 5 s. The metal plates aimed at ensuring plane and smooth surfaces of the resin composite before polymerization, and were removed at the end of the compression period. In this manner, the lower part of the resin composite was confined within the mold, flush with the surrounding mold surface, whereas the upper part of the composite gave rise to the production of a flash (Fig. 1). The flash was intended to act as the site of greatest attachment of the polymerizing material. The inner walls of the mold would provide an attachment that was not

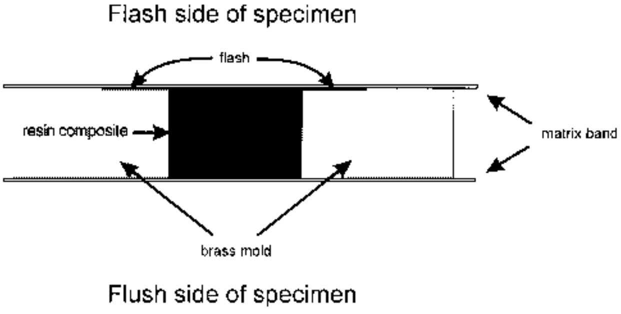

Fig. 1. Mold used to prepare the specimens. After being applied into the mold in small excess, the resin composite was covered with a transparent matrix band. A flash was produced with a paralleling device (not shown). The resin composite was now irradiated from either the flash side, or the opposite side, the flush side.

related to the one or the other side of the mold. With close contact between matrix band and light-tip (diameter = 8 mm), the assembly was now irradiated for 40 s with the curing lamp Visilux 2 (3M Company, St. Paul, MN, USA). The curing lamp gave maximum response in less than a second when the light intensity was tested with the radiometer CL-tester (CDB, Huddinge, Sweden), and resulted in a complete cure of the resin material. In one series, the irradiation took place from the side of the flash and in another series the irradiation was performed from the side having no flash—the flush side. In this manner, the site of greatest attachment was either the side of the mold closest to the light source or the side opposing the light source, depending on the position of the flash. The attachment provided by the flash when the specimen was irradiated from the flush side would stem from the material in the immediate vicinity of the hole polymerized by diffused light, even though not directly irradiated. Three series of molds were used, 3, 4, and 5 mm, respectively, in height. After the irradiation, the matrix bands were removed and the specimens (resin composite and mold) were stored dry for 1 day at 37°C to bring the polymerization to an end. Each specimen was then placed under a dial gauge as previously described (8). The specimen was affixed to a glass slide and placed on the moving table of an apparatus on which the dial gauge was mounted. The dial gauge allowed for the position of the stylus to be read with an accuracy of ±1 μm. The stylus was lowered onto the specimen 0.25 mm peripheral to the border of the circular hole in the mold and the reading recorded. By means of the moving table the specimen was now moved 0.50 mm and the dial gauge read again. In this way the changes in height across the diameter of the specimen were recorded every 0.5 mm. The changes in height were measured on the flash side and also on the flush side of each specimen. For each of the 3 thicknesses

