Characteristics and properties of composite pastes based on novel Bis–GMA analogues

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Summary

The relationship between filler type, filler content, and matrix resin composition and viscosity, and the flow characteristics of composite formulations were investigated. The structures of three newly synthesized monomers, i.e., 3F Bis–GMA, 6F Bis–GMA (Bis–GMA analogues) and IP AGDM (based on phthalic acid) were analyzed to determine their viscosity and polymerization shrinkage. Viscosities were determined with a cone and plate viscometer and shrinkages were determined gravimetrically. All three experimental monomers required less dilution than Bis–GMA, and they exhibited remarkable reductions in polymerization shrinkage relative to Bis–GMA. Bis–GMA and 4 experimental Bis–GMA analogues were also evaluated. For each monomer–filler combination, increasing filler contents resulted in monotonic plasticities. The maximum filler contents appeared to be an attribute of the particular filler rather than a matrix resin characteristic. Plasticity differences existed, unexpectedly, even at constant filler contents and matrix viscosities. Evidence was found that the plasticity varied with the base monomer composition and the amount of TEGDMA present.

Introduction

Our main thrust has been for the last several years on the investigation of various aspects of the relationships between the composition, structure and properties of dental composite matrix and the composites. We have been actively engaged in the studies dealing with the relationships between filler type, filler content, matrix resin composition and viscosity and the flow characteristics.

It is generally assumed that the more filler incorporated, the greater the effect on these properties up to a limit at which further additions result in a frangible, non–coherent mix. It is also assumed that the lower the viscosity of the uncured matrix the more filler can be incorporated for a given

paste consistency, with the property benefits noted above. This belief appears largely based on the behavior of 2, 2-bis (4-(2-hydroxy-3 Methacryloxy propyloxy) phenyl) propane (Bis-GMA) ; Fig.1) where the viscosity changes produced by dilution with Triethylene Glycol Dimethacrylate (TEG-DMA ; Fig.5) or Hydroxy Ethyl Methacrylate (HEMA) are great. The practical limit to the benefits of dilution is normally set by the lower strength, higher shrinkage and greater water sorption accompanying dilution. Because of the availability of a series of previously characterized monomers, we have been able to investigate the interaction of a number of these composition variables on the properties of composites. We have specifically investigated matrix composition, matrix viscosity, filler particle size, and filler content.

Dental composite filler materials are two-phase materials in which a particulate inorganic material is dispersed in a polymeric organic matrix. The addition of the inorganic filler results in a reduction of the curing shrinkage, thermal expansion coefficient, water sorption of the matrix material and an increase in the strength, hardness, and wear resistance to the cured composite. The influence of fillers of various fractions on mechanical properties of cured composites has been studied in order to test conformity with theoretical predictions that these properties of cured composites should increase monotonically as the volume fraction of the filler is increased. Properties evaluated by various authors which have been found to agree with this expectation include indentation hardness, compressive strength, yield strength and fatigue limit[1-5].

This manuscript primarily deals with those variables which determine the plasticity or viscosity of uncured composite pastes. These factors play an important role in determine the clinical handling characteristics of these materials and their adaptation to cavity walls. Among the several factors, matrix resin composition and viscosity, filler content and filler particle size are generally accepted one a qualitative sense as influence on the properties of the cured composite materials as well. In addition this paper also deals with the determination of glass transition temperatures (Tg) of these experimental low viscosity, hydrophobic monomers and unfilled polymers in dry and wet condition together with water uptake measurements of the unfilled polymers. Tg and viscosity values of these monomers will determine the extent to which they polymerized. Percent water uptake values are indicative of the hydrophobic or hydrophilic character of a given polymer although contact angle measurements provide a reasonable estimate of this property.

The objective of the present study is to establish structure-property relationship among novel dental composite matrix resins and to study the relationship between a filler type, filler content, matrix resin composition and the flow characteristics of composite paste formulations.

Materials and methods

Materials

The monomers used in the study are Bis-GMA, Tetra Ethylene Glycol Dimethacrylate (TEG-DMA) and four experimental structurally analogous monomers synthesized previously in our program and characterized by 1H nuclear magnetic resonance imaging (NMR) and Fourier transform infrared (FTIR) spectroscopic techniques[6,7]. The molecular structures of each of these are shown in Fig.1. The former two monomers are obtained from Polysciences Inc., and Aldrich Chemical Co., and used as-received.

