Composite restorative resins

Composition versus wall-to-wall polymerization contraction

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The purpose of the present work was to investigate a possible relationship between the composition of composite restorative resins and their wall-to-wall polymerization contraction. The investigated brands contained bis-gma diluted with varying amounts of one or two low-viscous monomers. The wall-to-wall contraction was measured microscopically as the maximum gap width in the dentin part of cavities in extracted human teeth. A positive correlation between gap width and amount of diluting monomer was found, with commercial composites and with non-composite, experimental resins. Addition of up to 75 % by weight of an inorganic filler to a monomer mixture similar to those found in commercial composites had no effect on the width of the contraction gaps. With non-composite experimental resins the gaps were found to increase with the amount of catalyst.

It was concluded that the composition of the organic phase is of primary importance for the size of the wall-to-wall contraction of composite restorative resins.

Key-words: Dental restoration, permanent; resins; dental materials

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It is an inherent property of restorative resins that they contract during polymerization. Under ordinary conditions this contraction always results in the formation of a gap between a central filling and the cavity walls, making ingress of bacteria, colouring matter, etc. into the cavity possible. Previous studies (*Asmussen & Jørgensen*, 1972) have demonstrated that non-composite resins do not necessarily show a greater wall-to-wall contraction than the composites, and that this contraction for the latter group of materials can vary significantly from brand to brand.

Restorative resins expand as a consequence of water absorption. This hygroscopic expansion will reduce the width of the polymerization contraction gaps, and it has been demonstrated by *Asmussen & Jørgensen* (1972) that the expansion for some but not all brands is sufficiently large to compensate completely for the wall-to-wall polymerization contraction. The w/w % water absorption of restorative resins has been measured by a number of investigators, e.g. *Viohl* (1974), and was found to vary significantly between brands.

An analysis of the literature published so far shows that an understanding of the

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observed variations in the wall-to-wall contraction and in the hygroscopic expansion is not possible when based exclusively on the filler content of the resins. On the other hand a recent study on the composition of the organic phase in several brands of restorative resins (*Asmussen*, 1975) may have provided, in part, a basis for a solution of this problem.

The purpose of the present work was to investigate a possible relationship between the composition of composite restorative resins and their wall-to-wall polymerization contraction in central cavities prepared in human teeth. The relationship between composition and hygroscopic expansion of the resins will be discussed in a following paper.

MATERIALS AND METHODS

Table I shows the brands used in the study.

The wall-to-wall polymerization contraction was measured as described in detail by *Asmussen & Jørgensen* (1972). Cylindrical cavities with a diameter and depth of approx. 2.5 mm were prepared in extracted human teeth. The prepared teeth were kept in water in a thermostat room at 37 ± 1°C, and immediately prior to filling the cavities were dried by compressed air. The filling materials were mixed at room temperature and were then carried into the thermostat room, where the cavities were filled in less than 45 seconds. The fillings were covered with a matrix during setting. Ten minutes after the filling of the cavities the restored teeth were placed in water at 37°C for 10 minutes. The teeth were thereafter abraded under water at 37°C approx. parallel to the surface of the fillings until dentin could be seen all the way round the cavities. The gaps were measured in a microscope at the point of greatest width, and the gap widths were expressed in percent of the cavity diameter. The gap measurements took place in the thermo-

Table I. *List of brands used in the investigation*

No.	Name	Batch No.	Manufacturer
A	Compact®	catalyst: 740121 universal: 740422	Svedia Dental Industri Enköping, Sweden
B	Compolite®	catalyst: 721102 universal: 721116	William Getz Corporation Illinois, USA
C	Opotow®	720420	Opotow Dental Mfg. New York, USA
D	Concise®	331317	3M Company Minnesota, USA
E	Concise cap-c-rynge®	3176 1G 212	3M Company Minnesota, USA
F	HL 72®	liquid: HL 0034 powder: HL 0038	Lee Pharmaceuticals California, USA
G	Prestige®	catalyst: HPR 0063 universal: HPR 0059	Lee Pharmaceuticals California, USA
H	Addent 12®	03 6301 C	3M Company Minnesota, USA
I	Blendant®	catalyst: 230.77 universal: 32410483	Kerr Manufacturing Company Michigan, USA
J	Smile®	catalyst: 330.05 universal: 923125.73	Kerr Manufacturing Company Michigan, USA
K	Exact®	47 303	S. S. White Pennsylvania, USA
L	DFR®	liquid: 581170 powder: 30 11 70	Surgident, Ltd. California, USA

stat room 30 to 45 minutes after start of mixing of the materials.

