Structure/Property of Model Dentin Adhesive Exposed to Wet Environments

P. Spencer[1,2], Q. Ye[1], R. Parthasarathy[1], V. Singh[1,2], A. Misra[1,3], J. S. Laurence[1,4]
[1]University of Kansas Bioengineering Research Center, [2]University of Kansas, Department of Mechanical Engineering
[3]University of Kansas, Department of Civil Engineering, , [4]University of Kansas, Department of Pharmaceutical Chemistry

Statement of Purpose: In the wet, oral environment, the hydrophobic and hydrophilic composition of dentin adhesives can lead to phase separation which compromises the integrity of the adhesive/dentin bond.[1] Due to differences in penetration of hydrophobic and hydrophilic components, and phenomena like diffusion, and convection, various polymer phases are formed by mixing of dentinal fluid with adhesive monomers. The separated phases can be broadly categorized into hydrophobic-rich resin and hydrophilic-rich aqueous phases.[2,3] Characterization and development of structure-property relationships for these phases is important for understanding the mechanical behavior of dentin adhesives under conditions relevant to the wet, oral environment. The objective of this research was to correlate the chemo-mechanical behavior of the phase-separated polymer system with water miscibility and monomer-water phase compositions for the model dentin adhesive.

Methods: Experimental adhesives containing 2-hydroxyethyl methacrylate (HEMA, Acros Organics, NJ), bisphenol-A diglycidyl ether dimethacrylate (bisGMA, Polysciences, Warrington, PA) were photo-polymerized in the presence of water close to the miscibility limit. 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. Bose Electroforce 3200 was used in a 3-point bending configuration for the mechanical testing. Monotonic tests were performed on rectangular beam specimens (1x1x15 mm^3) in both dry and wet conditions to obtain the stress-strain curves. Dynamic mechanical analyses were performed using DMA Q800 (TA Instruments, New Castle, USA). The test temperature was varied from 0 to 200 ^oC with a ramping rate of 3 ^oC/min at a frequency of 1 Hz. The microscale morphologies of cured specimens were observed using X-ray µCT (MicroXCT-400, Xradia Inc. Concord, CA).

Results: The water miscibility, as well as the hydrophilic/hydrophobic ratio decreases along the phase boundary line in the ternary phase diagram (Fig. 1). There was little difference in DC of these formulations, but there were substantial differences in the mechanical and chemical properties of the polymers along the phase boundary (Fig. 2). In the dry case, the mechanical behavior varies with chemical composition, as well as crosslink density. In the wet case, the mechanical behavior is strongly influenced by water sorption, which again depend on the chemical composition and pore structure.

Conclusions: In view of the viscosities, diffusion properties, and miscibility, the penetration of hydrophilic and hydrophobic adhesive components into the wet dentin are different. As a result, the phase compositions along the phase boundary represent all possible phases that could exist (for this simplified formulation). Structure-property relationships from this research will enhance our understanding of adhesive behavior and contribute to identifying strategies for reducing the detrimental effects associated with adhesive phase separation. The mechanical model simultaneously incorporates several inter-dependent factors such as porosity, composition, and physical parameters. These structure/property relationships can be input into a micromechanical model of the adhesive/dentin interface.

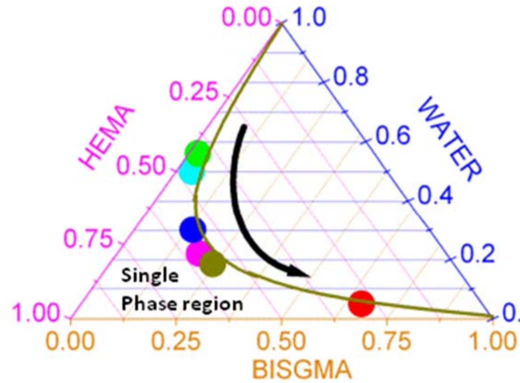

Figure 1. Ternary Phase Diagram and corresponding formulation along the phase boundary to represent the hybrid layer.

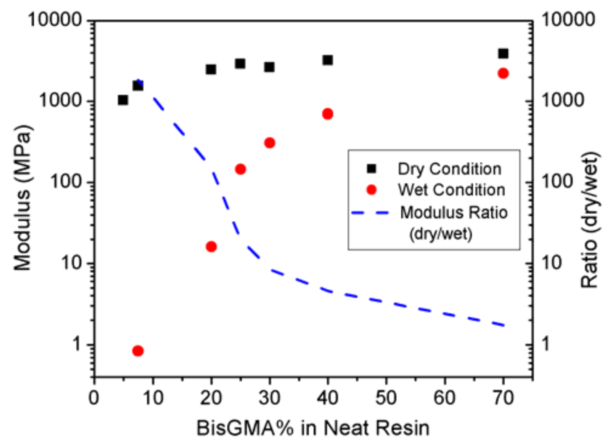

Figure 2. Mechanical testing in dry vs. wet conditions.

References:
1. Spencer P, Wang Y. J Biomed Mater Res 2002, 62:447
2. Ye Q et al. J Dent Res 2011, 90(12):1434-1438
3. Ye Q et al. J Biomed Mater Res Part B 2012, 100B:1086-1092

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