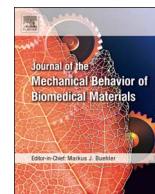




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Synthesis, characterization and evaluation of a fluorinated resin monomer with low water sorption

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ABSTRACT

A fluorinated acrylate monomer (4-TF-PQEA) without BPA (bisphenol-A) structure was synthesized and mixed with triethylene glycol dimethacrylate (TEGDMA) to used as dental resin system in order to achieve lower water sorption and reduce human exposure to BPA derivatives. Double bond conversion (DC) was measured using Fourier transform infrared spectroscopy (FTIR). Water sorption (WS), water solution (WL) and depth of cure (DOC) were evaluated according to ISO 4049:2009. Water contact angle (CA) was measured using contact angle analyzer. Polymerization shrinkage (PS) was evaluated according to the Archimedes' principle and ISO 17304:2013. Flexural strength (FS) and flexural modulus (FM) were measured by three-point bending test with a universal testing machine according to ISO 4049:2009. Comprehensive strength (CS) and vickers microhardness (VM) were also investigated. Thermal stability test was measure by Thermogravimetric analyzer. Cytotoxicity of three resin systems was tested through MTT (3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyltetrazoliumbromid) cytotoxicity method according to the ISO 10993-5:2009. Bisphenol-A glycidyl dimethacrylate (Bis-GMA)/TEGDMA resin system was used as a control. The results show that 4-TF-PQEA/TEGDMA resin system had lower PS, lower WS and higher DC values than those of Bis-GMA/TEGDMA resin system except some mechanical properties, such as FS, FM and CS. Moreover, properties of other 4-TF-PQEA-containing resin systems were also comparable with those of Bis-GMA/TEGDMA resin system. In particular, the overall performance of resin system consisted of 4-TF-PQEA/Bis-GMA/TEGDMA is optimized when the mixture ratio is 30/40/30(wt/wt/wt). Therefore, the 4-TF-PQEA has potential to be used as resin monomer for dental resin composites to achieve lower water sorption.

1. Introduction

Resin composites play a crucial role in dental restoration treatment and have been increasing popular among clinicians and patients on account of their favorable clinical performance, such as superior esthetics, adhesive properties and desirable performance (Da Rosa Rodolpho et al., 2011; Demarco et al., 2012; Mei et al., 2016). Generally, the composition of resin composites is based on methacrylate monomers, for instance, bisphenol A- glycerolate dimethacrylate (Bis-GMA), which has been prevalently used as organic matrix resin since it was synthesized by Bowen in 1965. Owing to the high viscosity of Bis-GMA which limits the practical application, triethylene glycol dimethacrylate (TEGDMA) is thus used as a diluent monomer to improve the clinical operation as well as permit higher filler loading. Even though the Bis-GMA/TEGDMA resin system provides numerous advantages like good mechanical properties and biocompatibility for resin

composites materials in restoration treatment, the defects still remain and therefore lead to the failure of restoration. Higher polymerization shrinkage is one of drawbacks, as it can bring on interfacial bonding microleakage between tooth structure and resin materials when the polymerization stress generates (Sunbul et al., 2016). Meanwhile, the subsequent contraction stress may result in enamel microcracks, post-operative sensitivity and even recurrent caries (Marchesi et al., 2010). Several studies have found that the shrinkage of resin composites depend on many factors, such as the structure of matrix monomer, the introduction of TEGDMA, degree of conversion and filler loading (Catel et al., 2017; Chen et al., 2006; Peutzfeldt and Asmussen, 2004). Therefore, many methods have been proposed to decrease polymerization shrinkage. Amounts of researches have confirmed that polymerization shrinkage is associated with shrinkage stress, thus, one way to reduce that is to decrease the reactive group concentration of monomers (Shah et al., 2017). As evident from previous studies, He J

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et al. developed the monomers with big molecule weight and large molecule volume used as matrix monomer could reduce polymerization shrinkage of resin restoration composites (He et al., 2010, 2012). It is also desirable to achieve low shrinkage by synthesizing a new monomer with low viscosity, such as Hong's work (Hong et al., 2017). Another strategy to endow dental composites with a low shrinkage is to reduce monomers content by the introduction of inorganic fillers in resin composites. Especially, the higher inorganic fillers can improve mechanical properties (Pick et al., 2011; Sideridou et al., 2011).

