Supporting Information

Self-Regulating Colloidal Co-Assemblies That Accelerate Their Own Destruction via Chemo-Structural Feedback

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1. **Abbreviations**

| Abbreviation | Description |
|--------------|-------------|
| AZ           | AZ-PEG-NH₂  |
| CD           | 3A-Amino-3A-deoxy- (2A S, 3A S)-α-cyclodextrin |
| DMEDA        | N, N-dimethylethylenediamine |
| MG           | Parent microgel |
| MG₉(Me)₂      | Parent microgel functionalized with DMEDA |
| MGₐZ         | Parent microgel functionalized with AZ |
| MGₐD         | Parent microgel functionalized with CD |
| MGₐZ(Me)₂     | MGₐZ functionalized with DMEDA |
| MGₐD(Me)₂     | MGₐD functionalized with DMEDA |
| INV          | Invertase |
| GOX          | Glucose Oxidase |
| MGₐZ,GOX     | MGₐZ functionalized with GOX |
| MGₐD,INV     | MGₐD functionalized with INV |
| MGₐZ(Me)₂,GOX| MGₐZ(Me)₂ functionalized with GOX |
| MGₐD(Me)₂,INV| MGₐD(Me)₂ functionalized with INV |
2. Materials

All chemicals and reagents are purchased from Sigma Aldrich or Merck and used without further purification unless otherwise stated:

2,2,2-trifluoroethyl methacrylate (ABCR GMBH, 99 %), N-isopropylacrylamide (97 %), 2’,2’-azobis(2-methylpropionamidine) dihydrochloride (ABCR GMBH, 96 %), divinylbenzene (80 %), acryloxyethyl thiocarbamoyle Rhodamine B (POLYSCIENCES, INC.), Silica60 (0.04 - 0.063 mm, MACHEREY-NAGEL GMBH CO. KG), N,N’-methylenebis(acrylamide) (99%), Glycidyl methacrylate (>97%), Boc-TOTA (IRIS BIOTECH GMBH), Al₂O₃ 90 neutral (CARL ROTH GMBH), EDC (ABCR GMBH, 98 %), trifluoroacetic acid (ABCR GMBH, 99 %) and NaHCO₃ (CARL ROTH GMBH, >99.5 %), CDCl₃ (Euriso-top, 99.80 %), dichloromethane (VWR INTERNATIONAL, >99 %), ethyl acetate (VWR INTERNATIONAL, >99 %), MeOH (FISHER CHEMICALS, 99.8 %), cyclohexane (VWR INTERNATIONAL, >99 %), 3A-Amino-3A-deoxy- (2A S, 3A S) -α-cyclodextrin (TCI, >90.0%), Atto 488 amine (ATTO-TEC GMBH), Azobenzene-4-sulfonyl chloride (TCI, ≥98%), Triethylamine (≥99.5%), citric acid monohydrate (CA, BioXtra, ≥99.5%), trisodium citrate dihydrate (Na₃C, BioXtra, ≥99.0%), sodium dihydrogen phosphate (BioXtra, ≥99.0%), disodium hydrogen phosphate (BioXtra, ≥99.0%), disodium hydrogen phosphate (BioXtra, ≥99.0%), (Tris, BioXtra, ≥99.9%), hydrochloric acid (36.5-38.0%, bioreagent), N, N-dimethylethylenediamine (95%), invertase from baker’s yeast (S. cerevisiae) (Inv, grade VII, ≥300 units/mg solid), glucose oxidase from Aspergillus niger (GOX, Type VII, lyophilized powder, ≥100,000 units/g solid), sucrose (≥99%), glucose (BioXtra, ≥99.5%). Activity assay kits of invertase and glucose oxidase were bought from BioVision (Catalog #K674-100). MilliQ water was used throughout all experiments.

3. General Characterization Methods and Instruments

DLS measurements were performed on the LS Instruments NanoLab 3D at 25 °C operating with a red laser (λ = 685 nm) and a scattering angle of Θ = 90 ° using standard disposable PS cuvettes (BRAND GmbH & Co. KG). The distributions of the hydrodynamic radii were obtained by a CONTIN mode analysis.