The study was divided in four sections in which the influence on the wall-to-wall contraction of the individual factors specified below under I—IV was studied. For each value of the variables studied, five fillings were investigated.

I. Diluent content of the organic phase of commercial brands. Before polymerization all brands contain bis-gma diluted with one or two low-viscous monomers (*Asmussen*, 1975). In brand A to G the bis-gma is diluted with tedma (tri-ethyleneglycol-di-methacrylate), in brand H to L other monomers serve as diluents. Table II shows the content of bis-gma and diluent(s) of the restoratives at the moment of mixing. The values in this table were obtained partly from the results in the above mentioned paper, partly from the calculations a—c specified below.

a. Paste-paste brands (A, B, D, G and K). The content of diluent was found as the mean value of this component in the two pastes.

b. Paste-liquid brands (H, I and J). The total amount of diluent was calculated in per cent of the resin component in a standard proportioned mix.

c. Liquid-liquid brands (L). The total amount of diluent was calculated in per cent of all the resin in a standard proportioned mix.

II. Diluent content of non-composite, experimental resins. Two series of non-composite resins were prepared, one series formed by mixtures of bis-gma and tedma, another by mixtures of bis-gma and edma (ethyleneglycol-di-methacrylate). The tedma content in the first series of mixtures was 0, 12.8, 24.7, 37.2, 49.9, 62.1 and 75.1 mole%, respectively, and the edma content of the second series 0, 22.8, 41.9 and 60.1 mole%, respectively. Each mixture was divided in two parts, forming a pair; 0.5 % by weight of dppt (di-isopropyl-p-toluidine) was dissolved in the first part and 3 % by weight of bpo (benzoyl-peroxide) in the second part. Equal amounts by volume of each pair of solutions were mixed for 30 seconds and used to produce fillings.

Table II. *Composition of the organic phase of the investigated brands*

Mole %	bis-gma	diluent(s)
A	83	17
B	82	18
C	79	21
D	64	36
E	53	47
F	47	53
G	42	58
H	68	32
I	66	34
J	55	45
K	50	50
L	31	69

III. Filler content of experimental resins. A mixture of 75 mole% bis-gma and 25 mole% tedma was divided in two parts, 0.5 % by weight of dppt was dissolved in one part and 3 % by weight of bpo in the other part. Each part was then divided in four parts to which 0, 25, 50 or 75 % by weight of filler was added. The powder of brand F served as filler after the catalyst and silane coupling agent had been eliminated by combustion. Fillings were made from each pair of mixtures with the same filler content. In addition, fillings were made from a similar experimental resin mixture containing 75 % by weight of a silanized filler.

IV. Catalyst content of non-composite, experimental resins. A mixture of 75 mole% bis-gma and 25 mole% tedma

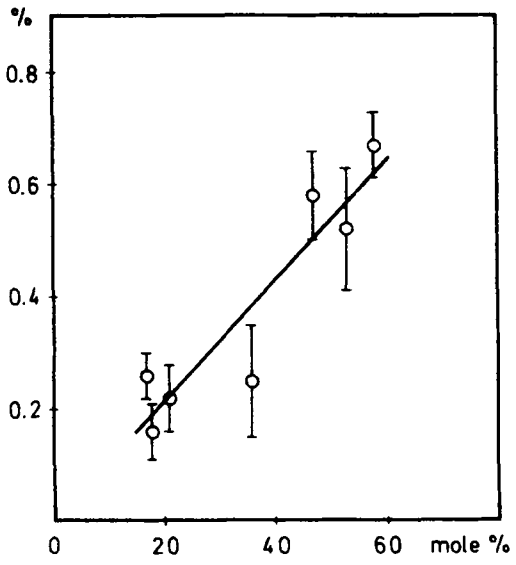

Fig. 1. Gap width in percent of cavity diameter in relation to content of tedma in commercial composite resins.