Composite pastes were formulated from each of these monomer dilutions using two types of filler, one of which was a 'hybrid' filler, and the other a 'micro' filler. Both were silanated before use. The
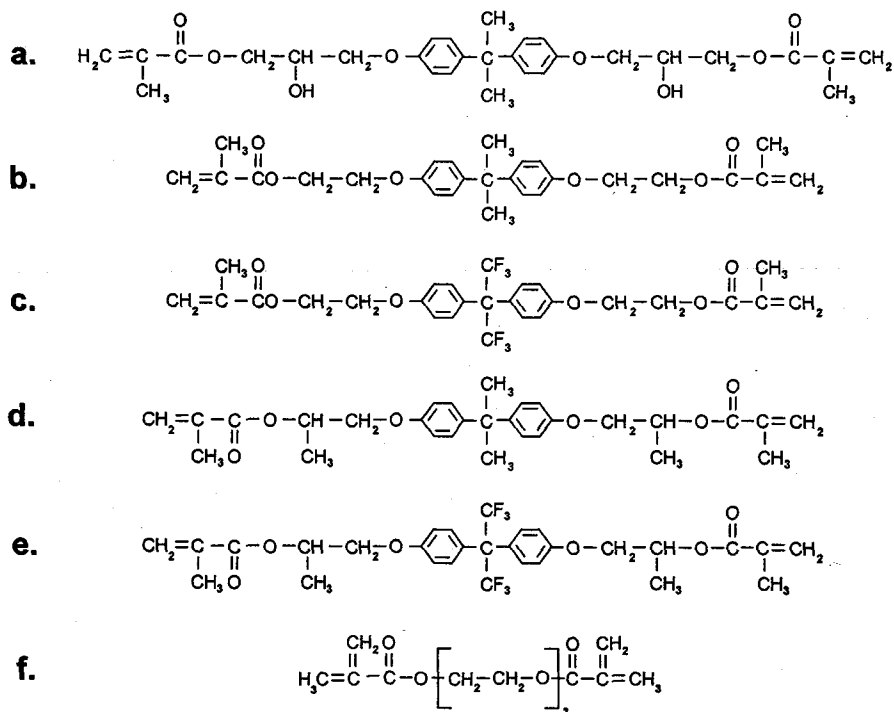
a. $H_2C=C-\overset{\overset{\displaystyle O}{\|}}{C}-O-CH_2-\underset{\underset{\displaystyle OH}{|}}{CH}-CH_2-O-\langle\!\!\!\bigcirc\!\!\!\rangle-\underset{\underset{\displaystyle CH_3}{|}}{\overset{\overset{\displaystyle CH_3}{|}}{C}}-\langle\!\!\!\bigcirc\!\!\!\rangle-O-CH_2-\underset{\underset{\displaystyle OH}{|}}{CH}-CH_2-O-\overset{\overset{\displaystyle O}{\|}}{C}-\underset{\underset{\displaystyle CH_3}{|}}{C}=CH_2$

b. $CH_2=\underset{\underset{\displaystyle CH_3}{|}}{C}-\overset{\overset{\displaystyle O}{\|}}{C}O-CH_2-CH_2-O-\langle\!\!\!\bigcirc\!\!\!\rangle-\underset{\underset{\displaystyle CH_3}{|}}{\overset{\overset{\displaystyle CH_3}{|}}{C}}-\langle\!\!\!\bigcirc\!\!\!\rangle-O-CH_2-CH_2-O-\overset{\overset{\displaystyle O}{\|}}{C}-\underset{\underset{\displaystyle CH_3}{}}{C}=CH_2$

c. $CH_2=\underset{\underset{\displaystyle CH_3}{|}}{C}-\overset{\overset{\displaystyle O}{\|}}{C}O-CH_2-CH_2-O-\langle\!\!\!\bigcirc\!\!\!\rangle-\underset{\underset{\displaystyle CF_3}{|}}{\overset{\overset{\displaystyle CF_3}{|}}{C}}-\langle\!\!\!\bigcirc\!\!\!\rangle-O-CH_2-CH_2-O-\overset{\overset{\displaystyle O}{\|}}{C}-\underset{\underset{\displaystyle CH_3}{}}{C}=CH_2$