ζ-potential of microgels was measured using a Zetasizer Nano-ZS (Malvern Panalytical GmbH) at 25 °C. All samples were measured in triplicate in disposable folded capillary cells. pH was maintained using a 907 Titrando auto-titrator (Metrohm, Switzerland), and titrations were performed using 0.1 M NaOH and 0.1 M HCl at 25 °C.

Confocal laser scanning microscopy (CLSM) was performed on Leica Stellaris 5 microscope with four laser lines and three HyD S detectors using plan-apochromat objectives (63×, 1.40 numerical aperture, oil immersion).

The evolutions of the pH profiles were recorded on a 12-channel pH monitoring system by EA Instruments at 25 °C. Electrodes were calibrated by calibration buffers (pH= 4, 7, 10) each time before using them.

The temperature-controlled fluorescence measurements were performed on a TECAN (Spark model) microplate plate reader using Costar® 96-Well black polystyrene plate.

p values are calculated by performing a t-Test (Two-Sample Assuming Equal Variances) in Microsoft excel using a built-in Data analysis tool pack.
4. Methods

4.1 Synthesis of surfactant-free, poly(2,2,2-trifluoroethyl methacrylate)-Rhodamine labeled core particles:

Divinylbenzene (DVB) and 2,2,2-trifluoroethyl methacrylate (tFMA) were purified using column chromatography (Al₂O₃, neutral). The initiator 2,2’-azobis(2-methylpropionamide) dihydrochloride (V50, 160 mg, 590 μmol) was dissolved in deionized water (44 mL), degassed (25 min) and thermostated at 70 °C for 15 min. To a solution of DVB (15.4 mg, 118 μmol, 1 mol %) and TFMA (1.98 g, 11.8 mmol), a solution of acryloxyethyl thiocarbamoyl Rhodamine B (1.5 mg) and N-isopropylacrylamide (NIPAM, 150 mg, 1.3 mmol) in water (4.3 mL) were added. The mixture was ultrasonicated for 2 minutes and degassed (10 min). The resulting mixture was added dropwise (over a period of 5 min) to the initiator solution starting the polymerization. The reaction mixture was stirred at 70 °C for 6 h (stirring rate = 600 rpm). The resulting dispersion was filtered while hot and dialyzed against deionized water (MWCO 8000 Da), and dispersion with a solid content of 35.6 mg/mL was observed upon freeze-drying.

4.2 Synthesis of the PNIPAM shell to yield PtFMA-Rhodamine labelled-core-PNIPAM-co-GMA-shell microgel (MGs) particles:

NIPAM (1.4g, 12.8 mmol, 80.5 wt %) was dissolved in deionized water (100 mL) together with the cross-linker N,N’-methylenebis(acrylamide) (MBA, 12.6 mg, 81.7 μmol, 1.0 wt %). The core particles (4.7 mL, 35.6 mg/mL, 166 mg solids content, 9.5 wt %) were added and the mixture was degassed for 30 min and heated to 72 °C. The initiator V50 (75.9 mg, 0.3 mmol) was dissolved in water (30 mL) and degassed (15 min). The polymerization was initiated by dropwise addition of initiator solution to heated reaction mixture and stirred (stirring rate = 350 rpm). After 10 minutes, degassed Glycidyl methacrylate (GMA, 162 mg, 150.7 µL, 1.1 mmol, 9 wt %) after purification by column chromatography (Al₂O₃, neutral) was added dropwise into the reaction mixture. The stirring rate was increased to 600 rpm and reaction was allowed to run for 4 hours. The resulting core/shell MG particles (amount of GMA moieties assuming full conversion = 633.8 μmol per g of MG) were filtered while hot and purified by dialysis (MWCO 8000 Da) against deionized water. The resulting core-shell MGs were further purified via centrifugation (5 x 25 min, 11000 rpm, 15 °C replacement of the supernatant with Milli Q water per centrifugation step).

