Supporting Information

Soft, strong, tough, and durable bio-hydrogels via maximizing elastic entropy

*Dani Liu, Shi Feng, Qingqiu Huang, Shuofei Sun, Gening Dong, Feifei Long, Mario Milazzo, Mingkun Wang**

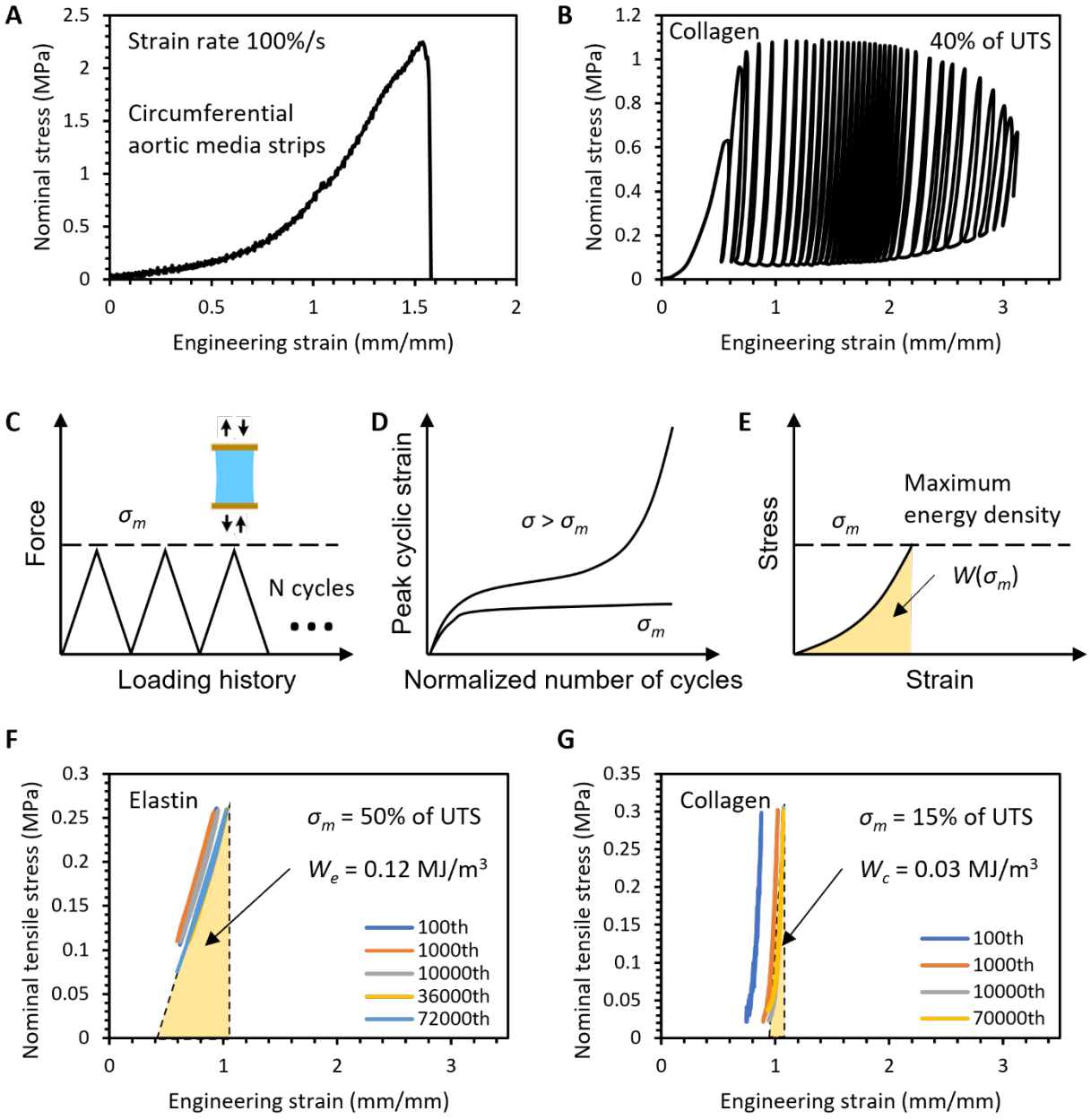

Figure S1. Biomechanical tests and measurement of maximum energy density. (**C**) To measure the maximum energy density, samples were loaded in creep-fatigue at 1 Hz until failure or for 20 hrs. (**D**) Samples showed creep behaviors when the applied stress was above critical point σ_m: the peak strain increased with loading cycles, and samples eventually ruptured. The peak strain stabilized with loading cycles and maintained stable when the applied stress is below the critical point (**E**) The maximum energy density $W(\sigma_m)$ is the area under the strain-stress curve when samples were loaded at their critical stress σ_m. (**F**) Creep-fatigue tests of purified elastin, elastin samples could sustain a creep stress up to 50% of their ultimate tensile stress (UTS), giving a maximum energy density as large as 0.12 MJ m^{-3}. (**G**) Creep-fatigue tests of isolated collagen, collagen samples could only sustain a creep stress around 15% of their UTS, giving a maximum energy density less than 0.03 MJ m^{-3}.

