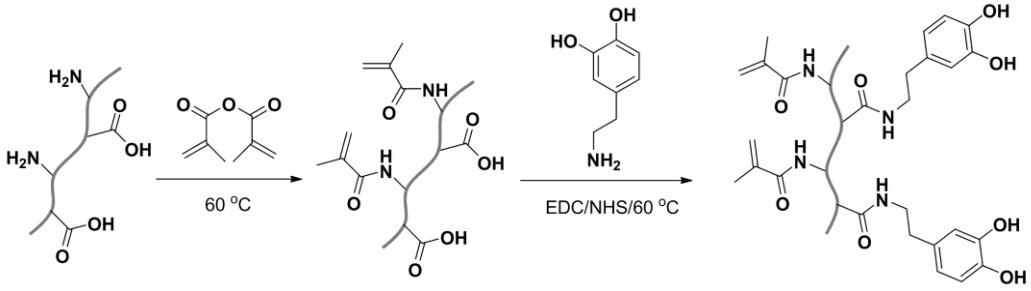
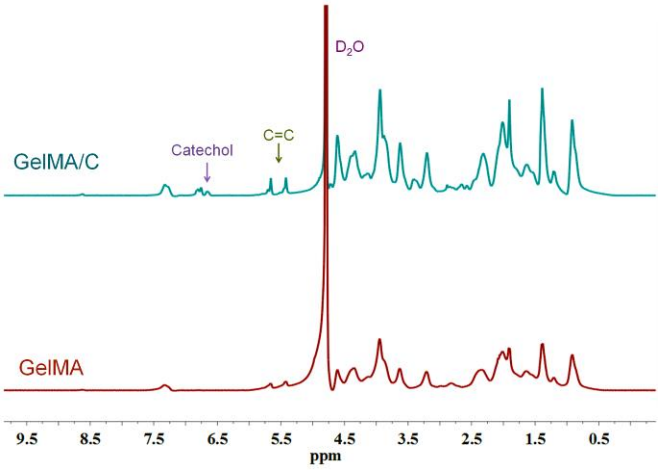
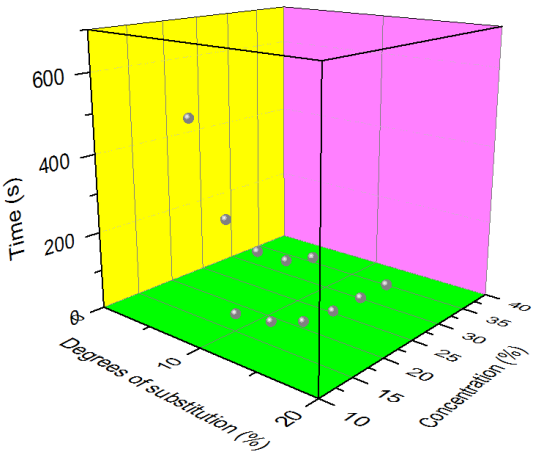
Supplementary Material



Supplementary Figure 1. Synthesis route of GelMA/C bioink.

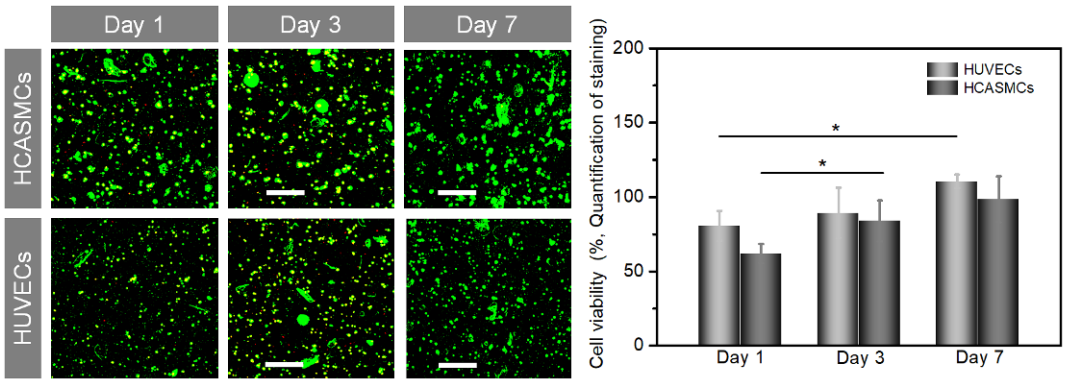


Supplementary Figure 2. ^{1}H NMR spectra of GelMA and GelMA/C copolymers.