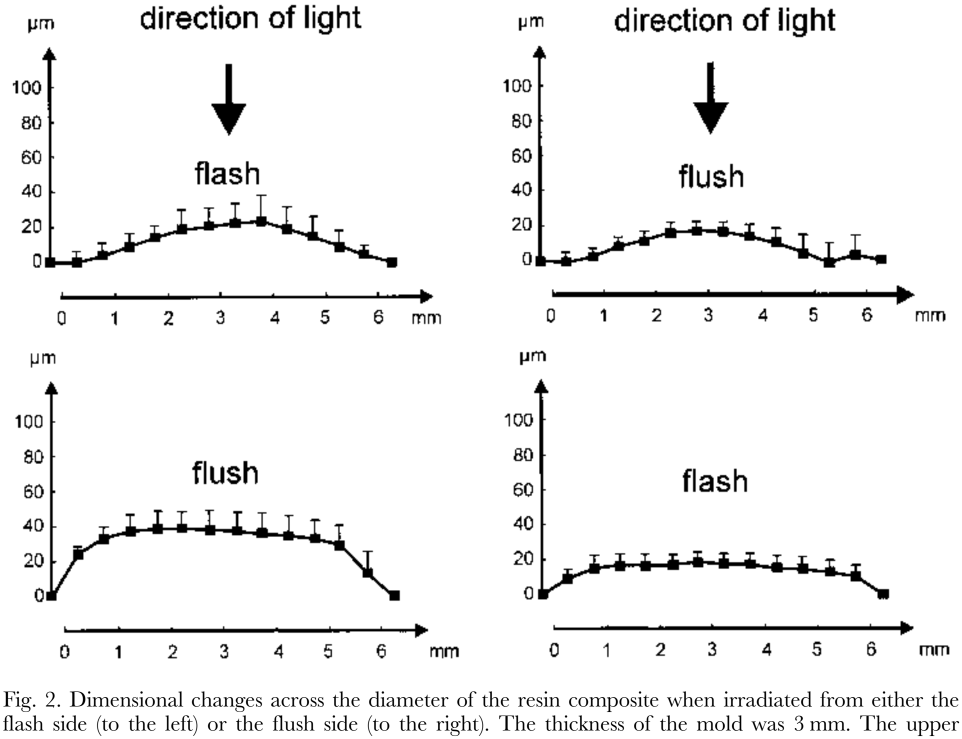

Fig. 2. Dimensional changes across the diameter of the resin composite when irradiated from either the flash side (to the left) or the flush side (to the right). The thickness of the mold was 3 mm. The upper diagrams give the dimensional changes of the irradiated sides of the specimens, with positive values indicating convexity. The lower diagrams give the dimensional changes at the sides away from the light source, with positive values indicating concavity. Mean values and standard deviations are given ($n = 5$). Analysis of variance showed significant differences ($P < 0.05$) between the right and the left side diagrams.

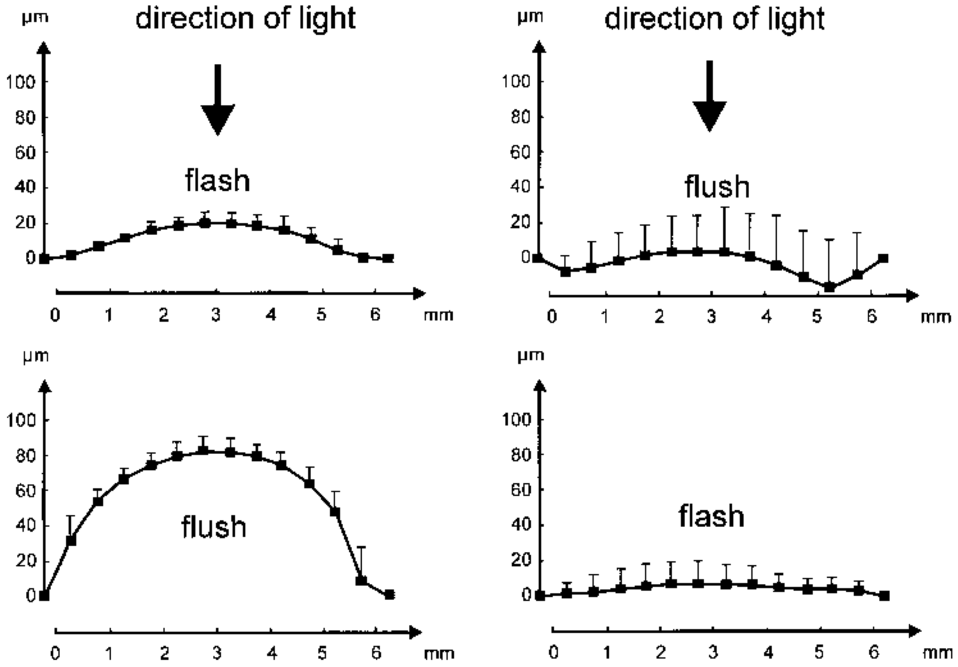

Fig. 3. Dimensional changes across the diameter of the resin composite when irradiated from either the flash side (to the left) or the flush side (to the right). The thickness of the mold was 4 mm. The upper diagrams give the dimensional changes of the irradiated sides of the specimens, with positive values indicating convexity. The lower diagrams give the dimensional changes at the sides away from the light source, with positive values indicating concavity. Mean values and standard deviations are given ($n = 5$). Analysis of variance showed significant differences ($P < 0.05$) between the right and the left side diagrams.

of the molds, 5 specimens were investigated. The means and standard deviations were calculated, and the results compared by analysis of variance (9).

Results

The results are presented in Figs 2–4. The black squares represent the mean values of the measurements. The T-shaped figures on top of the black squares represent the standard deviation. The curves are constructed so that they give a direct visual impression of the way the upper and lower surfaces changed shape as a consequence of the curing. Thus, positive values in the upper diagrams indicate a convexity of the surface, while positive values in the lower diagrams indicate a concavity.