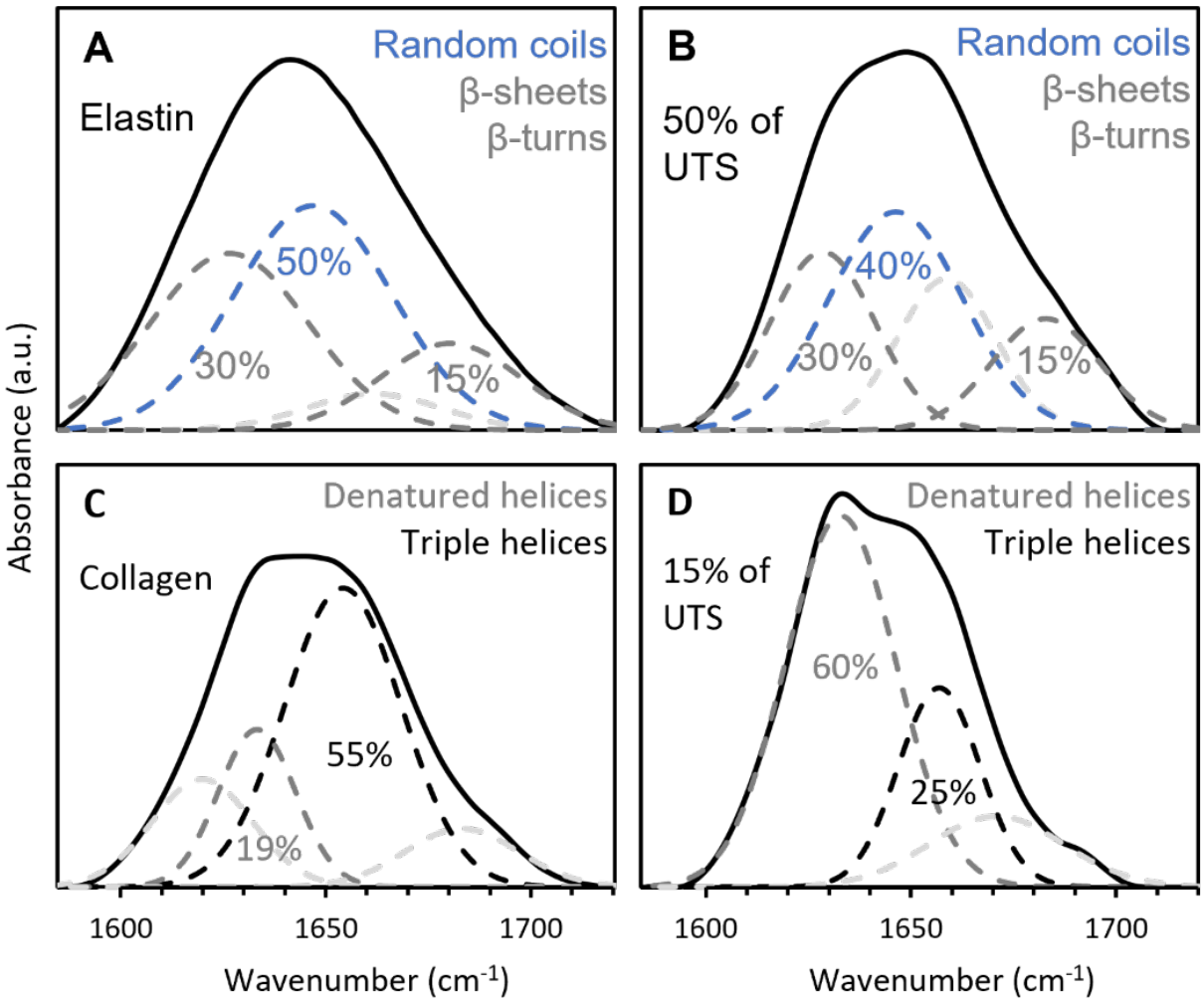

Figure S2. Deconvoluted FTIR amide I band of (**A**) unloaded elastin and (**B**) elastin after having been loaded in creep-fatigue to 50% of their UTS for 20 hours, (**C**) unloaded collagen and (**D**) collagen after having been creep-fatigue loaded to 15% of their UTS for 20 hours.

