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ORIGINAL ARTICLE

# Conjugation of diisocyanate side chains to dimethacrylate reduces polymerization shrinkage and increases the hardness of composite resins



Yih-Dean Jan <sup>a</sup>, Bor-Shiunn Lee <sup>b,c</sup>, Chun-Pin Lin <sup>a,b,c,d</sup>,  
Wan-Yu Tseng <sup>c,d,\*</sup>

<sup>a</sup> Graduate Institute of Clinical Dentistry, National Taiwan University, Taipei, Taiwan

<sup>b</sup> Graduate Institute of Oral Biology, National Taiwan University, Taipei, Taiwan

<sup>c</sup> School of Dentistry, National Taiwan University, Taipei, Taiwan

<sup>d</sup> Department of Dentistry, National Taiwan University Hospital, College of Medicine, National Taiwan University, Taipei, Taiwan

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## KEYWORDS

composite resin;  
degree of conversion;  
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urethane-modified  
resin

**Background/Purpose:** Polymerization shrinkage is one of the main causes of dental restoration failure. This study tried to conjugate two diisocyanate side chains to dimethacrylate resins in order to reduce polymerization shrinkage and increase the hardness of composite resins.

**Methods:** Diisocyanate, 2-hydroxyethyl methacrylate, and bisphenol A dimethacrylate were reacted in different ratios to form urethane-modified new resin matrices, and then mixed with 50 wt.% silica fillers. The viscosities of matrices, polymerization shrinkage, surface hardness, and degrees of conversion of experimental composite resins were then evaluated and compared with a nonmodified control group.

**Results:** The viscosities of resin matrices increased with increasing diisocyanate side chain density. Polymerization shrinkage and degree of conversion, however, decreased with increasing diisocyanate side chain density. The surface hardness of all diisocyanate-modified groups was equal to or significantly higher than that of the control group.

**Conclusion:** Conjugation of diisocyanate side chains to dimethacrylate represents an effective means of reducing polymerization shrinkage and increasing the surface hardness of dental composite resins.

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\* Corresponding author. 1, Chang-Te Street, Taipei 10048, Taiwan.

E-mail address: [yeshes@gmail.com](mailto:yeshes@gmail.com) (W.-Y. Tseng).

## Introduction

Composite resins are currently the most popular dental restorative materials worldwide. Composite resins provide certain advantages such as good esthetics, easy application, and lower costs. However, there remain some disadvantages to their use, such as polymerization shrinkage, low wear resistance, and marginal discoloration. Composite resins are composed of organic monomers and inorganic fillers. High molecular weight dimethacrylate monomers with low polymerization shrinkage and high strength, such as bisphenol A-glycidyl dimethacrylate (bis-GMA), are most commonly used. The high viscosity of bis-GMA reduces the loading of fillers and also the degree of conversion of the monomers in the absence of other low viscosity diluents.<sup>1</sup> Low molecular weight diluent monomers, such as triethylene glycol dimethacrylate, are often added to reduce viscosity and increase the reactivity and conversion rate.<sup>1</sup> However, the diluent monomers also increase polymerization shrinkage,<sup>2</sup> leading to polymerization stress, debonding at the restoration–tooth interface, secondary caries, postoperative sensitivity, pulpal irritation, and marginal discoloration.<sup>3</sup>

Polymerization shrinkage is the principal cause of failure of clinical dental composite resin fillings. Reducing this shrinkage, thus, represents one of the most important goals in the development of new matrices for composite resins. Investigators have made several efforts to achieve this goal, such as by adding inorganic inert fillers to reduce the volume of the matrix and, therefore, the polymerization shrinkage.<sup>4</sup> However, the viscosity of the resin matrix can limit filler loading. Jeon et al<sup>5</sup> synthesized a low-viscosity matrix by substituting the alkoxy for hydroxyl groups in bis-GMA to minimize the amounts of diluent monomers and the shrinkage. Jingwei et al<sup>6</sup> developed a new matrix with multibranches or multifunctional groups. However, resin matrices with multibranches or multifunctional groups display high viscosity and a low degree of conversion.