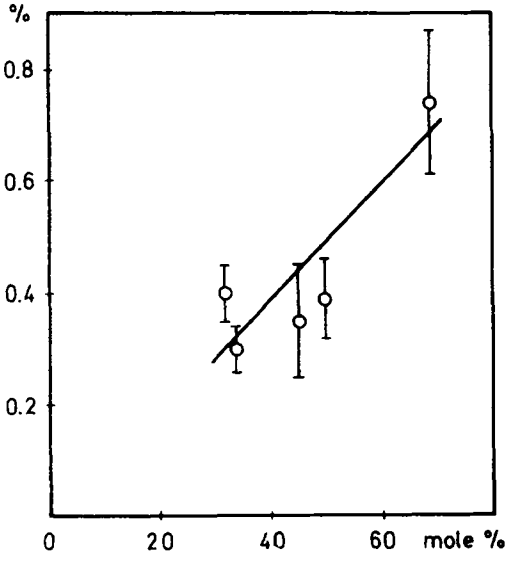

Fig. 2. Gap width in percent of cavity diameter in relation to content of diluting monomers (different from tedma) in commercial composite resins.

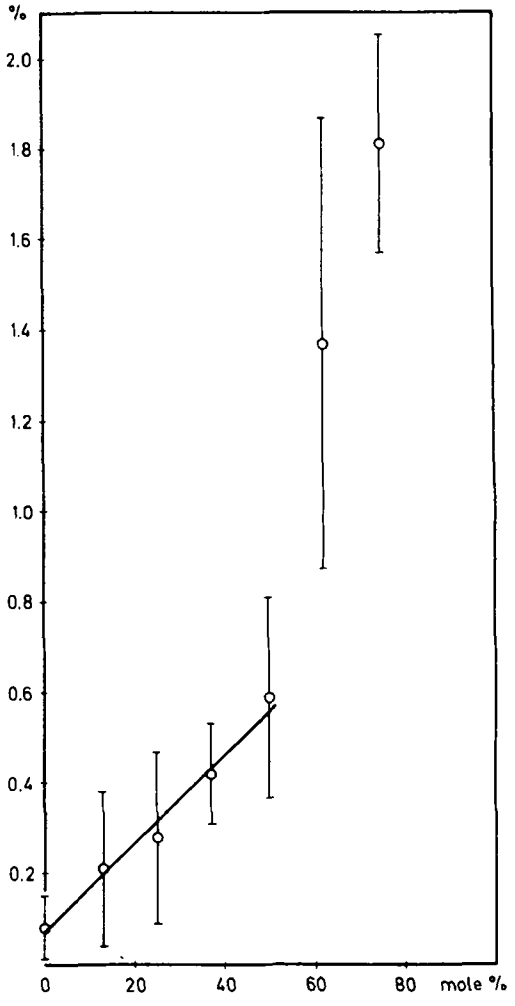

Fig. 3. Gap width in percent of cavity diameter in relation to content of tedma in experimental, non-composite resins.

was divided in four parts. Each part was then divided in two, forming a pair. In the one part of the four pairs 0.375, 0.5, 0.75 or 1 % by weight of dppt was dissolved, in the other part of the pairs 2.25, 3, 4.5 or 6 % by weight of bpo. Fillings were made from each pair of the resins.

RESULTS

The results are presented in Figs. 1—6.

Section I and II. Figs. 1—4 show the relationship between the diluent content in the organic phase (abscissa) and the mean value of the measured gap width (ordinate). The standard deviations are represented by the vertical lines. In Fig. 1

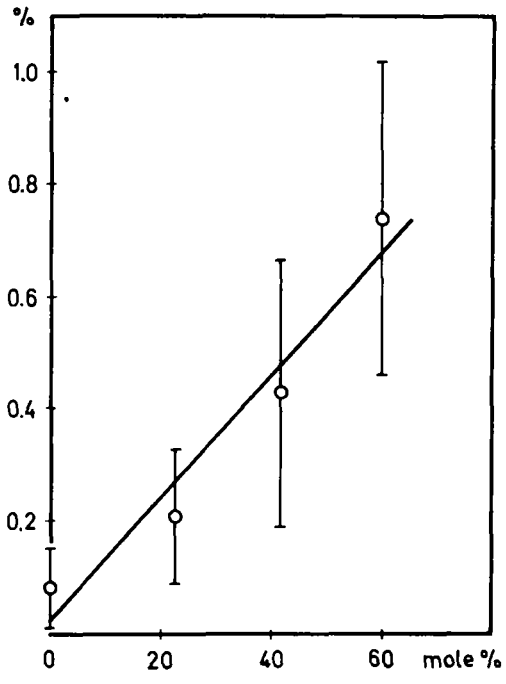

Fig. 4. Gap width in percent of cavity diameter in relation to content of edma in experimental, non-composite resins.