d. $CH_2=\underset{\underset{\displaystyle CH_3 \; O}{|}}{C}-C-O-\underset{\underset{\displaystyle CH_3}{|}}{CH}-CH_2-O-\langle\!\!\!\bigcirc\!\!\!\rangle-\underset{\underset{\displaystyle CH_3}{|}}{\overset{\overset{\displaystyle CH_3}{|}}{C}}-\langle\!\!\!\bigcirc\!\!\!\rangle-O-CH_2-\underset{\underset{\displaystyle CH_3}{|}}{CH}-O-\overset{\overset{\displaystyle O}{\|}}{C}-\underset{\underset{\displaystyle CH_3}{|}}{C}=CH_2$

e. $CH_2=\underset{\underset{\displaystyle CH_3 \; O}{|}}{C}-C-O-\underset{\underset{\displaystyle CH_3}{|}}{CH}-CH_2-O-\langle\!\!\!\bigcirc\!\!\!\rangle-\underset{\underset{\displaystyle CF_3}{|}}{\overset{\overset{\displaystyle CF_3}{|}}{C}}-\langle\!\!\!\bigcirc\!\!\!\rangle-O-CH_2-\underset{\underset{\displaystyle CH_3}{|}}{CH}-O-\overset{\overset{\displaystyle O}{\|}}{C}-\underset{\underset{\displaystyle CH_3}{|}}{C}=CH_2$

f. $H_3C=\overset{\overset{\displaystyle CH_2}{\|}}{C}-\overset{\overset{\displaystyle O}{\|}}{C}-O-\Big[CH_2-CH_2-O\Big]_3\overset{\overset{\displaystyle O}{\|}}{C}-\overset{\overset{\displaystyle CH_2}{\|}}{C}=CH_3$

Fig.1 a : 2, 2–bis (4–(2–hydroxy–3–methacryloxyprop–1–oxy) phenylpropane (Bis–GMA)
   b : ethoxylated Bis–GMA (Bis–GMA (H))
   c : 6F ethoxylated Bis–GMA (F Bis–GMA (H))
   d : propoxylated Bis–GMA (Bis–GMA (CH₃))
   e : 6F proxylated Bis–GMA (Bis–GMA (CH₃))
   f : triethylene glycol dimethacrylated (TEGDMA)

'hybrid' filler was obtained from 3M and had an average particle size of 0.76 micron meters but with 40% by weight exceeding one micron in diameter. Details of the particle size distribution are given in Table 1. This filler did not have a bimodal size distribution and appears not to have been the result of blending fillers of differing particle size, but the particle sizes extend from multi–micron to sub–micron diameters characteristic of true blended hybrids. It is referred to here as 'hybrid' only for convenience and brevity in distinguishing it from the other, sub–micron, filler employed.

 The 'micro–filler' was obtained from Corning Glass Co., and was nominally 100%<0.5 micro–meters particle size SEM (Scanning Electron Microscope) examination revealed that the individual particles of this filler were thin platelets which had clumped together into aggregation as large as 1 micron meters in diameter, apparently as a result of the silanization process. It is assumed that

Table 1 : Particle size distribution for 3 MZ 100 hybrid filler

1.	Wt% less than 2 μm	87.2
2.	Wt% less than 1 μm	60.1
3.	Wt% less than 0.3 μm	24.1
4.	Wt% less than 0.1 μm	7.7
5.	Average particle size	0.76

some re-dispersion of the original particles occurred during incorporation into the resin matrix, but the extent and uniformity of that dispersion is not known.

Synthesis

Other properties and characteristics of these experimental monomers have been described in previous publications[6-11]. Structures of Bis-GMA, four newly synthesized experimental monomers and TEGDMA were shown in Fig.1. The air trapped during this mixing process was removed under reduced pressure.

