SUPPORTING MATERIAL

Table S1. Microgel composition and key properties.

Abbreviation	**Nominal composition**			**Size**[a]	**pKa**[b]	**%MAA**[b]	**Ionization**[b]
	EA (wt%)	MAA (wt%)	BDDA (wt%)	(nm)			(at pH 7.4)
MAA20	79.0	20.0	1.0	76.9 ± 2.8	7.0	22.1 ± 1.1	0.72
MAA33	66.0	33.0	1.0	67.7 ± 2.8	6.4	36.9 ± 0.4	0.91
MAA60	39.0	60.0	1.0	180.1 ± 31.4	6.5	63.3 ± 1.5	0.89

a) Microgel hydrodynamic diameter determined with NTA, in 10mM Tris HCl, pH 7.4
b) Mettler Toledo titrator, 0.01M NaCl, initial pH 3.5, n=2.

Figure S1. Microgel degree of dissociation (charge) as a function of pH. Microgel solution (1.0 wt%, in 0.01M NaCl supporting electrolyte) was titrated using Mettler-Toledo Dl 15 titrator (Mettler Toledo, Columbus, USA).

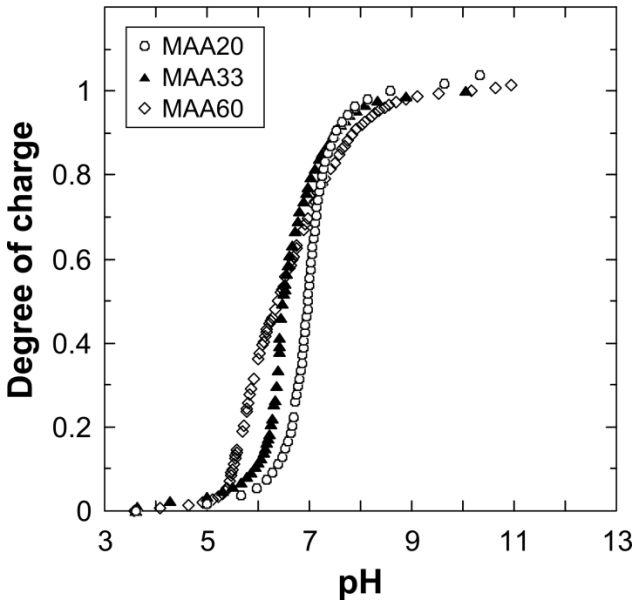

Figure S2. (A) 3D-rendering of 20 CLSM images of Texas Red-labeled MAA33 microgels supported to a GOPS surface. (B) Binding of fluorescently labeled pLys 1 kDa to MAA33 microgels supported either physicochemically on an untreated glass coverslip or a GOPS-treated glass coverslip and covalently immobilized. Microgels equilibrated in 7.5µM pLys 1kDa overnight prior measurements, followed by rinsing.