Lower double bond conversion and higher water sorption are also responsible for the failure of restoration treatment. Polymerization process of light-cured resin composites is a free-radical polymerization reaction as well as it is induced by light-irradiation components, which can result in the generation of reactive free radicals. When the resin composites are cured in the air, the polymerization reaction will be inhibited by oxygen due to the fact that oxygen has greater ability to react with free radicals than monomer molecules (Bijelicdonova et al., 2015; Suh, 2004). As a consequence, the incomplete polymerization process gives rise to the leach of monomers, which can affect the biocompatibility between resin materials and oral tissues. Moreover, it turned out that the leachable monomers were cytotoxic (Jagdish et al., 2009b). In addition, the higher viscosity of resin composites leads to the lower double bond degree due to the weak mobility of monomer molecules and trapped functional groups (Cornelio et al., 2013). Furthermore, it is well known that the utilization of TEGDMA in resin composites will increase the water sorption (Miao et al., 2012). Both the low double bond conversion and water sorption phenomenon show negative effects on the longevity of dental restoration materials. Because when the resin composites are exposed to oral environment, the unreacted leachable monomers will suffer hydrolysis and even the filler bonding agent will be degraded (Gajewski et al., 2012; Gonçalves et al., 2014). Subsequently, the poor mechanical properties accelerate restorations failure. Usually, the amount of water that dental resin composites could absorb depends on the hydrophilicity of polymeric matrix and filler composition (Berger et al., 2009; Sideridou et al., 2003). Aim to those, various attempts have been done in order to improve properties of resin composites, including improving double bond conversion and reducing water sorption by synthesizing new dimethacrylate monomers (Kerby et al., 2009; Yin et al., 2015). Besides, it is reported that fluorine compounds exhibit water and oil repellency, chemical stability and potential resistance to microbial attachment (Kadoma, 2010; Kasuga et al., 2008). It's worth mentioning that fluorinated monomers have been utilized for dental materials with low water sorption (Hoshino et al., 2014; Kadoma and Tamaki, 2009; Yoshida et al., 2013).

Nowadays, people pay more attention on health care, the application of Bis-GMA and its derivatives have been suspected due to bisphenol-A (BPA) core structure of Bis-GMA, which is an endocrine-disrupting chemical (Program 2008; Vandenberg et al., 2012). Thus, more and more non-BPA structure methacrylate monomers were synthesized for dental materials with the aim to minimize human exposure to BPA (Ding et al., 2016; Luo et al., 2016; Yu et al., 2015). Although it's hard to eliminate the use of Bis-GMA for dental restoration materials, it's also necessary to do much effort to reduce the content of Bis-GMA in dental resin composites.

In this research, a fluorinated acrylate monomer 4-TF-PQEA without BPA structure was synthesized and mixed with TEGDMA to prepare dental resin. Both the physicochemical properties and the mechanical properties of 4-TF-PQEA/TEGDMA were evaluated in comparison with those of Bis-GMA/TEGDMA resin system. Meanwhile, the properties of other 4-TF-PQEA-containing resin systems were also investigated. The hypothesis is that 4-TF-PQEA could endow dental resin system with lower water sorption. And the 4-TF-PQEA/TEGDMA resin system has the comparable properties when compared with those of Bis-GMA/TEGDMA resin system.