Monomers viscosity adjustment was made using diluent comonomer such as TEGDMA. The viscosity of the matrix-forming monomers also a controlling effect on the amount of filler that can be mixed into a composite, and thus on the properties of the resultant material. Therefore the properties of Bis-GMA and the one experimental monomer (Bis-GMA (H)) were compared at equal viscosity.

A preliminary investigation indicated that viscosity of the uncured matrix of commercial dental composites was in the range of 1-2 Pas.

Bis-GMA and TEGDM were mixed in various proportions up to about 35 wt% of TEGDM (Table 1). Similarly, mixture of Bis-GMA (H) and TEGDM, up to about 28% were prepared. The amount of TEGDM required to produce useable viscosities of 1-2 Pas were determined from viscosity versus dilution curves and shown in Fig.2.

Fig.2 : Variation of viscosities of Bis-GMA and Bis-GMA (H) with content of TEGDMA (viscosity values are in centipoises ; $1CP = 10^{-3}$ Pas)

Methods

Viscosities of the monomers and the comonomer mixtures was measured by means of a Brookfield Cone-and-Plate viscometer at ambient temperature. Two additional customized tests were developed for characterization of the monomer paste by modification of common dental test methods.

In order to determine the maximum useful filler content, a test was developed resembling the Eames setting time test for dental amalgam[12]. Starting with a weighted amount of monomer, small amounts of filler powder were added by hand mixing with a nylon spatula until a thick paste was formed. The paste was then tested by division into two portions which were then recombined. This test was repeated after each addition until the mix no longer coalesced into a single smooth mass when recombined. The weight percent of filler in the last mix which formed a coherent mass was taken as the maximum filler content. For hybrid fillers this is presumably equivalent to a mass in which the volume of the liquid phase is just sufficient to fill the spaces between filler particles. Practically it corresponds to the direct mix capable of forming continuous, defect free, restorations.

Formulations

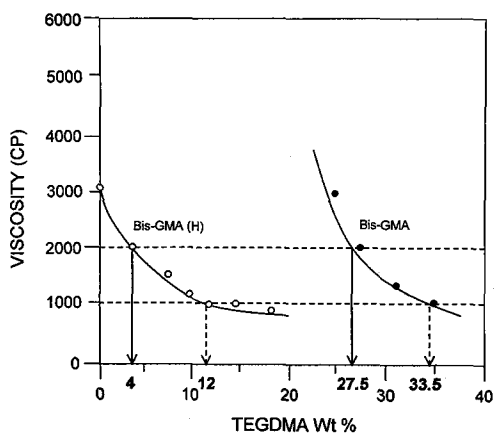Each of the monomers was diluted as needed to produce 1,000 and 2,000 cp solution. Because of

the inherently low viscosity of the synthesized base monomers, this provided seven 2,000 cp and nine 1,000 cp monomer dilutions and two low viscosity (<1,000 cp) undiluted matrix resins. These monomers were then combined with the hybrid and micro–fillers to create sets of composite pastes with varied filler levels. Each monomer–filler combination was first evaluated for maximum filler content, and then subsequently for plasticity at two standard filler contents. In addition, a more extended series of Bis (OH)–TEGDMA dilutions were tested to provide a detailed supplemental evaluation of the effect of matrix viscosity on plasticity. This composition was selected for this purpose because of a perceived relevance to commercial formulations. These monomer blends were also tested for maximum filler content and corresponding plasticity.

Determination of Paste Plasticity

It was expected that the plasticity of composite pastes containing less than the maximum filler content would increase as the filler content was reduced. The extent of this plasticity was measured by a modification of the consistency test first developed for silicate cement[15]. A measured volume of composite paste was placed in the center of a 5 cm x 5 cm slide glass, and covered with a second glass. A constant load was then applied centered over the sample, and maintained for 20 sec. For the purpose of measuring composite paste plasticities the volume of material tested was adjusted to 0.2 ml, and the load was adjusted to 5,000 gm. These values produced disk diameters of approximately 30 mm for commercial composites.

Characterization

The monomers were polymerized in steel moulds, sandwiched between two glass plates. Camphorquinone (0.25 wt%) was used as the photo–initiator and N, N'–dimethyl p–toluidine (0.3 wt%) was used a Triad (Dentsply model 2,000) photocuring unit. Water uptake are described in detail in a previous publication[7].

