

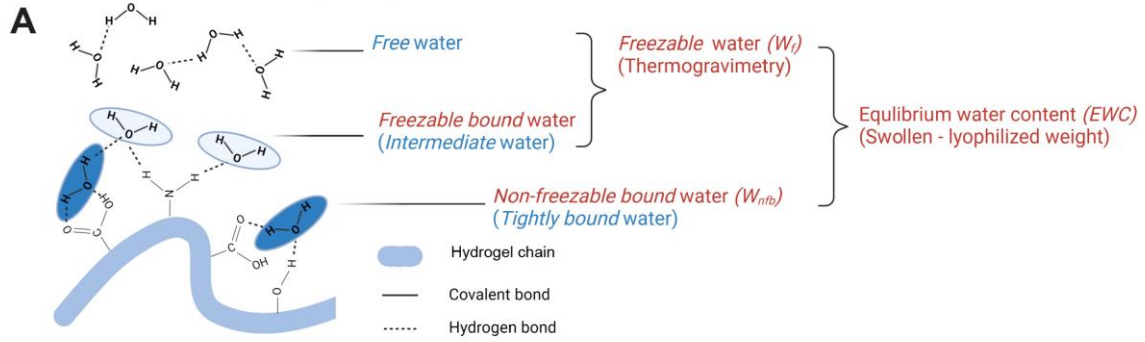
# SUPPLEMENTARY DATA

*Crosslinking substrate regulates frictional properties  
of tissue-engineered articular cartilage and  
chondrocyte response to loading*

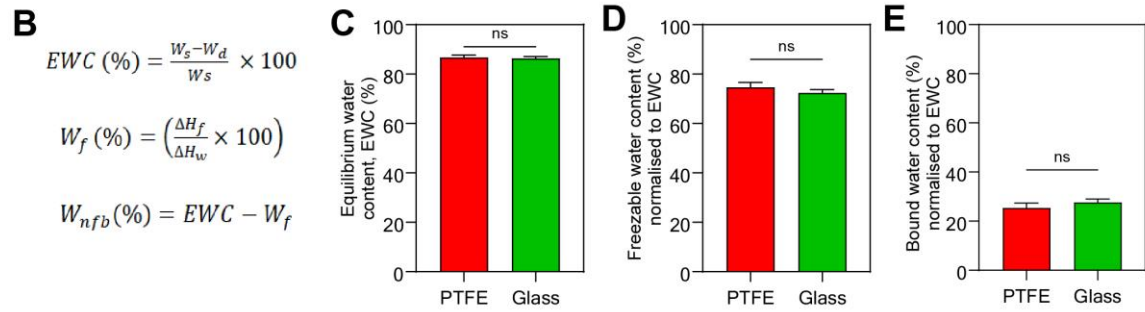
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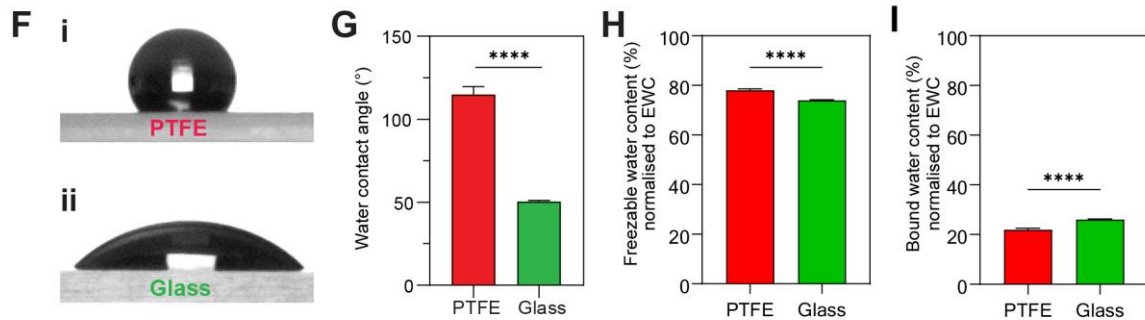
## Molecular Water & Hydrogel Interaction