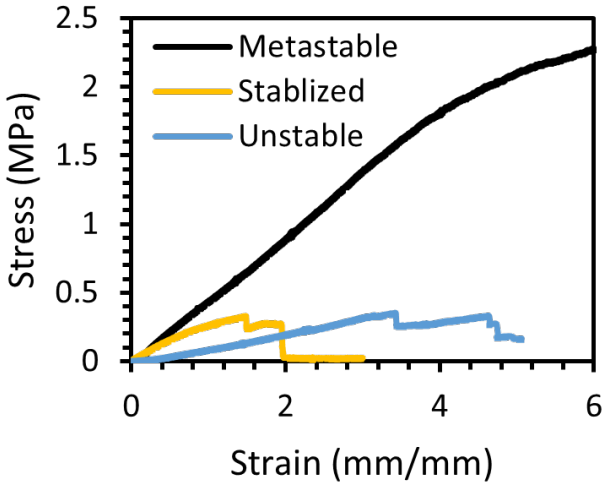

Figure S3. Stress-strain curves of MHEG prepared from different energy states.

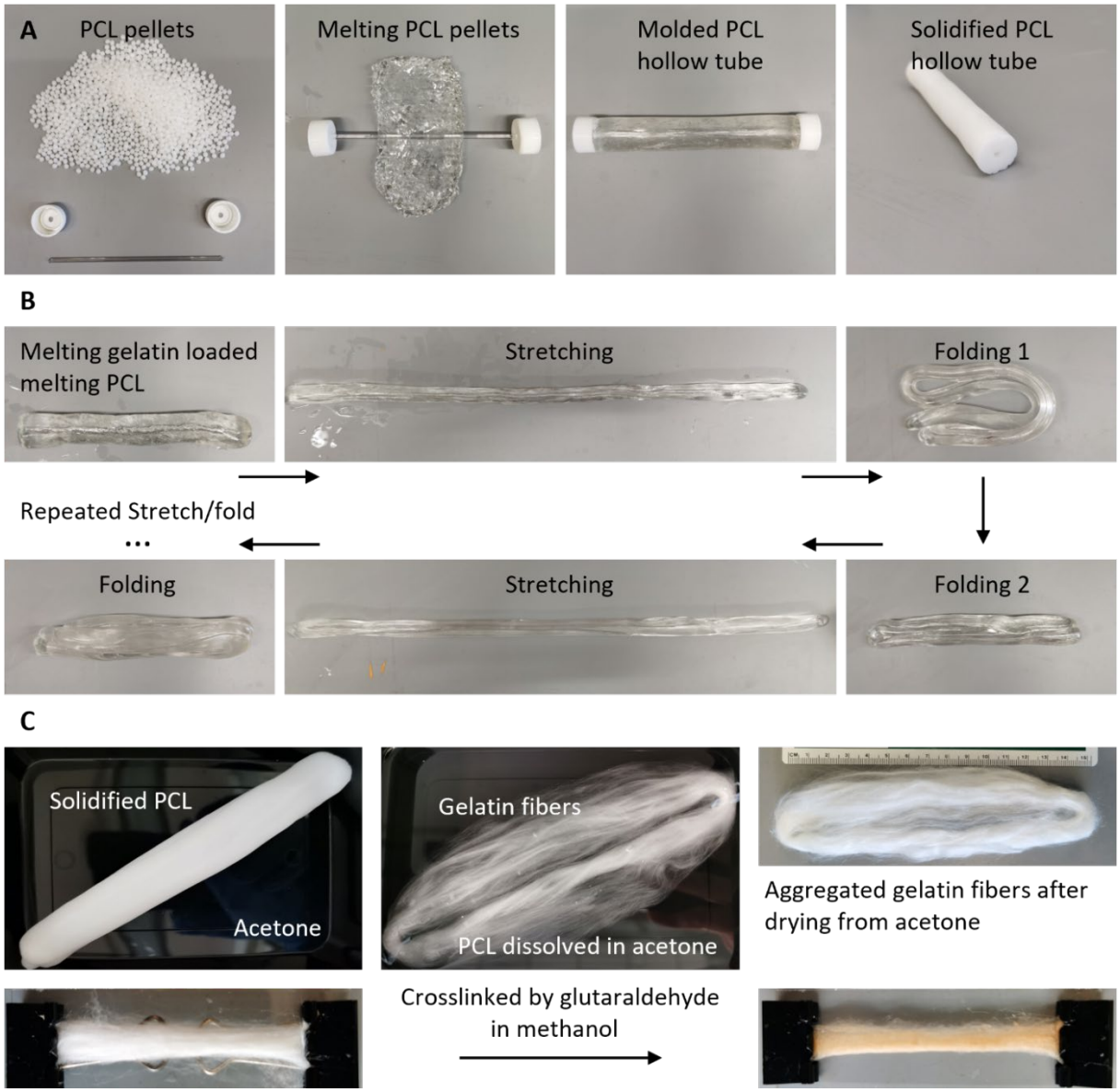

Figure S4. Kitchen table version of fabrication process. (**A**) A PCL shell was produced by melting PCL pellets and then molding and rolling them into hollow tubes. (**B**) The gelatin loaded PCL tube was first melted by heating to 65 °C and then repeatedly stretched and folded. (**C**) The solidified PCL was dissolved in acetone, and the retrieved fibers were crosslinked with glutaraldehyde in methanol overnight.