## Supplementary Material

## Spatiotemporal control of polymer brush formation through photoinduced radical polymerization regulated by DMD light modulation

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Fig. S1 <sup>1</sup>H NMR spectrum (a) and <sup>13</sup>C NMR spectrum (b) of the AMEMA monomer.



**Fig. S2** Digital grayscale image for fabricating a PGMA and PAMEMA binary geometric circular pattern



**Fig. S3** DMD spatial light modulation for initiating the radical polymerization process. (a) The designed digital grayscale image for generating spatially modulated light by DMD spatial light modulation, the substrate covered with monomer solution (b) or monomer/catalyst solution (c) was irradiated by modulated light, and fluorescence excitation was monitored via a CCD camera. (d) Excitation and emission spectra of the photoredox catalyst Ir(ppy)<sub>3</sub> in monomer/catalyst solution ( $\lambda ex = 435nm$ ,  $\lambda em = 538nm$ ). (e) Optical imaging of the resultant substrate in the case of monomer/catalyst solution was irradiated by modulated light.



Fig. S4 Optical images of PEGMA brushes on the glass and polystyrene substrates.



**Fig. S5** Influence of exposure energy, exposure intensity (a) and catalyst concentration (b) on the thickness of PEGMA brushes. The two-stage brush thickness curves showed pseudo-first-order kinetics with a relative linearity of brush growth followed by a "saturation" plateau of brush thickness, indicating the "pseudo-living" nature of Photo-ATRP chemistry.



**Fig. S6** Negative-ion TOF-SIMS spectra from micropatterned PAMEMA brushes on the silicon substrate.



**Fig. S7** Fluorescence images of chemical concentration gradients of fluorescein corresponding to different exposure strategies.



**Fig.S8** Optical image of micropatterned PGMA brushes and corresponding fluorescence images of actin- and DAPI-stained HUVECs after 1 day and 3 days of culture.