Supporting Information

Permeability Mapping of Gelatin Methacryloyl Hydrogels

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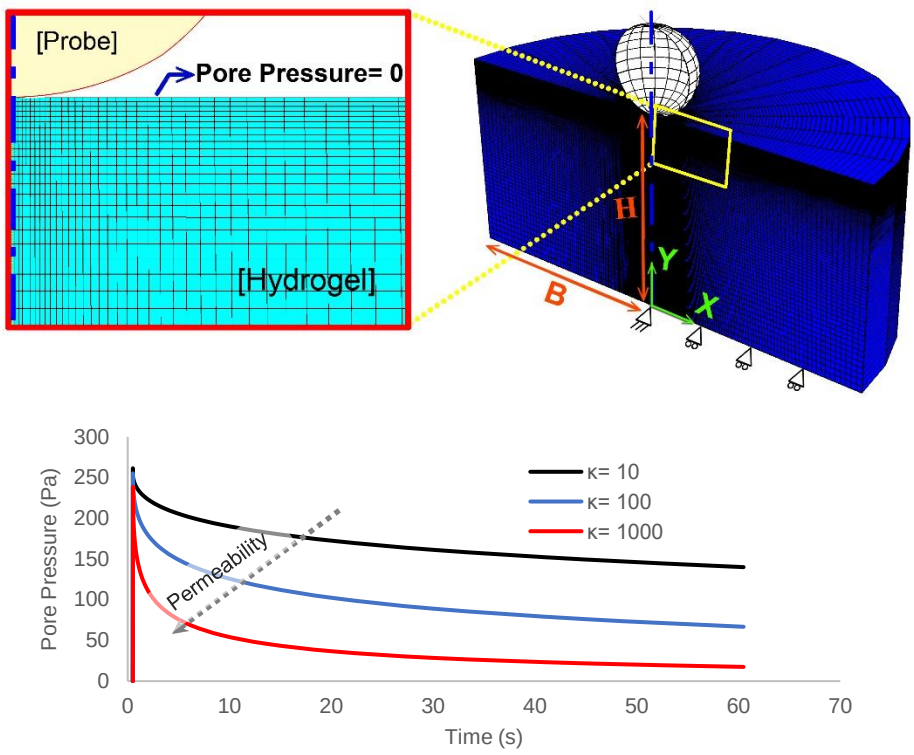
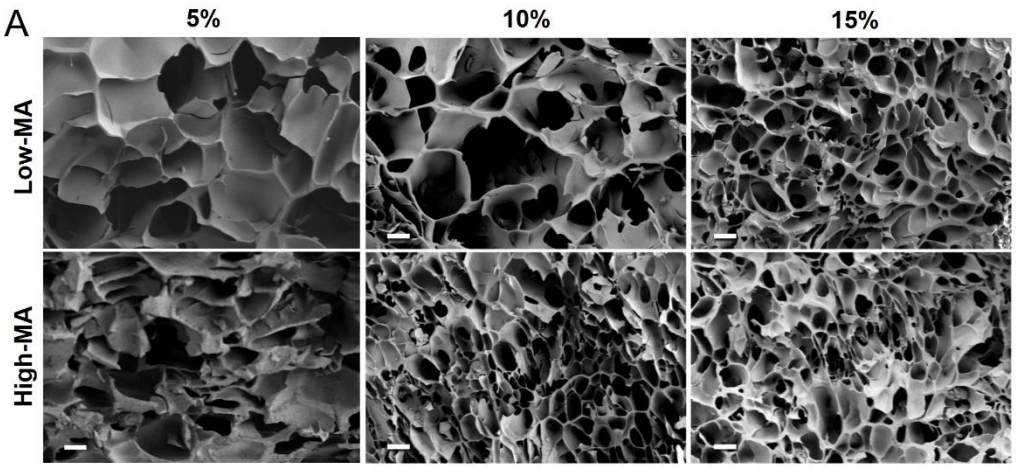


Figure S1. Geometry of simulation model and the used mesh system, and summation of fluid pressure in hydrogel elements over time.



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	5%	10%	15%
Low-MA	96 ± 48 μm	61 ± 29 μm	39 ± 21 μm
Medium-MA	62 ± 31 μm	43 ± 19 μm	30 ± 18 μm
High-MA	45 ± 27 μm	36 ± 17 μm	19 ± 14 μm

Figure S2. SEM of GelMA microstructure (scale bars: 50 μm): A) Role of GelMA concentrations and MA degrees at a fixed UV crosslinking time (all in 60 s). B) Role of UV crosslinking time at a fixed GelMA concentration and MA degree (10% GelMA, medium-MA). C) One quantification of void sizes from SEM images for 60-s UV-crosslinked samples.