A comparison of Figs 2–4 shows that the thickness of the molds influenced the shrinkage pattern. At a material thickness of 3 mm (Fig. 2), the direction of the shrinkage was towards the light source, and although statistically significant the difference between the position of the flash was small, i.e. the difference between the right and the left diagrams is not very pronounced. At a material thickness of 4 mm (Fig. 3), the shrinkage was towards the light source when the specimens were irradiated from the flash side, but at certain points *away* from the light source when the specimens were irradiated from the flush side. A comparison between the right and the left diagrams demonstrates a significant influence of the position of the flash. Finally, at a material thickness of 5 mm (Fig. 4), the shrinkage pattern demonstrated in Fig. 3 was accentuated. When the specimens were irradiated from the flush side, the mean movement of the flash side was away from the light source.

Discussion

The present study has demonstrated a complex pattern of shrinkage of a resin composite, with interactions between the direction of the curing light, the thickness of the polymerizing material, and the position of the flash. The differences in contraction pattern demonstrated in Figs. 2–4 may be discussed in terms of number of light quanta passing through the polymerizing resin material compared with the number of initiator molecules present in the material. To carry out this discussion, a few realistic assumptions have to be made. Assuming the intensity of the curing lamp to be $I = 700\,\text{mW/cm}^2$ (10), the wavelength of the blue light to be $\lambda = 470\,\text{nm}$ (11), the energy of 1 quantum of light to be $E = h \,.\, c/\lambda$ (12), in which expression h is Planck's constant and c is the velocity

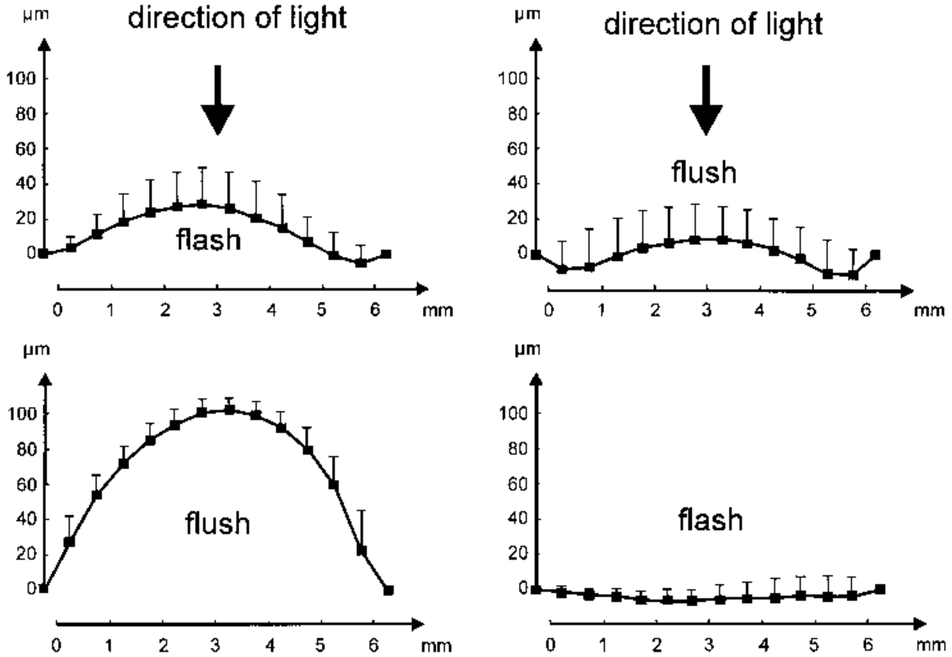

Fig. 4. Dimensional changes across the diameter of the resin composite when irradiated from either the flash side (to the left) or from the flush side (to the right). The thickness of the mold was 5 mm. The upper diagrams give the dimensional changes of the irradiated sides of the specimens, with positive values indicating convexity. The lower diagrams give the dimensional changes at the sides away from the light source, with positive values indicating concavity. Mean values and standard deviations are given ($n = 5$). Analysis of variance showed significant differences ($P < 0.05$) between the right and the left side diagrams.

of light, the number of quanta N emitted per s and per cm^2 can be estimated as $I_0 = I/E = 2 \,.\, 10^{18}$ quanta/(s . cm^2).