The glass transition temperatures (Tgs) of the monomers were measured using a Du Pont DSC (Model 1090) instrument, and those of the cured polymers were measured using a Perkin Elmer DMA Instrument (Model DMA–7) in three–point bending mode. The samples were heated at a rate of 2°C min^{-1} and tested at a frequency of 1 Hz.

The glass transition temperatures (Tgs) values of the monomers and monomer mixtures were determined using DSC (TA Instruments Model 2100) with a heating rate of 10 degree C/min using liquid nitrogen as coolant. The Tg values were determined from the onset of the heat capacity change and they are highly reproducible.

The extent of polymerization was determined by measuring the area of polymerization exothermicity using the following modified relationship developed by Miyazaki and Horibe[13,14].

$$EP\,(\%) = \frac{100\,\triangle H}{2\,Hm}\left(\frac{X_1}{M_1} + \frac{X_2}{M_2}\right)$$

where $\triangle H$ (kcal/mol) is the heat of polymerization of the mixture obtained from the area under the curve of the thermogram[9]. Hm is the heat of polymerization of the methyl methacrylate(13.1 kcal/mol).The factor 2 arises from the dimethacrylate reaction. The X_1 and M_1 are the weight fraction and molecular weights of comonomer 1 and comonomer 2,respectively.

Results

Values of glass transition temperatures (Tg) of monomers and their corresponding polymers (in wet and dry conditions), monomer viscosities, percent extent of polymerization (Ep) and water up-take were summarized in Table 2. Water uptake by the polymers was determined gravimetrically. The method was described in detail in a previous publication[6].

Table 2 : Some properties of experimental monomers and the corresponding monomers

Monomer	Tg (°C)	Viscosity (Pa s)*	%Ep**	Polymer		
				Tg (°C) Dry	Tg (°C) Wet	%water uptake
Bis GMA	−6.0	1200	36.1	64.5	57.6	3.05
Bis GMA (H)	−23.3	3.0	47.0	66.7	63.6	0.58
Bis GMA (CH₃)	−26.6	0.3	51.8	63.0	61.3	0.91
6 F BisGMA (H)	−17.5	1.5	50.6	67.2	66.1	0.14
6 F BisGMA (CH₃)	−25.0	0.8	53.6	63.8	61.5	0.18

* : At room temperature
** : Extent of polymerization

Several notable characteristics were observed in our earlier study with reference to the effect of the dilution of Bis−GMA with TEGDMA upon the viscosity of the monomer solutions associated maximum filler ("hybrid" and "micro") contents and plasticities. As expected increasing amounts of TEGDMA result in a decrease in the viscosity of the matrix resin. When the fillers were added very different results were obtained with the two fillings. Much higher amounts of the hybrid filler (~85 Wt%) can be incorporated in the resin than of the micro filler (~35 Wt%). In both cases, the maximum filler contents show little or no variation with matrix viscosity. The plasticity results for the two fillers again appeared to differ remarkably with disc diameter for the "micro" filled composite paste suggesting essentially independent of the matrix viscosity, while the "hybrid" pasted showed a consistent increase in plasticity as the matrix viscosity decreased[16].

The plasticity results in the resin formulation have been determined and tabulated as a function of matrix composition, matrix viscosity, filler type and filler content.

The plasticity results for each of the remaining resin formulations have been determined and tabulated as a function of matrix composition, matrix viscosity, filler type, and filler content. These results are summarized in Tables 3–6. Table 3 summarizes the data for variation of composite paste plasticity with filler content in 2000 cp–hybrid paste with reference to ethoxylated and propoxylated monomers namely Bis–GMA (OH), Bis–GMA (H), 6F Bis–GMA (OH), 6F Bis–GMA (H). Similar results were shown in Table 4 for the variation of composite paste plasticity with filler content in 2000 cp–micro pastes. Tables 5 and 6 show the results for variation of composite paste plasticity with filler content in 1000 cp–hybrid pastes and with reference to the filler content in 1000 cp–micro pastes respectively for the ethoxylated and propoxylated monomers.

