

Quantifying Degradation Process of Physical Gels

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Physical gels are soft materials formed through weak and reversible interactions such as hydrogen bonding and polymer entanglement, resulting in network structures capable of retaining large amounts of solvent. They are widely utilized in food, cosmetics, and biomedical fields, wherein similar structures are also found in biological systems. The spontaneous degradability is a characteristic phenomenon in physical gels, as well as their viscoelastic and self-healing properties that have been modeled quantitatively. In contrast, degradability in physical gels has been less understood and often described only qualitatively, partly due to the difficulty in controlling dissociation kinetics and limited temporal resolution in conventional measurements. In this study, we aim to understand degradation phenomena in physical gels, which is crucial for applications in degradable materials and tissue engineering.

To quantify the degradation properties in physical gels, we systematically measure the time evolution of gel diameter (d), total polymer number (N), and polymer concentration (c) under controlled dissociation kinetics in physical bonding, using DNA-crosslinked four-armed PEG gels as a model system [1]. In this system, by tuning the DNA sequence, the dissociation kinetics of crosslinkers can be precisely controlled over a wide range (1–1000 s) by changing temperature at 5–19 °C. Moreover, the use of fluorescently labeled DNA enabled non-contact, real-time monitoring under accurate temperature control.

Fig. A-C summarizes the degradation behavior in model physical gels. In Fig. A, d/d_0 initially increased under all conditions due to swelling, but subsequently, d/d_0 decreased at higher temperatures (e.g., 19 °C), suggesting network collapse. In Fig. B, N/N_0 decreased over time, except at 5 °C, indicating structural retention. In Fig. C, c/c_0 showed an initial rapid drop from dilution, then declined slowly. At 5 °C, c/c_0 followed a power-law decay ($c \sim t^{-1/3}$), while at higher temperatures, deviation occurred in later stages due to degradation.

We classified the degradation phenomena in physical gels into three modes: (i) pure swelling, (ii) restructuring-coupled swelling, where reversible crosslink dynamics reduce network density, and (iii) surface erosion, where polymers detach from the gel surface. These modes exhibit distinct time-dependent patterns in d , N , and c , enabling a mechanistic interpretation of degradation based on experimentally accessible parameters. While (i) and (ii) show increasing d and decreasing c with constant N , (iii) results in reduced d and N , with a slight change in c . We remark that our finding provides a comprehensive framework for understanding and designing degradable soft materials.

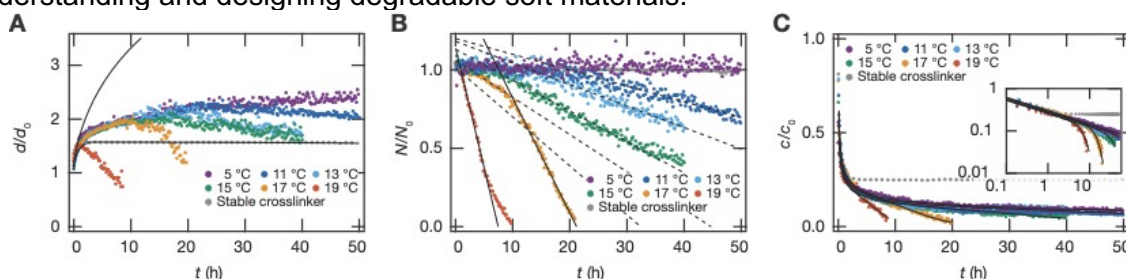


Fig. Time evolution of (A) normalized gel diameter d/d_0 , (B) total polymer number N/N_0 , and (C) polymer concentration c/c_0 under various temperatures. Inset in (C) shows a log-log plot.

References:

[1] M. Ohira, et al., *Adv. Mater.* 34, 2108818 (2022).