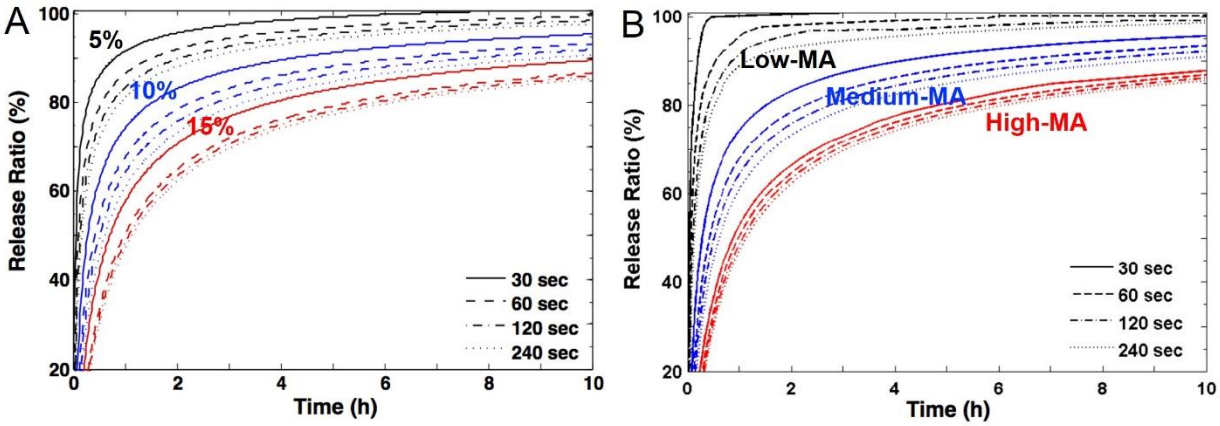

Figure S3. Release ratios computed by the theoretical model for A) medium-MA GelMA at different GelMA concentrations (black: 5%; blue: 10%; red: 15%) and B) 10% GelMA at different MA degrees (black: Low-MA; blue: Medium-MA; red: High-MA).

Mesh size calculation

We estimated the mesh sizes of medium- and high-MA GelMA hydrogels using the data presented by Yoon *et al.* [41] and by adapting the model derived by Zhou *et al.* [42]. It should be noted that, we were not able to directly adapt the model in Ref. [42] since their formulas were adopted for a zwitterionic hydrogel system where the chemical interaction between solutes and hydrogel was a dominant mechanism in mesh formation. However, in our system we neglected such interactions and the diffusion was more dominant. Specifically, we calculated the mesh size variations in our GelMA samples through the use of Eq. (S1) in Ref. [42]:

$$\varepsilon = {v_{2,s}}^{-1/3}\left[C_n\left(\frac{2\,M_c}{M_r}\right)\right]^{1/2} l \qquad \text{(S1)}$$

, where ε denotes the mesh size, $v_{2,s}$ represents the polymer volume fraction of hydrogel at equilibrium swelling state, C_n reflects the stretching degree of polymer chain in the solution, M_c demonstrates the number-averaged molecular weight of chains between crosslinks, M_r is the molecular weight of the repeating unit of GelMA (g/mol), and *l* represents the monomer backbone bond length. M_c is estimated from Eq. (S2) while the parameter χ is approximated from Eq. (S3):

$$\frac{1}{\overline{M_c}} = \frac{2}{\overline{M_n}} - \frac{\bar{v}/V_1[\ln(1-v_{2,s})+v_{2,s}+\chi v_{2,s}^2]}{v_{2,r}[\left(\frac{v_{2,s}}{v_{2,r}}\right)^{1/3}-\frac{v_{2,s}}{2v_{2,r}}]} \qquad \text{(S2)}$$

$$\chi = \frac{\frac{GV_1}{RT}-\ln(1-v_{2,s})-v_{2,s}}{v_{2,s}^2} \qquad \text{(S3)}$$

Here, M_n is the number-averaged molecular weight of chain without crosslinking, which sits around 4000-50000 g/mol in the case of GelMA, the experimental $v_{2,s}$ values for fish-derived GelMA hydrogels when medium and high degrees of methacryloyl substitution were used, are extracted from Ref. [2] and for estimation of swelling behavior at relaxation, a rough estimation of $v_{2,r} = 10\% * v_{2,s}$ was incorporated where $v_{2,r}$ represents the polymer volume fraction of the hydrogel at relaxation. The parameter *G* is the small strain shear modulus, V_1 is the molar volume of the solvent, and $\bar{v}$ is the specific volume of GelMA. Other parameters were roughly adopted from the data provided in Ref. [41] (i.e., $C_n = 16$, *l*= 0.154 nm, V_1=18 ml/mol, $\bar{v}$=1/39 ml/g). The calculated mesh sizes for the different medium- and high-MA hydrogel systems are illustrated in **Fig. S4** and were compared with the variations of the pore sizes obtained from SEM images. A

positive correlation was observed between the mesh size and the pore size despite the fact that the two phenomena belonged to different length scales.

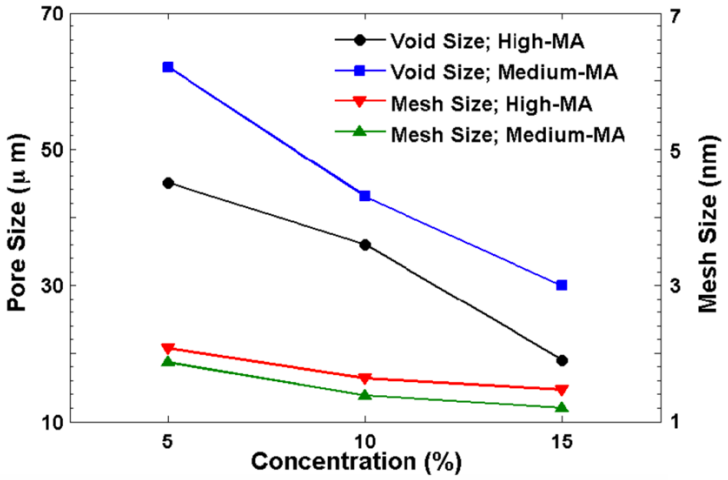

Figure S4. Estimated pore sizes (black: high-MA; blue: medium-MA) and mesh sizes (red: high-MA; green: medium-MA) for three concentrations of GelMA.