As regards the initiator, this may be assumed to be camphorquinone (9). The number of molecules N_M of camphorquinone in 1 cm^3 of monomer may be estimated if it is assumed that the density of the monomer is $\rho = 1.1$ g/cm^3 (13). The molar weight of camphorquinone is M = 166 g/mol, the concentration of camphorquinone C = 0.15 w% (13, 14), and Avogado's number is $N_A = 6 \,.\, 10^{23}$. The calculations give $N_M = 1 \,.\, \rho \,.\, C \,.\, N_A/M = 6 \,.\, 10^{18}$ molecules/cm^3. With a filler content of 67 vol%, the number of molecules of camphorquinone in 1 cm^3 of resin composite becomes $N_C = 2 \,.\, 10^{18}$ molecules/cm^3.

A typical transmittance diagram from the work of Harrington et al. (15) is characterized by an initial, almost instantaneous, increase in intensity, an intermediate period where the intensity increases linearly with time t until a time T, after which the intensity becomes constant. The linear increase in intensity for $t \leqslant T$ is best explained as due to the gradual consumption of camphorquinone, allowing an increasing amount of blue light to pass the specimen. At time T all camphorquinone in the 2-mm thick specimens used has reacted, which means that the intensity becomes constant. Lambert-Beer's law (16) gives the intensity as $I = I_0 \,.\, e^{-a \,.\, d}$, in which equation I_0 is the intensity at the surface of the material, a is the absorption coefficient, and d is the distance from the surface of the material. An expression for the intensity of the blue light which takes Lambert-Beer's law as well as the data of Harrington et al., (15) into account, may be written as:

$$(1)\ I = t/T \,.\, I_0 \,.\, e^{-a \,.\, d} \text{ for } t \leqslant T$$

$$(2)\ I = I_0 \,.\, e^{-a \,.\, d} \text{ for } t \geqslant T.$$

As mentioned, the linear increase in I with time is presumably caused by the consumption of camphorquinone, from which it follows that T must be proportional to the depth d. As T = 30 s at d = 2 mm (15), we have T = 30 . d/2.

The intensity of the light is related to the number of quanta N_Q as $I = dN_Q/dt$. Integration gives N_Q as a function $N_Q(t,d)$ of time and depth as

$$(3)\ N_Q(t,d) = \tfrac{1}{2}\, t^2/T \,.\, I_0 \,.\, e^{-a \,.\, d} \text{ for } t \leqslant T$$

$$(4)\ N_Q(t,d) = t \,.\, I_0 \,.\, e^{-a \,.\, d} - \tfrac{1}{2}\, T \,.\, I_0 \,.\, e^{-a \,.\, d} \text{ for } t \geqslant T.$$

From the data of Harrington et al. (15), "a" may be estimated from $I = I_0 \,.\, e^{-a \,.\, d}$, taking I_0 = 176 000 lx and I = 2 500 lx at a thickness of d = 2 mm. The calculation gives a = 2.15 mm^{-1}.

Using equations (3) and (4) and by taking $I_0 = 2 . 10^{18}$ quanta/(s . cm^2) as calculated above, the number of quanta $N_Q(30,2)$ passing a polymerizing resin composite of thickness d = 2 mm in 30 s can be calculated to be $0.4 . 10^{18}$ quanta/cm^2. The number of molecules of camphorquinone in 1 cm^3 of resin composite N_C was calculated to equal $N_C = 2 . 10^{18}$ molecules/cm^3. This is equivalent to a content of camphorquinone in a material thickness of 2 mm of $0.4 . 10^{18}$ molecules/cm^2. This number is in perfect agreement with the number of quanta just calculated to complete the consumption of camphorquinone. Of course, this very high degree of agreement should be regarded as incidental for several reasons. Firstly, quite a few assumptions were made. Other assumptions would have changed the fit of the agreement. Secondly, the light passing through a resin composite is not only absorbed by camphorquinone but is influenced by shade and to some degree dispersed by the filler particles (7). This would give a lower number of quanta than calculated although the dispersion of light is to some degree included in the "a" of equations (3) and (4). Further, in an electron spin resonance study it was found that the formation of radicals in 5-mm thick samples of light-cured resin in most cases did not reach a maximum unless the samples were irradiated for times longer than 40 s (17). This seems to indicate that the consumption of camphorquinone was not complete at T = 30 s, as estimated above. However that may be, the important thing is the general relationships that are expressed in equations (3) and (4). Credit to the general relationships in equations (3) and (4) may also be derived from the fact that the number of quanta passing in say 5, 10, and 20 s is the same for d = 2.0, 2.6, and 3.2 mm, respectively. This calculation is in agreement with measurements of depth of cure of resin composites as a function of curing time (18).