4.3 Synthesis of Guest, AZ-PEG-NH₂:

\[
\text{N} \quad \text{N} \quad \text{O} \quad \text{S} \quad \text{O} \quad \text{H₂N} \quad \text{O} \quad \text{O} \\
\text{O} \quad \text{O} \quad \text{N} \quad \text{N} \quad \text{O} \quad \text{H₂N} \quad \text{O} \quad \text{O}
\]

Step I: Azobenzene-4-sulfonyl chloride (1.3 g, 4.6 mmol) in DCM (dichloromethane) solution (65 mL) was dropped slowly (0.5 h) to a vigorously stirred 165 mL DCM solution containing 1-((t-Butyloxycarbonyl-amino)-4,7,10-trioxa-13-tridecanamine (Boc-TOTA, 3.0 g, 9.3 mmol, 2 eq.) and...
triethylamine (TEA, 0.5 g, 0.6 mL, 4.6 mmol) mixture in an ice bath, and then stirred at room temperature overnight. Subsequently, DCM was removed by a rotary evaporator, and the residue was dissolved into ethyl acetate. The solution was washed with a saturated NaHCO$_3$ solution, and the solvent was removed under reduced pressure. The resulting oil (AZ-PEG-NHBoc) was purified using column chromatography (SiO$_2$, DCM/MeOH, 8:2, $R_f = 0.8$), and the product (2.3 g) was obtained as an orange oil. $^1$H NMR (300 MHz, DMSO-d$_6$): d (ppm) = 7.96 (4H); 7.89 (2H); 7.48 (3H); 5.75 (1H); 4.85 (1H); 3.59 (6H); 3.47 (6H); 3.11 (4H); 1.67 (4H) and 1.35 (9H).

Step II: AZ-PEG-NHBoc (2.3 g, 4.1 mmol) was dissolved in DCM (12 mL), and the solution was cooled to 0 °C. Trifluoroacetic acid (TFA, 11.8 g, 103.6 mmol, 25 eq.) was added dropwise and the solution was stirred at RT for 3 h. Afterward, the solvent was removed under reduced pressure and the resulting oil was dissolved in water (40 mL) and washed with ethyl acetate (3 × 10 mL). The aqueous solution was freeze-dried, and the desired product was obtained as a deep orange liquid. $^1$H NMR (300 MHz, DMSO-d$_6$): d (ppm) = 8.01 (6H); 7.58 (3H); 6.82 (1H); 3.66 (12 H); 3.26 (2H); 3.13 (2H); 2.01 (2H) and 1.62 (2H). HRMS (ESI) calculated for [M + H]$^+$ C$_{22}$H$_{33}$N$_4$O$_5$S 465.2166, found 465.2165.

4.4 Functionalization reactions on parent MG

4.4.1 Functionalization of parent MG with $N$-$N$-dimethylethylenediamine (DMEDA) to produce MG$_{N(Me)_2}$.

First, 34.1 mg of DMEDA (ca. 10-fold excess with respect to GMA moieties in the MG shell) was dissolved in 5 mL of N-ethylmorpholine (NEM) buffer (200 mM, pH = 8.2). To this, 5 mL of parent MG suspension (12.2 mg/mL) was added. The mixture was stirred overnight at room temperature. The resulting modified core-shell MGs, MG$_{N(Me)_2}$ were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C replacement of the supernatant with 5 mL Milli Q water per centrifugation step).

4.4.2 Sequential functionalization of parent MG to produce MG$_{AZ}$ and MG$_{AZ/N(Me)_2}$.

First, 215.7 mg of AZ-PEG-NH$_2$ (ca. 12-fold excess with respect to GMA moieties in the MG shell) was dissolved in 5 mL of N-ethylmorpholine (NEM) buffer (200 mM, pH = 8.2). To this, 5 mL of parent MG suspension (12.2 mg/mL) was added. The mixture was stirred overnight at room temperature. The resulting modified core-shell MGs, MG$_{AZ}$ were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C replacement of the supernatant with 5 mL Milli Q water per centrifugation step). The change in the color of the precipitated MG from light pink (from Rhodamine B in the cores) to orange/peach after functionalization with AZ provides macroscopic proof for the successful linkage (Figure S3). 2.5 mL of MG$_{AZ}$ was reserved for further experiments, and the rest 2.5 mL was further functionalized with DMEDA (ca. 10-fold excess with respect to GMA moieties in the MG shell) as described in section 4.4.1 to produce MG$_{AZ/N(Me)_2}$.