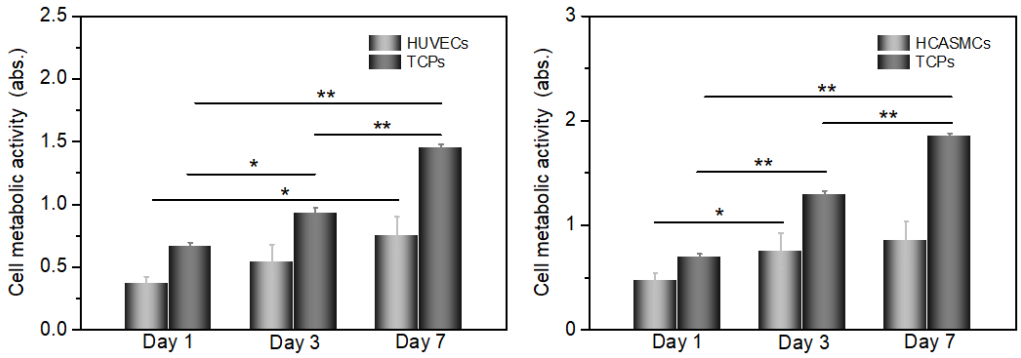


Degrees of Substitution (%)	Bioink Concentration (wt%)					
	10	15	20	25	30	35
6	--	~500 s	~220 s	~100 s	~40 s	~15 s
13.3	~120 s	~60 s	~18 s	~5 s	~2 s	~1 s

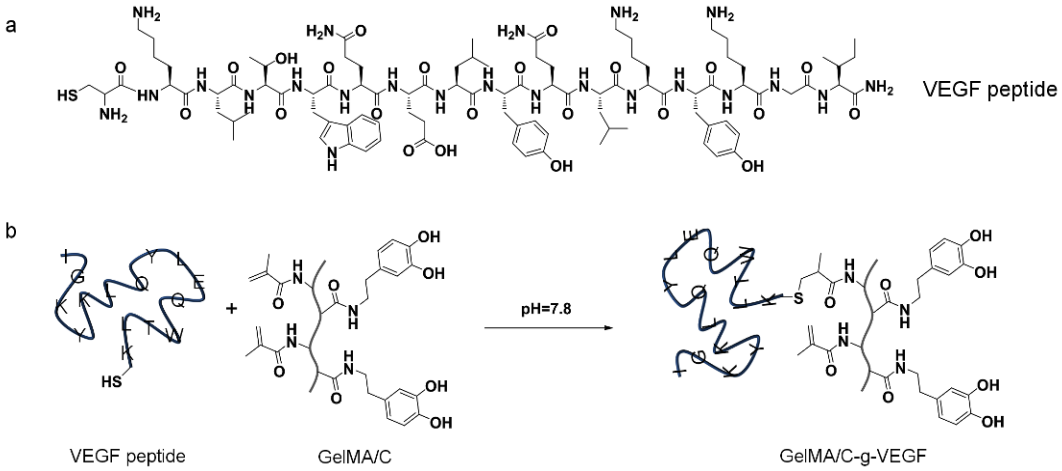
Supplementary Figure 3. The gelation time of GelMA/C solutions by varying DS of catechol and solution concentration.