Table 3 : Variation of composite paste plasticity with filler content in 2000 cp hybrid pastes

Monomer	Filler(Wt%)	Diameter (D) in cms
	81.2	2.4
Bis–GMA (OH)	75.0	3.0
	70.0	3.4
	78.9	2.0
Bis–GMA (H)	75.0	2.4
	70.0	3.0
	82.0	2.5
6F Bis–GMA (OH)	75.0	3.3
	70.0	3.8
	81.9	1.8
6F Bis–GMA (H)	75.0	2.6
	70.0	3.3

Table 4 : Variation of composite paste plasticity with filler content in 2000 cp–micro pastes

Monomer	Filler(Wt%)	Diameter (D) in cms
	34.0	1.53
Bis–GMA (OH)	30.0	1.81
	25.0	2.15
	33.1	1.50
Bis–GMA (H)	30.1	1.70
	25.1	2.00
	33.7	1.43
6F Bis–GMA (OH)	29.9	1.57
	24.8	1.20
	31.6	1.60
6F Bis–GMA (H)	30.0	1.71
	25.2	1.91

Table 5 : Variation of composite paste plasticity with filler content in 1000 cp hybrid pastes

Monomer	Filler(Wt%)	Diameter (D) in cms
	86.6	2.70
Bis–GMA (OH)	80.0	3.98
	75.0	5.80
	81.4	2.00
Bis–GMA (H)	80.0	2.20
	75.0	2.80
	70.0	3.30
	84.4	2.50
6F Bis–GMA (OH)	80.0	2.88
	75.0	4.70
	81.4	1.70
6F Bis–GMA (H)	80.0	2.88
	75.0	2.50
	70.0	3.15

Table 6 : Variation of composite plasticity with filler content in 1000 cp–micro pastes

Monomer	Filler(Wt%)	Diameter (D) in cms
	34.0	1.90
Bis–GMA (OH)	30.0	2.24
	25.0	2.68
	31.0	1.70
Bis–GMA (H)	30.0	1.88
	25.0	2.16
	20.0	2.45
	37.4	1.50
6F Bis–GMA (OH)	35.0	1.55
	30.0	1.70
	25.0	2.40
	36.2	1.62
6F Bis–GMA (H)	30.0	1.81
	25.0	2.06

Curves of plasticity filler for 1000 cp ethoxylated monomers with micro–filler Bis–GMA (H), 6F Bis–GMA (H) were shown in Fig.3. Similar curves for 1000 cp with hybrid micro–filler Bis–GMA (CH₃), 6F Bis–GMA (CH₃) were shown in Fig.4. Figs.5 and 6 represent respectively the plasticity curves for micro–filler and hybrid filler with reference to propoxylated monomers [Bis–GMA (CH₃) and 6F Bis–GMA (CH₃) for 1000 cp.

For ease in comparison and interpretation of the results portion of the information in the tables are also presented in graphic format. In each instance filler content (in wt%) is given as the abcissa and plasticity (measured in disk diameter in centimeters) is given as the ordinate. All of these graphs are for 1000 cp dilutions. The corresponding 2000 cp information is available but omitted for the sake of brevity. Each graph presents the results for a selected relates series of materials.
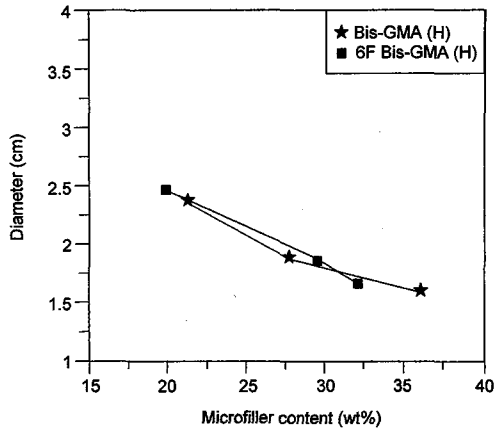
Fig.3 : Curves of plasticity–filler for 1000 cp ethoxylated monomers with microfiller – Bis – GMA (H), 6F Bis–GMA (H)
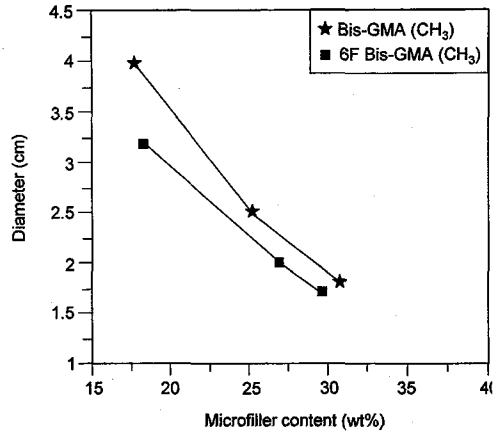