2. Materials and methods

2.1. Materials

4-Aminobenzotrifluoride was purchased from Energy Chemical Co., St. Shanghai, China. Sodium hydrogen carbonate, sodium nitrite, toluene, acetone, hydrochloric acid, sodium hydroxide, anhydrous ethanol and ethyl acetate were purchased from Beijing Chemical Reagent Co. Ltd., China. Zinc powder and epichlorohydrin were available from Tianjin Guangfu Fine Chemistry Research Institute. Tetrabutyl ammonium bromide was purchased from Huisheng Chemical Reagent Co. Ltd., China. Bis-GMA, TEGDMA, camphorquinone (CQ) and 2-(N, N-dimethylamino)ethyl methacrylate (DMAEMA) were purchased from Sigma-Aldrich Chemical Co., St. Louis, USA. Acrylic acid and hydroquinone were received from Xilong Chemical Reagent Co. Ltd., Guangzhou, China. Triphenylphosphine and 4-benzoquinone were available from Aladdin Industrial Corporation, Shanghai, China. HL-IV curing light was purchased from Hangzhou Zhongrun medical instrument Co., LTD. L-DMEM medium (Gibco, USA), FBS (Gibco, Australia), trypsin (sigma, USA), penicillin-streptomycin (Hyclone, USA), HEPES (Sigma, USA), MTT (Amresco, USA) centrifuge (5810R, Eppendorf, Germany), microplate reader (synergy HT, BioTek, USA) were used as received.

2.2. Synthesis route of monomers

2.2.1. Synthesis of (4-trifluoromethyl) phenylbenzoquinone (4-TF-PBQ)

The mixture of 4-aminobenzotrifluoride (12.7 mL, 0.1 mol) and H₂O (50 mL) were stirred in 500 mL beaker equipped with a thermometer. Next, hydrochloric acid (43 mL, 11.8 M) was added into the solution. Until the temperature of mixture solution was below 5 °C, the concentrated water solution (50 mL) of sodium nitrite (7.25 g, 0.1 mol) was added dropwise into the mixture with a dropping funnel. To keep the temperature below 5 °C, ice was added. After the concentrated water solution of sodium nitrite aqueous solution was added completely, a transparent solution was obtained. After that, the mixture solution was filtered and added dropwise into a mixture of sodium hydrogen carbonate (33.9 g, 0.4 mol), 4-benzoquinone (12.96 g, 1.2 mol) and deionized water (100 mL). It was added completely below 10 °C in 1 h. Then the reaction solution was stirred at 5–10 °C for another 4 h. The resulting product was filtered and then washed with deionized water for 8 times. Finally, the obtained yellow solid product was dried overnight at room temperature. Yield: 80%. Melting point: 106 °C (DSC). *m/z*: 252.

2.2.2. Synthesis of (4-trifluoromethyl) phenylhydroquinone (4-TF-PQ)

The dried yellow product (10 g, 0.01 mol), zinc powder (7.7 g, 0.03 mol) and deionized H₂O (60 mL) were added in a 250 mL three-necked flask equipped with a mechanical stirrer, heating mantle, a thermometer, a condenser and a dropping funnel. When the color of reaction turned purple and the temperature of the mixture was around 96 °C, HCl (10 mL, 11.8 mol/L) was added dropwise into the three-necked flask and then kept the reaction for 4 h. After that, the toluene (60 mL) was added into the mixture for 2 h in order to remove the distilled water. Finally, the hot mixture was filtered and cooled. The yellow crystals were recrystallized from toluene. Yield: 65%. Melting point: 121 °C (DSC). *m/z*: 254.

2.2.3. Synthesis of (4-trifluoromethyl) phenylhydroquinone epoxy (4-TF-PQE)

A 100 mL, three-necked flask equipped with a mechanical stirrer was placed into the oil bath. The product from step 2 (4-TF-PQ) (2.54 g, 0.01 mol) reacted with the epichlorohydrin (784 mL, 0.1 mol) and TBAB (0.0967 g, 0.001 mol) at 90 °C for 8 h. Subsequently, unreacted epichlorohydrin was removed by means of reduced pressure distillation. Then toluene (7.84 mL) and sodium hydroxide solution (3 g, 30 wt %) was added into the three-necked bottle for several times and kept

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