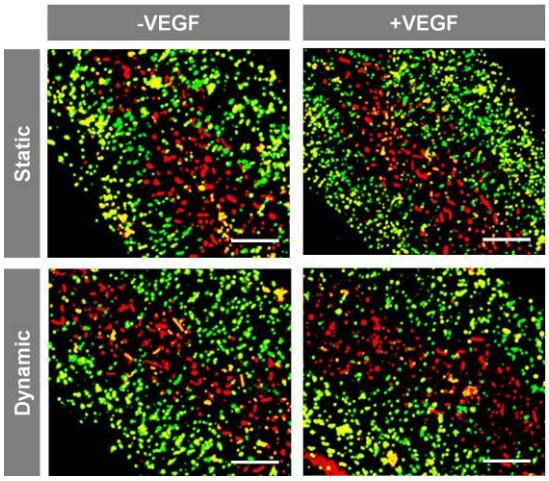
Supplementary Figure 4. Viability assay of HUVECs and HCASMCs in the vasculature using Live-Dead staining after 1, 3, and 7 days of culture. The results were normalized to the whole fluorescence area on day 1. The scale bars indicate 100 μm. Data are the mean ± sd., n≥9, *P <0.05.



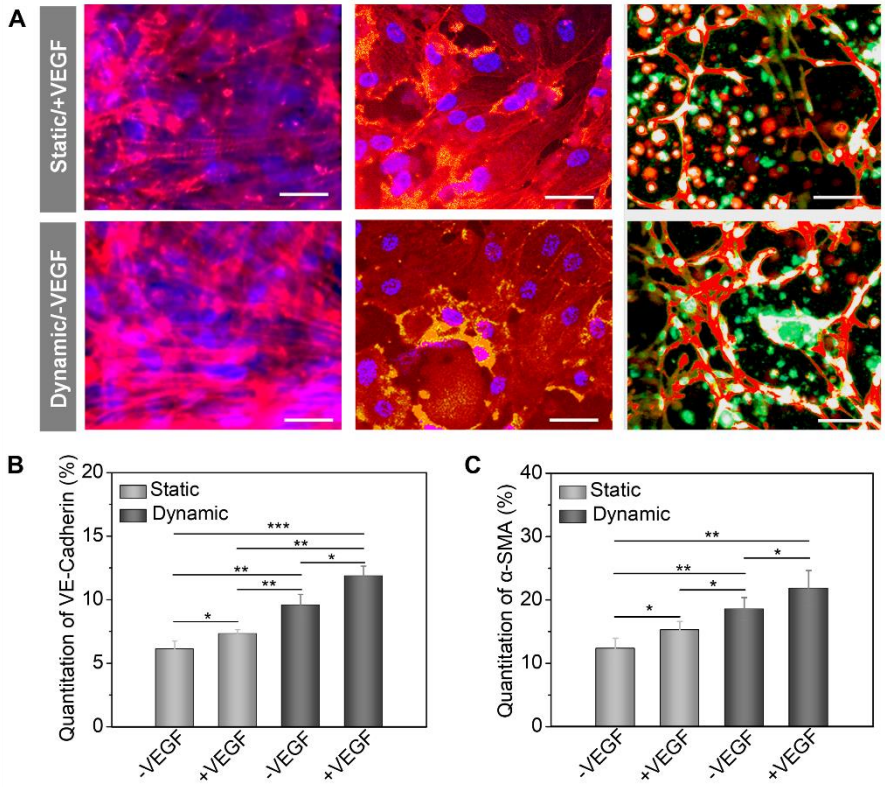
Supplementary Figure 5. Metabolic activity of HUVECs and HCASMCs in the vasculature and on the TCPs using CCK-8 kit after 1, 3, and 7 days of culture; the mean ± sd., n≥9, *P <0.05, **P <0.01