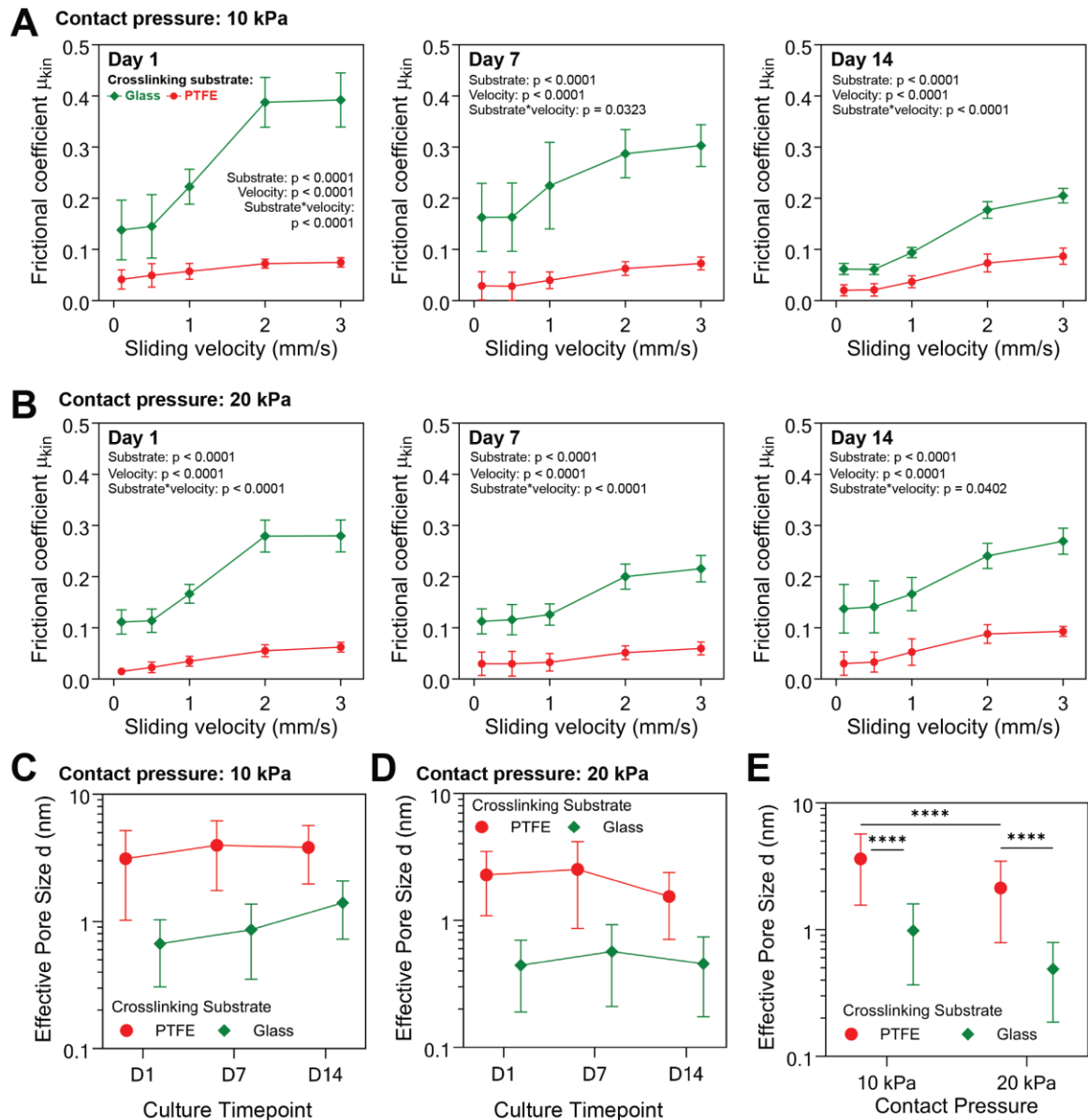
## Hydrogel DSC Water Analysis



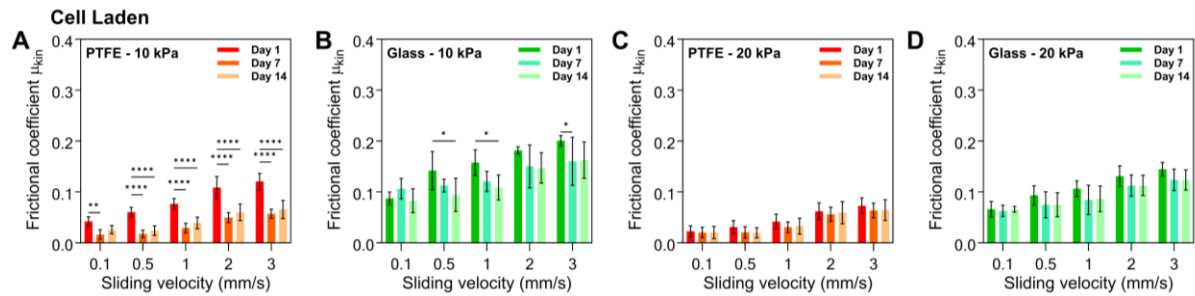
## Substrate Contact Angle & DSC Analysis



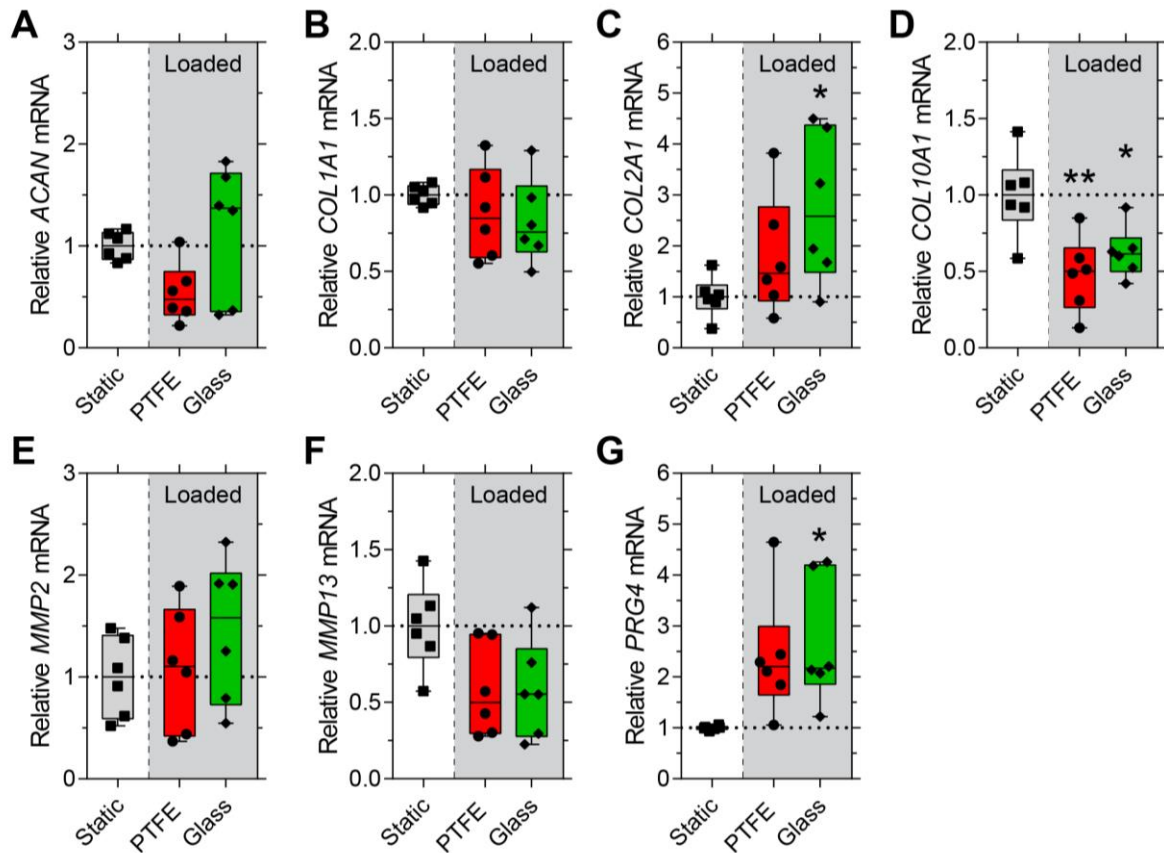
**Figure S1. Analysis of hydrogel water content & interfacial substrate hydrophobicity.** (A) Schematic representation of the molecular interactions at the interfacial surface of swollen hydrogels, depicting the various types of water. (B–E) Determination of equilibrium water content ( $EWC$ ), freezable water ( $W_f$ ) and non-freezable bound water ( $W_{nfb}$ ) based on DSC analyses of GelMA/HAMA hydrogels crosslinked on either PTFE or glass substrates, depicting negligible differences in bulk water content with the hydrogels. (F–I) Determination of substrate hydrophobicity via contact angle analysis, alongside calculation of the relative percentage of freezable water ( $W_f$ ) and non-freezable bound water ( $W_{nfb}$ ) directly interfaced with either PTFE or glass substrates, based on DSC analyses. Values reported as mean  $\pm$  SD, where symbols indicate significance (\*\*\*\* $p < 0.0001$ ).