4.4.3 Sequential functionalization of parent MG to produce MG$_{CD}$ and MG$_{CD/N(Me)_2}$.

First, 451.5 mg of 3A-Amino-3A-deoxy- (2A $S$, 3A $S$) -$\alpha$-cycloaddextrin ($\alpha$-CD, ca. 12-fold excess with respect to GMA moieties in the MG shell) was dissolved in 5 mL of N-ethylmorpholine (NEM) buffer (200 mM, pH = 8.2). To this, 5 mL of parent MG suspension (12.2 mg/mL) was added. The mixture was stirred overnight at room temperature. The resulting modified core-shell MGs, MG$_{CD}$ were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C replacement of the supernatant with 8 mL Milli Q water per centrifugation step). 2.5 mL of MG$_{CD}$ was reserved for further experiments, and the rest 2.5 mL was further functionalized with DMEDA (ca. 10-fold excess with respect to GMA moieties in the MG shell) as described in section 4.4.1 to produce MG$_{CD/N(Me)_2}$. There is no color change in precipitates of MG$_{CD}$ since $\alpha$-CD is a colorless solid.
For preparing CLSM samples, MG$_{CD}$ was functionalized with Atto-488-amine in the shell. 0.1 equivalent of Atto-488-amine (4 mg/mL, stock solution prepared in DMSO) with respect to GMA moieties in the MG shell was dispersed in PBS buffer (pH = 7.4), to which respective MG suspension was added. The reaction mixture was stirred overnight at 25°C. The resulting dye-labeled core-shell MGs were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C replacement of the supernatant with Milli Q water per centrifugation step). Further functionalization reaction with DMEDA was performed as described in Section 4.4.1.

It is important to mention that an exact quantification of functionalizations at each step is hardly feasible due to the small contents and overlap in elemental analysis or spectroscopic techniques. However, all reaction protocols maintain 12 equivalents of CD/AZ pairs and 10 equivalents of DMEDA with respect to GMA in the MG shell to achieve identical functionalizations.

4.5 Enzyme Immobilization:

4.5.1 Immobilization of Invertase (INV) on MGs (MG).\[1\]

The stock solution of INV (2 g/L) was freshly prepared as needed in the citric acid/sodium citrate (CA/Na$_3$CA) buffer (1.5 mM, pH = 5.5). Enzyme functionalization was done on core-shell beads, bearing epoxide groups in their shell. First, 5 mg of the INV was dissolved in 3 mL of respective buffer. Next, 1 mL of MG suspension (12.2 mg/mL) was added to 2 mL of respective buffer. To this mixture, 1 mL of enzyme solution was added, and the reaction was stirred overnight at room temperature. The functionalized core-shell MGs were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C, replacement of the supernatant with Milli Q water per centrifugation step).

4.5.2 Immobilization of Glucose Oxidase (GOX) on MG.\[2\]

A three-step reaction achieves the spacer-mediated immobilization of GOX on MGs. In the first step, a spacer arm was introduced on the MGs for which 1 mL of MG suspension was stirred in phosphate buffer solution (PBS, 10 mM, pH = 7.4) containing 0.1 equivalent of ethylenediamine (with respect to GMA groups on MGs) overnight at 25°C. After the completion of reaction, MGs were purified via centrifugation (3 x 10 min, 11000 rpm, replacement of the supernatant with PBS buffer per centrifugation step). Next, activation of carboxyl acid groups in GOX (6 mg) was carried out by stirring...
with 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide hydrochloride (EDC, 1.5 mg) in 2.4 mL of 2-(N-morpholino) ethanesulfonic acid buffer (MES, 10 mM, pH = 4.5) for 25 minutes at 25 °C. Finally, 1.2 mL solution of activated enzyme is added to aminated MGs prepared in first step and mixture is stirred overnight at 25 °C. The GOX immobilized MGs were purified via centrifugation (3 x 10 min, 11000 rpm, 15 °C, replacement of the supernatant with Milli Q water per centrifugation step).