Calculated on the basis of equations (3) and (4), Fig. 5 shows the relationship between number of quanta/cm^2 as a function of time, traversing resin composites of 2, 3, 4, and 5 mm thickness. In a pilot study it was shown that about 30 s of illumination was sufficient to cure 5 mm of Z100 in the experiments reported here. In 30 s, the number of quanta reaching a depth d = 5 mm can be seen to be $N_Q(30,2) \approx 3 . 10^{14}$ quanta/cm^2. Further, from Fig. 5 it can be seen that this number of quanta, necessary to bring about polymerization, passes d = 4, 3, and 2 mm in approximately 8, 2, and 1 s. The contraction of a resin composite cannot be instantaneous. The irradiated molecules of camphorquinone must diffuse to react with the co-initiator or reducing agent (11), and then diffuse to react with the first molecule of monomer. Also, the presence of inhibitor will retard the beginning of polymerization (13, 19). Measurements have shown that 1–3 s may go by before shrinkage commences (10, 13, 20). This implies that at d = 3 mm or less, the number of quanta sufficient to cause polymerization passes through the material in a period of time shorter than necessary for the resin to start contracting. In contrast, at d = 4 or 5 mm, the material closest to the light source will have started contracting before a sufficient number of quanta reaches the deeper levels of the resin composite.

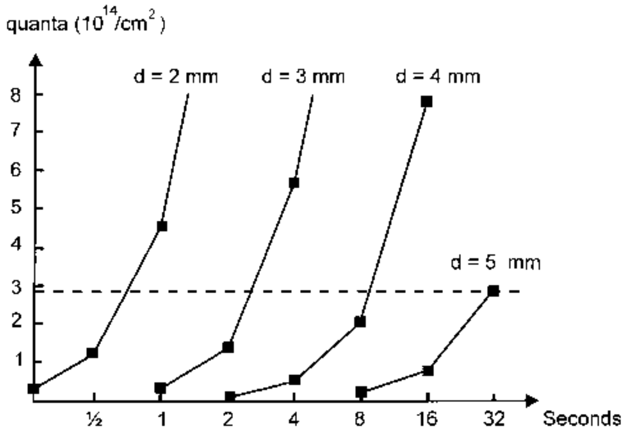

Fig. 5. Calculated number of light quanta passing 2, 3, 4, and 5 mm of resin composite in relation to time of irradiation. The dotted line indicates the number of quanta that will cause the resin composite to polymerize to a depth of 5 mm.

These considerations indicate that at a material thickness of 3 mm the direction of the light should not make much difference to the shrinkage pattern. However, this was not the case: in both the left and right sides of Fig. 2, the shrinkage was directed towards the light source and not, as expected, in both cases towards the flash side of the specimens. A possible explanation is that the flash did not act as an effective attachment in the situation to the right, where the flush side was irradiated. The specimen would have polymerized before the resin material in the flash close to the hole in the mold obtained sufficient strength to act as attachment. The observed expansion out of the mold is the result of complex interplay between forces of polymerization and attachment of the material. A fully satisfactory explanation cannot be given without further experimentation.

At material thicknesses of 4 and 5 mm, the above considerations indicate that the direction of the light should play a significant role in the shrinkage pattern. To the left in Figs 3 and 4, where the flash side of the specimens was irradiated, the flash and the material closest to the light source will polymerize before the material at the deeper parts of the specimen. The attachment provided by the flash will result in a shrinkage directed towards the light source and the flash. Also to the right in Figs 3 and 4, where the flush side of the specimens was irradiated, the material closest to the light source will polymerize before the material at the deeper parts of the specimen. As the material in the deeper parts of the specimen polymerizes, the attachment provided by the flash reduces the displacement of material towards the light source.

Returning to the hypothesis expressed in the introduction, this will have to be rejected: At small thickness of material, the shrinkage was not independent of the direction of the light, but in fact directed towards the light source. At larger thickness of material, the shrinkage was not in all cases directed towards the light source, but the direction of shrinkage also depended on the attachment of the material. However, the rejection of the hypothesis may, in part, be due to the experimental conditions of the present study. Further studies are needed to relate the described quantum approach to the clinical situation.

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