Currently, there remains a lack of “non-shrinkage” composite resins worldwide. In this study, we aimed to develop low-shrinkage composite resins for dental application. As expected, the higher the molecular weight and volume of the monomer, the less extensive the shrinkage when polymerized. Most commercial dental composite resins are composed of bis-GMA or its derivatives. We increased the molecular weight and volume of the dimethacrylate molecule by conjugating functional side chains to the dimethacrylate structure. Urethane, which is a compound of diisocyanate and 2-hydroxyethyl methacrylate (HEMA), is a material suitable for use as a bis-GMA side chain. Polyurethane displays certain advantages, such as low shrinkage, high wear resistance, and good biocompatibility. We selected two diisocyanates with different chemical structures as side chain materials: 4,4′-diisocyanatodicyclohexylmethane (H<sub>12</sub>MDI) and toluene 2,4-diisocyanate (TDI). H<sub>12</sub>MDI contains two aliphatic rings (cyclohexane) linked by a methyl group, whereas TDI contains a toluene moiety. When conjugated to dimethacrylate, these two chemical structures reduced polymerization shrinkage and increased the mechanical strength of the composites. Different structures and numbers of side chains on dimethacrylate provided different results.

The hypothesis tested in this study was that monomers with higher molecular weight and steric hindrance (side chains on dimethacrylates) would display lower polymerization shrinkage and higher mechanical strength. In this study, we conjugated diisocyanate and HEMA side chains, in various proportions, to bisphenol A dimethacrylate (DM) to synthesize urethane-modified dimethacrylate composite resins, with the aim of reducing polymerization shrinkage because of the relatively high molecular weight and cohesive energy density of the urethane group. We also evaluated the effects of different diisocyanate side chain structures and densities on the dimethacrylate resin composites.

## Materials and methods

### Preparation of multifunctional dimethacrylate composite resins

TDI (T, 2 equivalents; Acros Chemical Co., Geel, Belgium) and H<sub>12</sub>MDI (H, 2 equivalents; Acros Chemical Co.) were poured into separate reaction bottles and HEMA (1 equivalent; Acros Chemical Co.) was then added to each bottle dropwise. Dry nitrogen gas was continuously purged during the reaction procedure to remove oxygen and moisture. A Fourier transform infrared (FTIR) spectrometer was used to monitor the peak of the isocyanate group (NCO–). When the peak intensity of NCO– had reached half of its original value, this indicated that the hydroxyl group (OH–) of HEMA had fully reacted with the isocyanate group (NCO–) of TDI or H<sub>12</sub>MDI. Bisphenol A DM (Double Bond Chemical Ind. Co. Ltd., Taipei, Taiwan) was then added to the reaction mixtures as 4, 2, 1, or two-thirds equivalents in the nitrogen atmosphere. When the peak of the isocyanate group had disappeared, this indicated that the NCO– group of TDI or H<sub>12</sub>MDI had fully reacted with the OH– group of dimethacrylate. Their formulae are presented in Figs. 1 and 2.

For the control group, DM was mixed with dimethacrylate and 20 wt.% tripropylene glycol diacrylate (TPGDA; Double Bond Chemical Ind. Co. Ltd.) as a diluent. The urethane-modified dimethacrylate resin matrices were synthesized using a mixture of dimethacrylate, HEMA, and H<sub>12</sub>MDI (or TDI) with molar ratios of 1:0.25:0.25, 1:0.5:0.5, 1:1:1, and 1:1.5:1.5. All experimental groups—DM-H-0.25, DM-H-0.5, DM-H-1.0, DM-H-1.5, DM-T-0.25, DM-T-0.5, DM-T-1.0, and DM-T-1.5, and DM-0 as the control group—are listed in Table 1. The urethane-modified resin matrices were added to a 20 wt.% TPGDA diluent monomer. All groups of the resin matrix were then added to 1 wt.% camphorquinone (Acros Chemical Co.), 0.5 wt.% ethyl 4-dimethylaminobenzoate (Acros Chemical Co.), and 50 wt.% silanized silica fillers (average 1.6 μm; Sibelco Bao Lin Co. Ltd., Taichung, Taiwan), and mixed well using a planetary mixer (Kurabo, Mazerustar, KK-250S, Osaka, Japan) to form the composite resins.

### Viscosity of resin matrices

The viscosity of resin matrices was measured using a viscometer (Brookfield Engineering Laboratories, Middleboro, MA, USA) at 25°C.

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