Toughness and Modulus of Photopolymerizable Acrylate-based Networks are Altered under Physiological Conditions

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Statement of Purpose: Photopolymerizable acrylate networks serve as promising biomaterials due to their demonstrated use in minimally invasive procedures (1) and tunable physical and thermomechanical properties (2). The design of novel biomaterials for implementation in high load bearing environments such as the knee or spine is driven by two key properties, the elastic modulus and toughness. It is known that the mechanical properties of polymers are influenced by temperature and humidity (3), but a relationship between toughness, modulus and physiological conditions (i.e. body temperature in saline solution) has yet to be determined. In order to build an understanding of how environmental factors influence network toughness, specifically in photopolymerizable acrylates, stress strain measurements were performed on several model acrylate systems in air and phosphate bufferec saline (PBS) and compared with a linear acrylate (PMMA), a common biomedical grade polymer.

Methods: Model acrylate networks consisting of at least one monofunctional acrylate and a difunctional acrylate (crosslinker) were copolymerized under 365nm UV light using 2,2 dimethoxy 2-phenylacetophenone as a photoinitiatior. The chosen monofunctional acrylates were methyl methacrylate (MMA), methacrylate (MA), and 2-hydroxyethyl methacrylate (2HEMA) while the crosslinker was poly (ethylene glycol) dimethacrylate (PEGDMA) with Mn=750. Poly(methyl methacrylate) (PMMA) was obtained in 1mm thick sheets. Polymer sheets were laser cut into ASTM D638 dogbones and tensilely strained to failure at 5% strain/minute under isothermal conditions. To simulate physiological conditions, samples were soaked in PBS for 24 hours and strained under the same testing regime in an environmental bath filled with PBS. The mass of each sample was measured before and after soaking, and the swelling ratio (q) was calculated as the ratio of the wet mass to initial dry mass. Elastic modulus was calculated as the slope of the initial linear portion of the stress strain curve while toughness was calculated as the area under the stress strain curve.

Results: The swelling ratios of MMA-co-PEGDMA, 2HEMA-co-PEGDMA, and MA-co-MMA-co-PEGDMA were 1.14±0.01, 1.58±0.05, and 1.02±0.01, respectively. The elastic moduli of the acrylate networks at 40C decreased in PBS compared to values in air (Figure 1a). For MMA-co-PEGDMA and 2HEMA-co-PEGDMA, toughness decreased at least one order of magnitude when strained in PBS. On the other hand, MA-co-MMA-co-PEGDMA showed no difference in toughness in air and PBS. PMMA did not exhibit any change in modulus or toughness in PBS compared with testing in air.

Conclusions: The modulus and toughness of acrylate networks is influenced by the environment, particularly at body temperature in saline solution. 2HEMA-co-PEGDMA had the highest swelling ratio and experienced the largest decrease in toughness while MA-co-MMA-co-PEGDMA had a small swelling ratio and experienced no change in toughness between testing in PBS and air. These results demonstrate that the presence of aqueous conditions must be taken into consideration when developing photopolymerizable acrylate networks for biomedical applications. Future studies should address how network properties such as crosslinking and monomer chemistry can be tailored to optimize toughness and maintain modulus under aqueous conditions. From these results, novel acrylate networks can be designed with the appropriate modulus and enhanced toughness for implementation in high-load bearing applications.

References: (1) Elisseef J et al, Plas Reconst Surg 104: 1014-1022, 1999; (2) Gall K et al, J Bio Mat Res A: 73A: 339-348, 2005; (3) Bolon et al, J Appl Pol Sci 25(4): 543-553, 1980.

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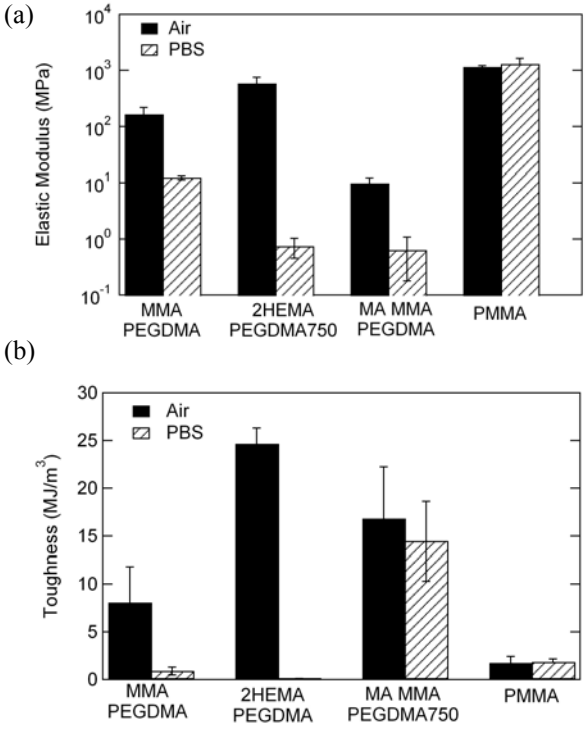

Figure 1. The effect of PBS on the elastic modulus (top) and toughness (bottom) of 3 acrylate-based networks and PMMA. Data are Mean±SEM.