4.6 Activity determination of immobilized enzymes by fluorometric assay

4.6.1 INV activity measurement

INV activity assay kit from BioVision (Catalog #K674-100) was adopted to measure the activity of immobilized INV, MCD,INV and MCD/N(Me)2,INV. We followed the protocol of the assay kit but did not take background control as it was insignificant for our samples. The assay kit has the following components: INV hydrolysis buffer, INV assay buffer, INV stop solution, OxiRed™ probe in DMSO, INV enzyme mix, INV substrate, glucose standard (100 mM), and positive control.

Calibration curve: 1 mM glucose standard solution was prepared by diluting 10 μL 100 mM glucose standard with 990 μL MilliQ water. 1mM glucose standard solution was further diluted with MilliQ water to make 0.1mM stock solution of glucose standard. 0, 2, 4, 6, 8, and 10 μL of 0.1 mM glucose standard solution was pipetted to a 96-well plate, INV hydrolysis buffer was added to adjust the total volume to 50 μL. 10 μL INV stop solution, 36 μL INV assay buffer, 2 μL INV enzyme mix, and 2 μL probe were added to the glucose solution to make a total volume of 100 μL. The well plate was incubated at 25 °C for 30 min in dark place, then the fluorescence (Ex/Em = 535/585 nm) was measured with a plate reader. Zero standard record was subtracted from all records and calibration curves were plotted. The calibration curve is shown in Figure S5 (a).

INV activity measurement: i) Preparation of INV sample: 250 μL of respective MG suspension (0.2 mg/mL) was centrifuged (5min, 11000 rpm) and the Milli Q water was replaced with INV hydrolysis buffer to make a final volume of 40 μL. Positive control of INV sample: 4 μL positive control solution was diluted with 36 μL hydrolysis buffer. ii) Substrate hydrolysis: 10 μL of INV substrate was added into wells containing INV sample and positive control. Then the plate was incubated at 25 °C (same as the temperature at which all experiments in the main text were carried out) for 30 min. After the incubation, 10 μL of INV stop solution, a mixture of 36 μL assay buffer, 2 μL INV enzyme mix, and 2 μL probe was sequentially added to each well, including INV sample and positive control. The well plate was incubated at 25 °C for 30 min in a dark place, then fluorescence (Ex/Em = 535/585 nm) was measured with a plate reader. iv) Calculation: Zero standard record was subtracted from all records and the calibration curve was plotted. The fluorescence record of INV sample was applied to calibration curves to get the amount of glucose generated by sample INV during the incubation at 25 °C. The activity of sample INV was calculated using the following equation:

\[
\text{Activity} = \frac{n}{(m \times t)} \text{nmol/} (\text{min*mg})
\]

where: \(n\) is the amount of glucose generated by sample INV (nmol); \(m\) is the amount of sample INV added into the well plate (mg); \(t\) is the incubation time at 25 °C (min). Figure 3c in the main text presents the activity of MCD,INV and MCD/N(Me)2,INV.

4.6.2 GOX activity measurement

GOX activity assay kit from BioVision (Catalog #K788-100) was used to measure the activity of immobilized GOX, MGOX and MGOX/N(Me)2,GOX. We followed the assay kit protocol but did not take background control as it was insignificant for our samples. The assay kit has the following components:
GOX assay buffer, OxiRedTM probe, GOX substrate, GOX developer, GOX positive control, and H$_2$O$_2$ standard (0.88 M).

Calibration curve: 10 μL of 0.8 M H$_2$O$_2$ standard was added to 870 μL MilliQ water to make an H$_2$O$_2$ solution with a concentration of 10 mM. Then 5 μL 10 mM H$_2$O$_2$ solution was diluted with 45 μL assay buffer to a concentration of 1 mM. 1mM glucose standard solution was further diluted with MilliQ water to make 0.1mM stock solution of H$_2$O$_2$ standard. 0, 2, 4, 6, 8, and 10 μL of 0.1 mM H$_2$O$_2$ standard solution was pipetted to a 96-well plate, and assay buffer was added to each well to adjust the volume to 50 μL. A mixture of 36 μL assay buffer, 2 μL GOX developer, 2 μL OxiRedTM probe, and 10 μL GOX substrate was added to the H$_2$O$_2$ solution and homogeneously mixed. The plate was incubated at 25 °C for 5 min and then the fluorescence (Ex/Em = 535/585 nm) was measured with a plate reader. The zero standard record was subtracted from all records and the calibration curve was plotted. Calibration curves is shown in Fig S5 (b).