Fig.4 : Curves of plasticity–filler for 1000 cp proxylated monomers with microfiller – Bis – GMA (CH₃), 6F Bis–GMA (CH₃)
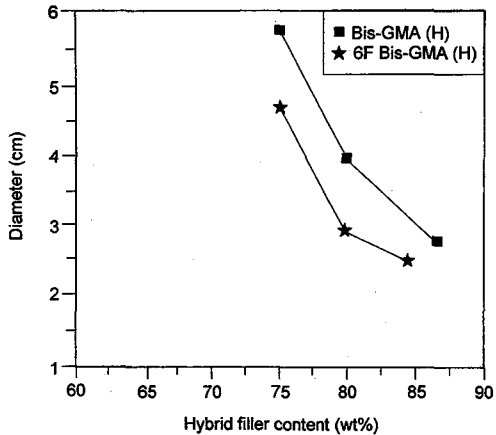

Fig.5 : Curves of plasticity–filler for 1000 cp ethoxylated monomers with hybrid – filler Bis – GMA (H), 6F Bis–GMA (H)
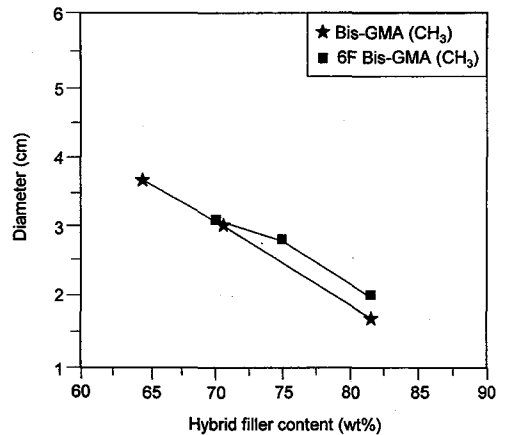

Fig.6 : Curves of plasticity–filler for 1000 cp proxylated monomers with hybrid–filler–Bis – GMA (CH₃), 6F Bis–GMA (CH₃)

Discussion

It may be observed from the Table 2 that among all the monomers investigated, Bis–GMA exhibited the lowest extent of polymerization (conversion). It may also be noted that the Tg and viscosity values of the new monomers are much lower than the Tg and viscosity of the control Bis–GMA (Fig.1a). This suggested that higher conversions should be possible using the less viscous and lower Tg monomers. In other words, the difference in the Ep values of the experimental monomers is probably due to different vitrification/conversion relations. The monomer with a lower viscosity may allow for an enhanced diffusion of reactive groups during the initial vitrificaiton at a higher extent of conversion. Fluorine contacting monomers exhibited higher Tg values than the others. It may be explained as due to the rigidity introduced by the relatively bulky fluorine groups in the 6F monomers.

The Tgs of the dry and water saturated samples were measured using DMA (Table 2). The glass transition temperatures of the dry polymers were very similar [65±2.2℃] indicating that vitrification at the ambient temperature during polymerization of the samples limited the complete conversions. However, the Tgs of the corresponding water saturated samples were found to be strongly influenced by the hydrophilicity of the monomers. For example, Bis–GMA, which takes up 3.05 wt% of water, exhibits a reduction of 7 degrees compared to other systems where the reduction of Tg on saturation with water is the only 3 degrees or less. Similarly among the ethoxylated and propoxylated analogues of Bis–A and 6F polymers, the reduction of Tg due to water sorption is lower and similar. The reduction of the Tg by water sorption may be due to the fact that the absorbed water may act as plasticizers in these systems[17].

