

Supplementary Materials

## Development of Neutral pH-Responsive Microgels by Tuning Cross-Linking Conditions

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**Longitudinal relaxation time ( $T_1$ ) and transverse relaxation time ( $T_2$ ).**

$^1\text{H}$ -MRI signal intensity depends on two relaxation processes of the net magnetization ( $M_0$ ) of water proton spins.  $M_0$  is initially parallel with static magnetic field (Z-direction) but there is no net magnetization on XY plane. By applying radio frequency (RF) pulse, proton spins are excited to a higher energy state involved with phase coherence and  $M_0$  is tilted out from the Z-direction. At the end of RF pulse, the excited proton spins gradually return to thermal equilibrium state and the net magnetization is also relaxed to the initial state  $M_0$ . This relaxation process is separated into two components, Z-direction and XY-direction. The net magnetization recovery of the Z-direction is termed longitudinal relaxation defined as  $M_z = M_0[1 - \exp(-t/T_1)]$ ;  $T_1$  is the time required for  $M_z$  to approach 63% of  $M_0$ . Longitudinal relaxation is energy exchange between proton spins and surrounding lattice. On the other hand, the net magnetization decay on the XY-plane is termed transverse relaxation. This process is a loss of phase coherence and defined as  $M_{xy} = M_0 \exp(-t/T_2)$ ;  $T_2$  is the time required for  $M_{xy}$  to approach 37% of  $M_0$ . General MRI contrast agents significantly reduce  $T_1$  and  $T_2$  due to a dipole-dipole interaction between their electron spins and proton nuclear spins and/or distortion of local magnetic field.

**Table S1.** Mean diameters of the microgels calculated from TEM images.

Group	Microgels cross-linked by	Mean diameter $\pm$ SEM
1	40mol% EGDMA	120 $\pm$ 4 nm ( $n = 69$ )
1, 2	20mol% DEGDMA	161 $\pm$ 2 nm ( $n = 76$ )
1	20mol% TEGDMA	293 $\pm$ 4 nm ( $n = 31$ )
1	20mol% TETEGDMA	326 $\pm$ 13 nm ( $n = 54$ )
2	30mol% DEGDMA	227 $\pm$ 5 nm ( $n = 44$ )
2	40mol% DEGDMA	564 $\pm$ 16 nm ( $n = 35$ )
2	45mol% DEGDMA	579 $\pm$ 15 nm ( $n = 14$ )