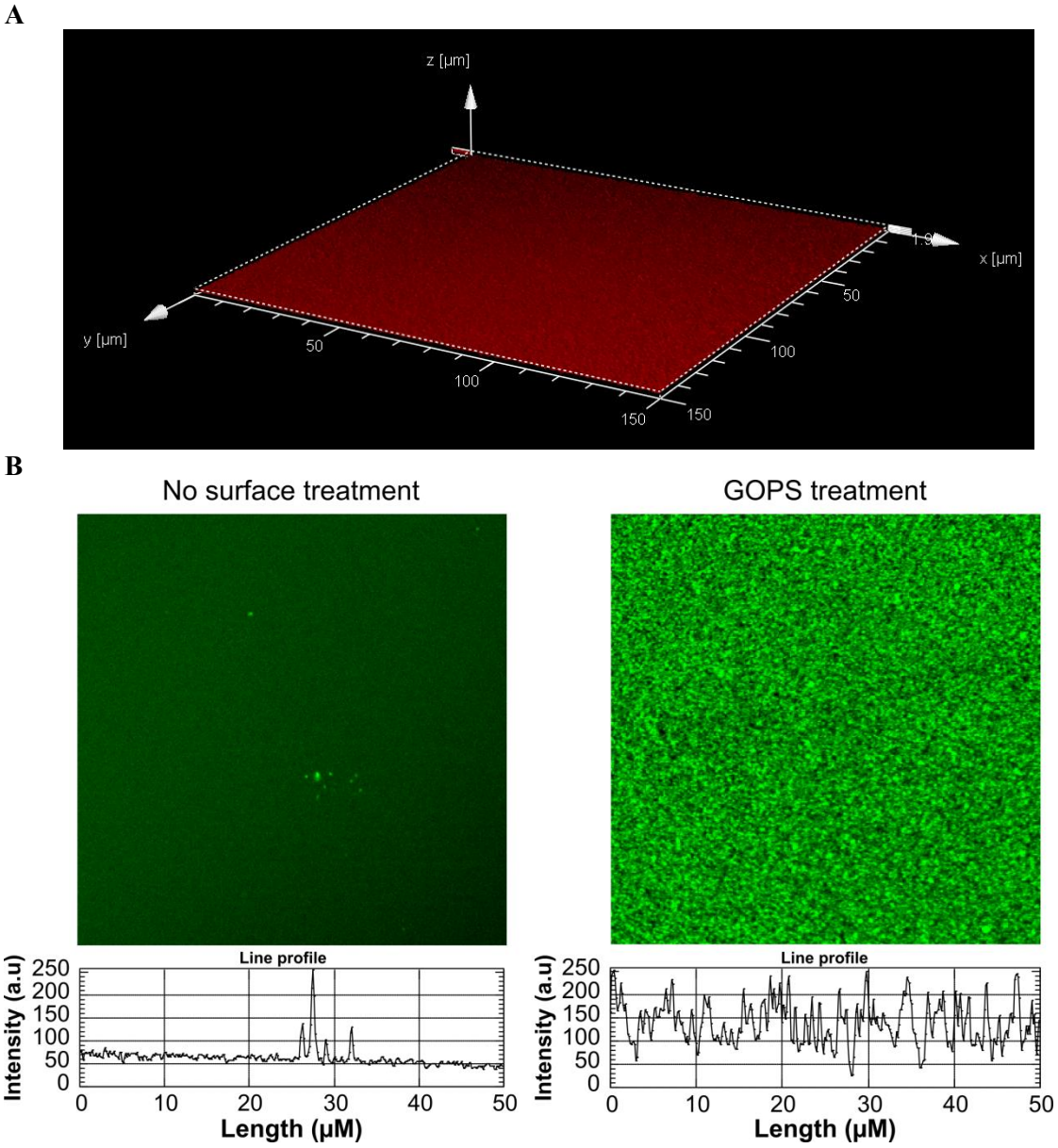

Figure S3. Time-resolved ellipsometry of peptide loading to surface-bound MAA33 microgels. The concentration of peptide (pLys 1, 10, 150 kDa) was sequentially increased in the cuvette after 1 hour incubation. All experiments performed in 10mM Tris HCl, pH 7.4.

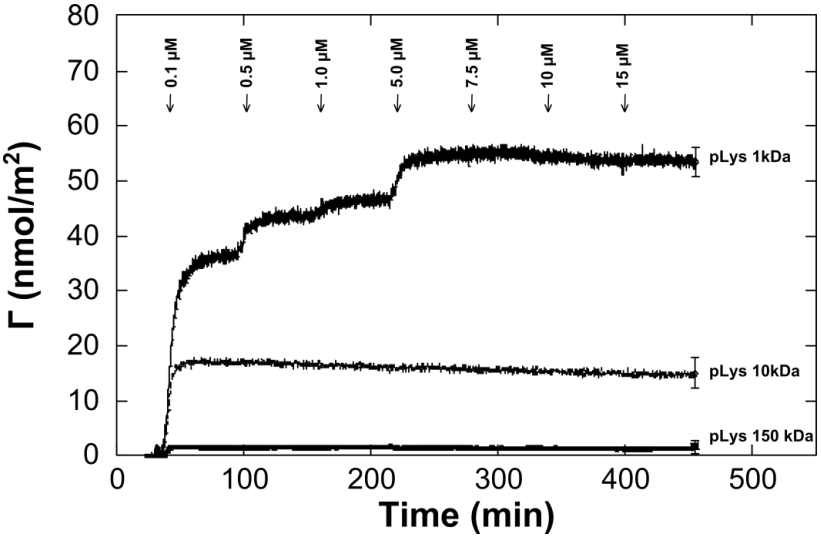

Figure S4. Effect of microgel charge density on peptide binding (pLys 10 kDa, 7.5µM in 10mM Tris HCl at pH 7.4). Results are shown for confocal fluorescence intensity of Alexa 488-labeled pLys and non-labeled pLys adsorbed amount obtained using ellipsometry.