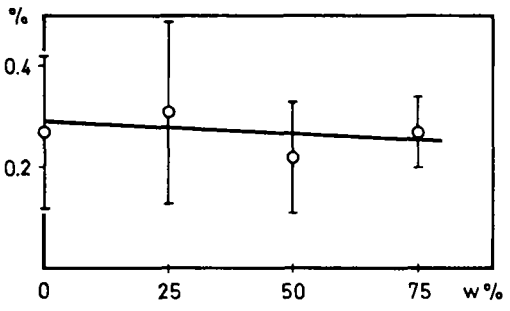

Fig. 5. Gap width in percent of cavity diameter in relation to amount of filler added to an experimental resin composed of 75 mole% bis-gma and 25 mole% tedma.

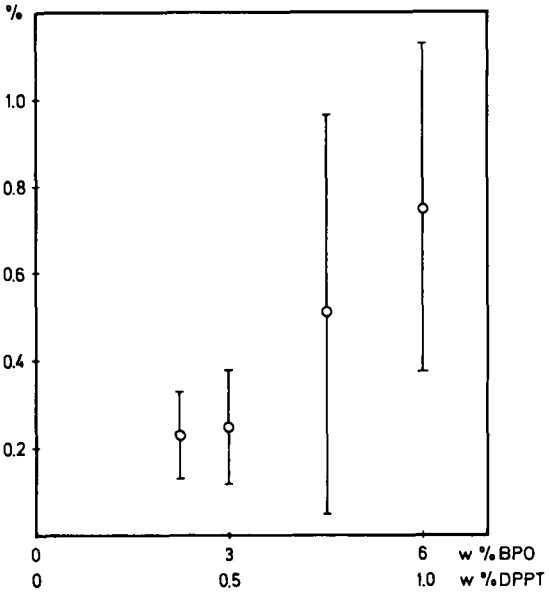

Fig. 6. Gap widt in percent of cavity diameter in relation to amount of catalyst added to an experimental, non-composite resin composed of 75 mole% bis-gma and 25 mole% tedma.

the abscissa gives the content of tedma and in Fig. 2 the content of other diluting monomers in the commercial composite resins. In Figs. 3 and 4 the abscissas give the content of tedma and edma, respectively, of the non-composite, experimental resins. Assuming a straight-line relationship between the gap width and the content of diluting monomers (in Fig. 3 this was assumed only in the interval 0 to 50 mole% tedma), regression analyses were performed. The slope of the regression lines in Figs. 1—4 differed from zero at the $p = 0.001$ level; the differences between the regression lines were not statistically significant at the $p = 0.05$ level. The coefficients of correlation in Figs. 1—4 were 0.87, 0.80, 0.76 and 0.80 respectively.

Section III. Fig. 5 shows the relationship between the content of non-silanized filler (abscissa) and the mean of the gap width (ordinate). Assuming a straight-line relationship between the variables a regression analysis showed that the slope of the regression line did not differ significantly from zero. The coefficient of correlation was —0.10. The resin with 75 % silanized filler gave a gap width identical with the one found for the resin with 75 % non-silanized filler.

Section IV. Fig. 6 shows the relationship between the amount of catalysts (abscissa) and the mean of the gap width. The difference between the gap width obtained with 3 % bpo and 6 % bpo was statistically significant at the $p = 0.025$ level.

DISCUSSION

The foregoing has shown that the wall-to-wall polymerization contraction of restorative resins is related to the composition of the materials. A complete understanding of the demonstrated relations is difficult to obtain, since the influence of the investigated factors on the basic variables determining the size of the wall-to-wall contraction is only known in part. As proposed by *Jørgensen* the polymerization contraction of a restorative resin in a cavity can be discussed on the basis of the model shown in Fig. 7. A represents adhesive forces between resin and cavity walls, and P contraction forces due to polymerization. It is not until P becomes larger than A that a gap will be formed between filling and cavity walls. As long as P is smaller than A, the polymerization shrinkage will appear as a sinking in of the free surface of the filling. The longer the period of time in which P is smaller than A, the larger is the part of the total contraction that has taken place before a gap is formed, and the smaller the gap will be. A high volumetric contraction tends to increase P, whereas the flow of material from the surface of the filling (thin arrows in Fig. 7) tends to reduce P. The amount of material that flows into the cavity is large when the viscosity during polymerization is low and when the polymerization period is long.

