

Temporal Control of Mechanical Properties of Degradable Poly(β -amino ester) Networks

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Statement of Purpose: Current biodegradable polymers have been used in a multitude of applications, such as tissue scaffolds, orthopedic devices, and drug delivery devices. The ability to degrade is beneficial because a drug can be released eliminating the need for additional surgeries to remove the device; however, there is always a loss in mechanical properties for these polymers¹. A material that can degrade over time, and maintain or improve its mechanical properties would be beneficial in load-bearing biomedical applications.

The mechanical properties of nondegradable crosslinked amorphous networks are highly dependent upon the thermal properties, where changes in the glass transition temperature (T_g) have shown to alter the toughness under immersed conditions². The objectives of this study are (1) control the degradation of poly(β -amino ester)-based networks by altering the chemical composition and (2) to alter the thermal properties of the network through degradation, thereby changing the mechanical properties.

Methods: Biodegradable poly(β -amino esters) macromers were formed from hexanediol diacrylate (HDDA), poly(ethylene glycol) diacrylate Mn~700 (PEGDA), and 3-methoxypropylamine in select ratios of PEGDA:HDDA ratio while maintaining a constant diacrylate to amine ratio of 1.15:1. The reaction proceeded at 90°C for 24 hours on a rotary shaker at 200 rpm. Macromers were mixed with methyl methacrylate at 55 wt % and photoinitiator, Irgacure 2959, at 0.5 wt%. 1 mm thick sheets were photopolymerized with a UV lamp at 365nm. Samples, ASTM D638 dogbones for mechanical testing and 1 cm² squares for degradation, were soaked in phosphate buffered saline (PBS), pH=7.4, at 37°C for up to 8 weeks. Dogbone samples were strained to failure on a MTS Insight 2 in a custom environmental chamber filled with PBS at 37°C at a strain rate of 10⁻³ s⁻¹. Square samples were dried for 24 hours and the change in mass was taken in order to determine mass loss. T_g was determined via dynamic mechanical analysis at a rate of 3°C/min from -100°C to 200°C on a TA Q800. Each point represents the mean \pm standard deviation (n=3).

Results: Networks were synthesized with 3 molar ratios of PEGDA:HDDA at a constant wt% MMA. Figure 1A shows the degradation profile over 8 weeks for the three networks. The mass loss increased as the PEGDA:HDDA ratio increased. The effect of PEGDA:HDDA ratio on the T_g during degradation is shown in Figure 1B. The T_g increases over time for each network and increases as the PEGDA:HDDA ratio increases. Figure 1C shows the toughness profile of the three networks during degradation. The changes in toughness during degradation are composition dependent.

Conclusions: The biodegradable networks are comprised of two components, a low T_g macromer and a high T_g methyl methacrylate component. The networks with increased PEGDA:HDDA ratio show increased degradation due to the presence of PEGDA, which is hydrophilic. The degradation caused an increase in the networks' T_g . The toughness changes for these networks because of the increasing T_g ; however, a high increase in T_g can be detrimental and cause brittle behavior. This system establishes a platform for tailoring toughness of biodegradable networks. Future work will include tailoring monomers to enable sustained toughness over long time periods.

References:

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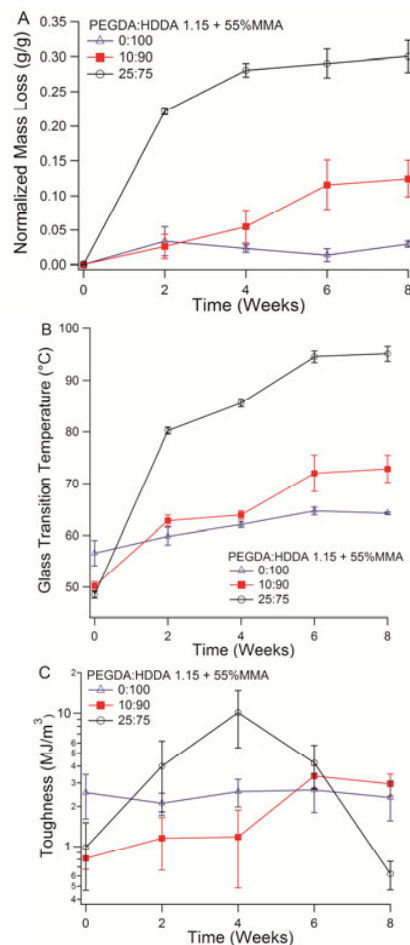


Figure 1. (A) Degradation profile, (B) T_g profile, (C) toughness profile of PEGDA:HDDA-co-MMA networks.