

Synthesis and Characterization of Nanostructured Dental Polymers through Phase Dispersion Control

Q. Ye, J Park, A. Misra, R. Parthasarathy, V. Singh, P. Spencer

¹University of Kansas Bioengineering Research Center, ²University of Kansas Department of Civil Engineering,

³University of Kansas, Department of Mechanical Engineering

Statement of Purpose: The breakdown of the tooth/composite bond has been linked to the failure of current adhesives to provide a durable seal at the interface with dentin. Water is a major interfering factor when bonding adhesives and/or composite to the tooth. Current commercial adhesives undergo phase separation in water. At the interface with the wet demineralized dentin matrix, this phase separation leads to limited infiltration of BisGMA, the critical dimethacrylate component. Phase separation in conjunction with partitioning of the adhesive components inhibits not only the formation of a durable hybrid layer, but suppresses adhesive infiltration throughout the width of the demineralized dentin matrix and the subjacent, intact dentin [1]. Clinical conditions demand a hydrophilic-hydrophobic balance, i.e. hydrophilic resin for initial miscibility with the wet tooth surface but for durability, a hydrophobic material must be formed during and after polymerization. The objective of this research was to use a phase dispersion control technique, e.g., to incorporate reactive dispersing agents (RDA), which enhance miscibility with the wet tooth surface while providing enhanced thermomechanical properties under wet bonding conditions.

Methods: Experimental adhesives containing bisphenol-A diglycidyl ether dimethacrylate (bisGMA, Polysciences, Warrington, PA), 2-hydroxyethyl methacrylate (HEMA, Acros Organics, NJ), and reactive dispersing agent Poly(oxy-1,2-ethanediyl), α -[4-nonyl-2-(1-propenyl) phenyl]- ω -hydroxy-, (Noigen RN, Dai-ichi Kogyo Seiyaku) were photo-polymerized with visible light and compared to control adhesives [HEMA/BisGMA, 45/55 w/w]. Water at concentrations of 5, 10 and 15% was selectively added into these neat resins. The miscibilities of adhesive resins were determined by the concentration of water that could be added before the resin solution became turbid. Turbidity of the mixture was measured using an UV-visible spectrophotometer (Thermo Scientific Evolution 600) at the wavelength of 600 nm. The prepared resins were poured into a glass-tubing mold and light-cured to form rectangular beam specimens (1x1x15 mm³). The degree of conversion (DC) was determined by using a LabRAM ARAMIS Raman spectrometer (LabRAM HORIBA Jobin Yvon, Edison, New Jersey) with a HeNe laser as an excitation source. Dynamic mechanical analysis were performed using DMA Q800 (TA Instruments, New Castle, USA) with a three-point bending clamp. The test temperature was varied from 0 to 200 °C with a ramping rate of 3 °C/min at a frequency of 1 Hz.

Results: The water miscibility of the control adhesive is about 10 wt%. The experimental adhesive containing 10% RDA showed 50% increase of water miscibility although

it has similar hydrophobic/hydrophilic composition compared with the control adhesive. There was little difference in DC of these adhesives cured in the presence of water, but there were substantial differences in the thermal-mechanical properties (Figure 1). The control resins cured in the presence of 10% water are selected to simulate homogeneous adhesives, i.e. based on visual examination these formulations present one solution phase prior to photopolymerization. At 10% water, the storage moduli were statistically significantly greater ($p < 0.05$) for the experimental resin as compared to the control resin. The storage moduli of the experimental resins cured in the presence of 15% water are greater than the control adhesive cured in the presence of 10% water. The 10% water concentration is the miscibility limit for the control adhesive. Storage moduli in the rubbery region were also significantly greater ($p < 0.05$) for the experimental as compared to the control adhesive; these results indicate higher crosslink density with the experimental adhesive.

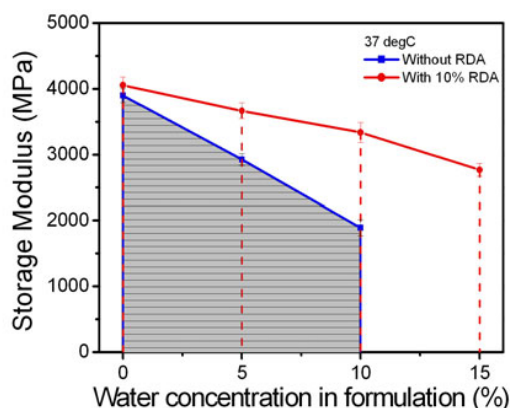


Figure 1. Influence of reactive dispersing agents (RDA) used in dentin adhesive on water miscibility and dynamic mechanical properties.

Conclusions: In this study, we synthesized novel dentin adhesives using reactive dispersing agents to meet the variable polarity requirement of polymeric materials that must provide durable function in the wet, oral environment. The novel nanostructured polymers provide a hydrophilic resin for initial miscibility with the wet tooth surface but during polymerization the material will develop the hydrophobicity required for durability.

References:

1. Spencer P, Wang Y. J Biomed Mater Res 2002 62:447

Acknowledgement: Supported by grant R01DE143923 and R01DE014392-08S109 (Spencer), from the NIDCR/NIH, Bethesda, MD 20892.