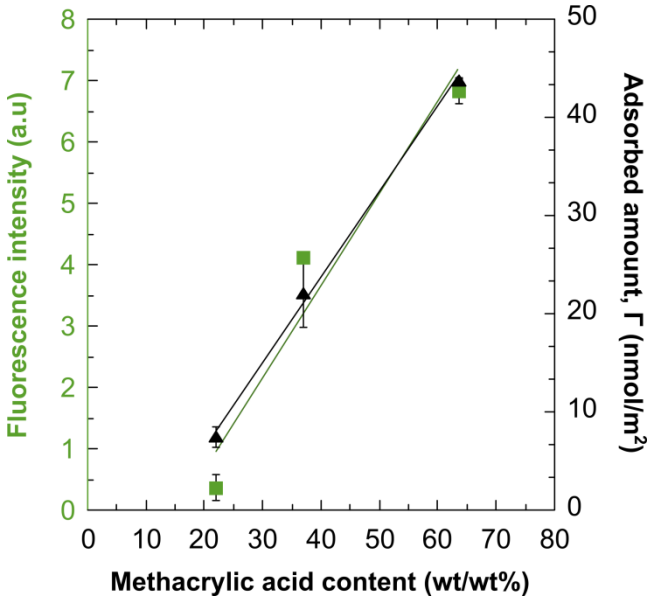

Figure S5. Peptide desorption kinetics from dispersed MAA33 microgels in 10 mM Tris, pH 7.4. The microgels were first loaded with the peptides indicated at 75 μM overnight. Just prior to measurement, microgels were diluted 100 times by 10 mM Tris, pH 7.5, after which the zeta potential was measured over time, the first measurement point representing a time of 30-60 s after dilution.

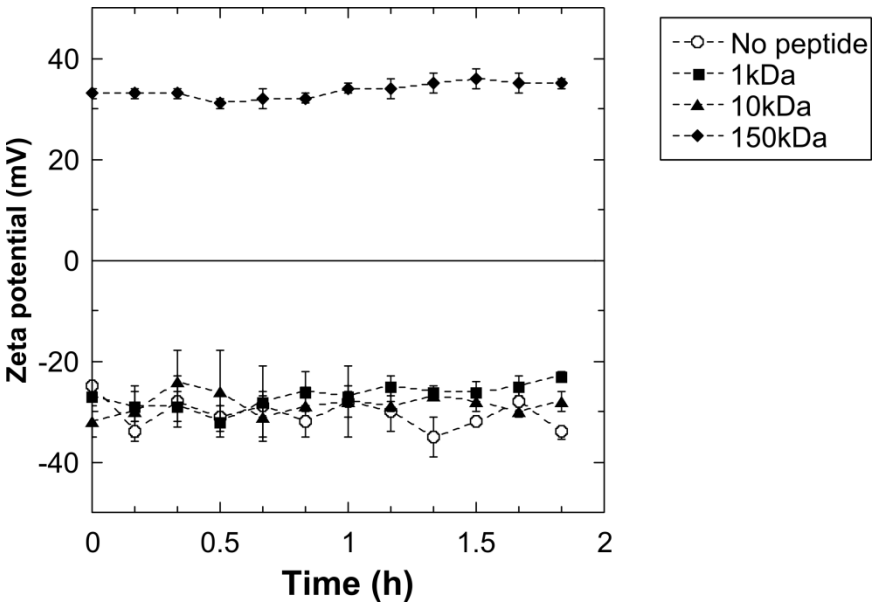

Figure S6. Peptide-induced deswelling of dispersed MAA33 microgels. Measurements were performed in 10 mM Tris buffer, pH 7.4, in the presence of 0.01 μM of the indicated peptides and after an overnight loading equilibration.