It was seen in Fig.2 that ethoxylated Bis–GMA [Bis–GMA (H) required only 4% and 12% of the diluent TEGDMA to adjust the viscosity to 2000 cp and 1000 cp respectively when compared with the amounts of TEGDMA (27.5% and 33.5%) needed to bring down the viscosities to 2000 and 1000 cp of Bis–GMA. In other words, the experimental monomer, Bis–GMA (H) did exhibit lower viscosity than Bis–GMA.

It became rather difficult to predict the behavior of these materials based on limited prior information. Nevertheless, there were certain general expectations. It was assumed that there would be some maximum amount of filler that could be incorporated in a coherent paste, and that there would be differences between the behavior of the mixes made with two fillers. From experience with other materials it was presumed that increasing filler the more finely divided filler would be more effective in increasing viscosity. However, no specific compositional effects were anticipated. To the extent that any behavior was expected, it was assumed that for any given filler and matrix viscosity the plasticity of the pastes would vary in some uniform fashion with filler content independent of resin composition. In the case of the ethoxylated monomers shown in Fig.3, these assumptions seemed reasonably valid. The two lines representing different materials appears to be straight or only slightly curved, and they appear to share a common rate of variation of plasticity with filler content. However, the results for the propoxylated monomers even at the same viscosity and with the same filler, were different as seen in Fig.4. The individual curves in this case represent variations in the core structure of the matrix resins. We have no explanation at this time as to why the identical differences produce no variation in the plasticity of the ethoxylated monomer pastes.

Somewhat similar results were seen for the formulations with hybrid filler (Figs.5 and 6). Once again, the ethoxylated monomer produces relatively straight curves with similar slopes.

It was seen, in addition to the relationship already noted, the data provide insight into other relationships. Among the basic of these is the overall relationship between the matrix resin viscosity and filler content. The primary factor that appears to control the maximum filler content is the nature of the filler. However, at lower filler content and hyper plasticities, considerable differences arise.

It can be seen clearly in Figs.4 and 6, formulations can be prepared which differ considerably in plasticity at constant filler content or filler content at constant plasticities. By holding either of these factors constant, it becomes possible to evaluate some additional effects.

The values for the hybrid formulations conform reasonably well to an assumption of close packing of the filler particles with the matrix resin filling the intervening spaces. The much lower values for the microfiller implied that some additional or alternative mechanism is involved in limiting the filler content.

Conclusions

1. The novel monomer (Bis–GMA analogues) lower viscosity and glass transition temperature with remarkable increase in conversions relative to control Bis–GMA.
2. Fluorine contacting monomers showed less water uptake compared to Bis–GMA.
3. For each matrix filler combination there is a characteristic maximum filler content which is predominantly an effect of the nature of the filler.
4. In order to produce a similar plasticity, less amount of micro filler than hybrid is needed.
5. For all formulations, decreasing the filler content resulted in increasing plasticity.

Acknowledgment

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抄録：新しい Bis-GMA 類似体を基調としたコンポジットレジンペーストの性質と特徴

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　歯科用コンポジットレジンのモノマーについて，フィラータイプ，フィラー含有量，マトリックスレジン組成と粘度並びに流れ（フロー）の関係を調査する目的で，今回新しく合成した3種類のモノマー（3F Bis–GMA，6F Bis–GMA（Bis–GMA 類似体），IP AGDM（フタリル酸ベース））について，粘調度と重合収縮を分析した．粘調性はコーン・プレート粘調度測定器を用い，重合収縮は重量測定法で算出した．3種類全ての新規合成実験用モノマーは通常のベースモノマー（Bis–GMA）よりも少ない希釈が必要とされ，通常の Bis–GMA と比較して重合収縮は著しく減少した．また Bis–GMA 単体と4種類の実験用 Bis–GMA 類似体の評価では，モノマーとフィラーの組み合わせにおいてフィラー含有量が増加すると単純に可塑性化した．フィラーを最大限含有したモノマーは個々のフィラーの特質が見られたというよりもむしろマトリックスレジンの特徴が見られた．一定量のフィラーを含有し，マトリックスの粘調性があるにもかかわらず可塑性の差が予期せず現れた．すなわち可塑性はベースモノマーと TEGDMA の量により変化することが明らかとなった．