In Section I and II a positive correlation between gap width and the content of diluting monomer was demonstrated. A

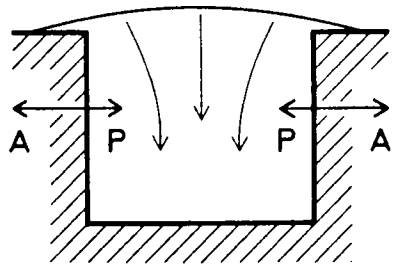

Fig. 7. Schematic presentation of a restorative resin polymerizing in a cavity. P represents contraction forces due to polymerization, and A adhesive forces between filling and cavity walls. The thin arrows indicate the flow of material from the free surface of the filling during polymerization (Courtesy of K. D. Jørgensen).

complete explanation of this relationship must be based on all the above mentioned properties that are involved in the gap-formation process, but only the influence of the diluent content on viscosity and volumetric contraction is known (qualitatively). Assuming a constant filler content the higher the content of diluting monomer the lower is the viscosity and the higher is the volumetric contraction. The latter is due to the fact that the diluent molecules are smaller than those of the bis-gma. As mentioned above a low viscosity tends to reduce the gap widths while a high volumetric contraction tends to increase the gap widths.

The experimental findings show that the influence of the increase in volumetric contraction and the unknown changes in adhesion and duration of polymerization time dominates the influence of the reduction in viscosity, so that the gap widths increase with the content of diluent. However, it must be emphasized that a large volumetric contraction does not necessarily lead to a large wall-to-wall contraction for all types of restorative resins. This was demonstrated by *Asmussen & Jørgensen* (1972) with the unfilled Sevriton

Simplified ®, the main component of which is the comparatively low molecular weight methyl-methacrylate monomer (*Asmussen*, 1975).

It is interesting to note that there is no statistical difference between the regression lines in Figs. 1—4. This suggests that the amount of filler has no influence on the wall-to-wall contraction when the content of diluent is low (<50 mole%). That this is indeed the case was demonstrated in section III (Fig. 5), using a resin containing 25 mole% of tedma. As above, to understand this finding, the influence of the filler content on the basic variables has to be analyzed. When filler is added to a non-composite resin, the volumetric contraction is reduced and the viscosity is increased, whereas the adhesion and the duration of the polymerization period may be assumed to be unaffected. The changes in volumetric contraction and viscosity tend, respectively, to reduce and increase the wall-to-wall polymerization contraction. The results show that the influence of these two variables cancels each other with the consequence that the gap widths are independent of the filler content.

In section IV the influence of the catalyst content was investigated. The effect of the catalyst on the viscosity and the adhesion is not known, but it may be assumed that the degree of polymerization (and hence the volumetric contraction) is increased and the duration of the polymerization period is reduced when the concentration of catalyst is raised. These changes in volumetric contraction and duration of polymerization period both tend to increase the wall-to-wall polymerization contraction and offer (in part) an explanation of the observed variation of gap width with content of catalyst.

To summarize, it was demonstrated that the composition of the organic phase is of primary importance for the wall-to-wall polymerization contraction.

Fillers are included in restorative resins, among other things, to lower the polymerization shrinkage and to reduce the coefficient of thermal expansion (*Bowen*, 1963). The present work demonstrated, however, that the wall-to-wall contraction was not affected by the filler content, and it is probable that the coefficient of thermal expansion for a number of brands of restorative resins is of only minor importance. Fillings of brands with moderate wall-to-wall contraction and a water absorption expansion sufficiently large to compensate for the contraction become expanded against the cavity walls. In such fillings elastic strain counteracts the formation of marginal gaps during cooling, thus reducing the risk of »thermal percolation». An unfilled resin will have a higher hygroscopic expansion than the same resin containing a filler. Since the wall-to-wall contraction is independent of the filler content, a non-composite resin filling will become more hygroscopically expanded against the cavity walls than the composite filling. Accordingly, it is probable that the risk of percolation caused by thermal fluctuations may be equally low with a non-composite and a composite restorative resin, as indicated by *in vitro* investigations (*Asmussen*, 1974).

Thus, it seems that the fillers in composite restorative resins are of importance mainly as reinforcing agents (*Bowen*, 1963), and to a lesser degree, if at all, in securing a good marginal adaptation.

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