**Figure S2. Tribological properties of cell-free GelMA/HAMA hydrogels photocrosslinked on glass or PTFE substrates.** Kinetic friction coefficients  $\mu_{kin}$  of cell-free 9.5% w/v GelMA / 0.5% w/v HAMA constructs photocrosslinked on glass (green) or PTFE (red) substrates at day 1, 7, and 14 incubation, with chondrocyte differentiation media used as lubricant, at contact pressures of (A) 10 kPa and (B) 20 kPa, respectively. Effective hydrodynamic pore size of hydrogel surfaces crosslinked on glass or PTFE at (C) 10 kPa and (D) 20 kPa, respectively, at day 1, 7, and 14. (E) Overall effective hydrodynamic pore size of hydrogel surfaces across the experiment. Values reported as mean  $\pm$  SD, where symbols indicate significance (\*\*\*\* $p < 0.0001$ ).



**Figure S3. Tribological properties of cell-cultured GelMA/HAMA hydrogels photocrosslinked on glass or PTFE substrates.** Kinetic friction coefficients  $\mu_{kin}$  of 9.5% w/v GelMA / 0.5% w/v HAMA constructs photocrosslinked on glass (green) or PTFE (red) substrates at day 1, 7, and 14 incubation, with chondrocyte differentiation media used as lubricant, at contact pressures of (A,B) 10 kPa and (C,D) 20 kPa, respectively. Values reported as mean  $\pm$  SD, where symbols indicate significance (\*\*\*\* $p$  < 0.0001, \*\*\* $p$  < 0.001, \*\* $p$  < 0.01, \* $p$  < 0.05).



**Figure S4. Chondrocyte gene expression in static and dynamic GelMA/HAMA hydrogels after 28-day culture, comparing gels photocrosslinked on PTFE and glass substrates.** Relative expression levels of (A) ACAN, (B) COL1A1, (C) COL2A1, (D) COL10A1, (E) MMP2, (F) MMP13, and (G) PRG4 in statically cultured (free swelling) and dynamic (intermittently loaded) hydrogels after 28 days culture. Box-plots presented with median center line and all points shown, with upper and lower quartiles presented and whiskers extending from minimum to maximum, where symbols indicate significance with respect to the unloaded static group as the control (\*\* $p$  < 0.01, \* $p$  < 0.05).