Physically and chemically gelling hydrogel formulations based on poly(ethylene glycol) diacrylate and Poloxamer 407

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Physical hydrogels are viscoelastic solids containing a polymer network held together by physical interactions and which is swollen in water. Due to the reversibility of the cross-links, physical hydrogels are often used as injectable hydrogels, as shear thinning materials in additive manufacturing of hydrogels, as drug delivery devices or in tissue engineering.[1] However, the mechanical strength of physical hydrogels is often limited. In order to fix the shape of a physical hydrogel after processing into a defined shape, there is a general demand for formulations of physical hydrogels which can be chemically cured. Chemical hydrogels resulting from the curing process then have improved form stability and mechanical strength. An attractive approach for the formulation of both physically gelling and chemically curable hydrogels is the combination of two materials which contribute the two desired properties. In this contribution, the success of this approach is demonstrated using the synthetic hydrogel precursors poly(ethylene glycol) diacrylate (PEG-DA) and Poloxamer 407 (P407).

To this end, hydrogel formulations containing P407, PEG-DA (M_n = 700 g mol^{-1}) and the photo-initiator 1-[4-(2-hydroxyethoxy)phenyl]-2-hydroxy-2-methyl-1-propan-1-one (Irgacure 2959) were characterized concerning their physical and chemical gelling behavior. Upon addition of PEG-DA to P407 solutions, their gel transition temperature T_{gel} was altered and could be adjusted between 10 °C and 39 °C. At P407 concentrations c_{Polox} of <22.5 wt.-%, T_{gel} increased compared to P407 solutions by adding PEG-DA. At c_{Polox}>22.5 wt.-% T_{gel} decreased. This could be explained by a decrease of P407 micelle size upon PEG-DA addition, as shown by dynamic light scattering. Chemical gelation of the formulations was investigated by FT-IR spectroscopy, by rheology, and by the equilibrium degree of swelling of the cured hydrogels. The shear moduli and equilibrium degrees of swelling of the cured hydrogels could be adjusted by the PEG-DA concentration in the formulation, similar to pure PEG-DA hydrogels.

In order to demonstrate the possibility of using the hydrogel formulations as injectable hydrogels, we used them as inks for the additive manufacturing technique of robotic dispensing. Thus we were able to produce various shapes of spatially well-defined hydrogels (see Figure 1). Our results give rise to the conclusion that the hydrogel formulations are well suited as injectable hydrogels with adjustable properties.

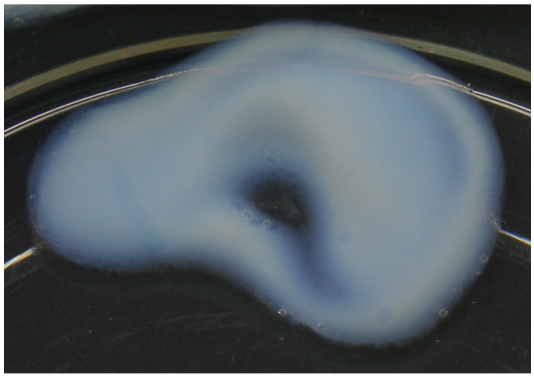

Figure 1 Ear shape produced from the hydrogel formulations by robotic dispensing.

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[1] T. Jungst, W. Smolan, K. Schacht, T. Scheibel, J. Groll, *Chemical Reviews* **2016**, *116*, 1496-1539.