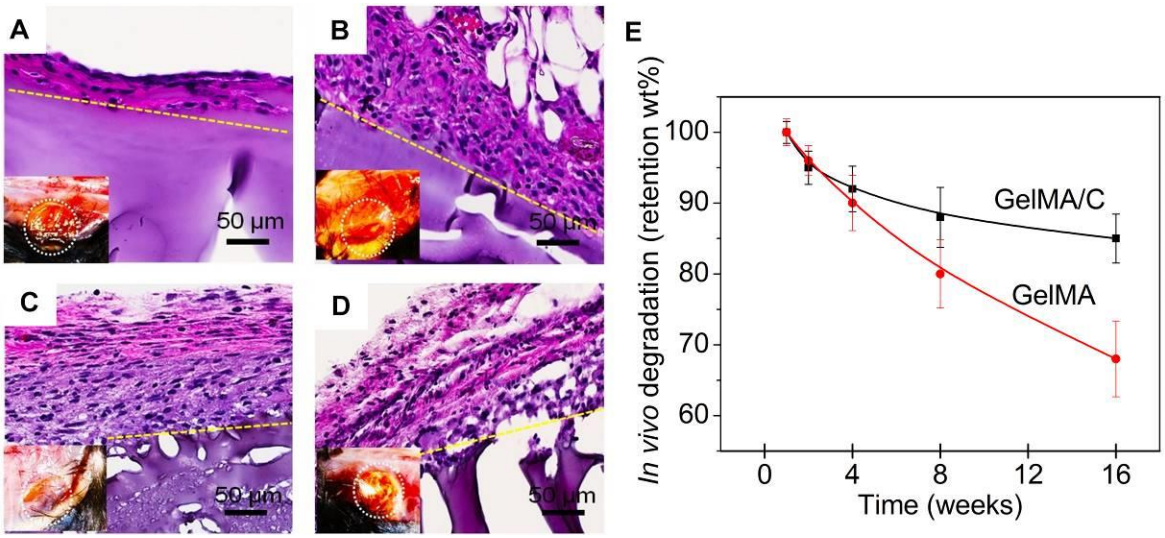
Supplementary Figure 6. Structural schematics of VEGF peptide and its immobilization onto GelMA/C copolymers.



Supplementary Figure 7. Immunofluorescence images of 3D bioprinted vasculature (CD31, red; α-SMA, green) including –VEGF/static group, –VEGF/dynamic group +VEGF/static group, +VEGF/dynamic group. The scale bars indicate 100 μm.



Supplementary Figure 8. (A) Immunofluorescence images of the endothelium (spreading and orientation) of 3D bioprinted vasculatures (vinculin, red; DAPI, blue) including +VEGF/static group, –VEGF/dynamic group. The scale bars indicate 20 μm. Immunofluorescence images of the endothelium (intercellular junctions) of 3D bioprinted vasculatures (VE-cadherin, green; F-actin, red; DAPI, blue) including +VEGF/static group, –VEGF/dynamic group. The scale bars indicate 20 μm. Immunofluorescence images of the smooth muscle of 3D bioprinted vasculatures (α-SMA, green; F-actin, red; DAPI, blue) including +VEGF/static group, –VEGF/dynamic group. The scale bars indicate 50 μm. (B) Quantitative analysis (avg. area % in immunofluorescence images) of VE-Cadherin expression in the endothelial layer of 3D bioprinted vasculature. (C). Quantitative analysis (avg. area % in immunofluorescence images) of α-SMA expression in the smooth muscle layer of 3D bioprinted vasculature.



Supplementary Figure 9. In vivo biocompatibility and degradation of GelMA/C hydrogels after subcutaneous implantation on the back of C57BL/6N mice. Macroscopic view of implanted hydrogel and H&E stained sections of the implants, harvested at week 1 (A), week 4 (B), week 8 (C) and week 16 (D). E shows the degradation rate of oxidative crosslinked GelMA/C and photo-crosslinked GelMA.

Supplementary Movie S1. 3D bioprinting process of vascular constructs using coaxial extrusion bioprinter. Fluorescence dyes were added to improve visualization of GelMA/C ink (Fluorescein) and crosslinking slurry (Nile red).

Supplementary Movie S2. Dynamic flow behavior in our 3D bioprinted vasculature, where cells were perfused into the channel.

Supplementary Movie S3. 3D bioprinted vasculature perfusion in dynamic culture device (customized bioreactor), including temperature controller, peristaltic pump, medium reservoir, gas exchange valve, and culture chamber.