GOX activity measurement: i) Preparation of GOX sample: 250 μL of respective MG suspension (0.2 mg/mL) was centrifuged (5min, 11000 rpm) and the Milli Q water was replaced with GOX assay buffer to make a final volume of 40 μL. Positive control: 4 μL of positive control solution was diluted with 46 μL GOX assay buffer. ii) Kinetics measurement: A mixture of 36 μL assay buffer, 2 μL GOX developer, 2 μL OxiRed™ probe, and 10 μL GOX substrate was added to GOX sample and positive control, and homogeneously mixed. The well plate was incubated at 25 °C for 30 min. The fluorescence (Ex/Em = 535/585 nm) of GOX sample and positive control was measured after incubation at 25 °C. iii) Calculation: Zero standard record was subtracted from all records and calibration curve was plotted. The fluorescence record of GOX sample was applied to calibration curves to get the amount of H$_2$O$_2$ generated by sample GOX during the incubation at 25 °C. The activity of GOX sample can be calculated using the following equation:

$$\text{Activity} = \frac{n}{(m*t)} \text{nmol/(min*mg)}$$

where: n is the amount of H$_2$O$_2$ generated by GOX sample (nmol); m is the amount of GOX sample added into the well plate (mg); t is the incubation period at 25 °C (min). Figure 3d in main text presents the activity of MG$_{AZ,GOX}$ and MG$_{AZ/N(Me)2,GOX}$.

4.7 Qualitative assessment of immobilized enzymes by pH assay

4.7.1 pH assay for INV

250 μL (0.2 mg/mL) of INV immobilized MG (MG$_{CD,INV}$ and MG$_{CD/N(Me)2,INV}$) was first diluted with 250 μL of MilliQ water. To this suspension, 750 μL of CA/Na$_3$CA buffer (1.5 mM, pH = 3.5) containing 0.2 mg of GOX was added. pH-assay was initiated by addition of 500 μL of tris buffer (20mM, pH 8.8) containing 200 mM sucrose. pH profiles are recorded to observe any change in the pH (Figure 3e in main text). All pH-profiles are an average of two measurements recorded simultaneously.

4.7.2 pH assay for GOX

250 μL (0.2 mg/mL) of GOX immobilized MG (MG$_{AZ,GOX}$ and MG$_{AZ/N(Me)2,GOX}$) was first diluted with 250 μL of MilliQ water. To this suspension, 750 μL of CA/Na$_3$CA buffer (1.5 mM, pH = 3.5) was added. pH-assay was initiated by addition of 500 μL of tris buffer (20mM, pH 8.8) containing 200mM sucrose. pH profiles are recorded to observe any change in the pH (Figure 3f in main text). All pH-profiles are an average of two measurements recorded simultaneously.
5. Supporting Figures

Figure S1. pH-dependent enzyme activities.\textsuperscript{[3]} Both INV (a) and GOX (b) have bell-shaped pH-dependent activity curves with weak activity at high pH (8-9), which increases rapidly as the pH decreases below 7. Thereafter, the activity remains nearly constant until pH 4, and decreases again at lower pH. Therefore, the formation of gluconic acid during the deactivation pathway first exerts a positive feedback loop, self-accelerating the metabolic chain of the EC, and ultimately provides a negative feedback loop to the enzymatic activity, flattening off the pH response.

Figure S2. DLS CONTIN plots of: poly(2,2,2-trifluoroethyl methacrylate) (PtFMA) core, parent MG, MG$_{N(Me)_2}$, MG$_{AZ}$, MG$_{AZ/N(Me)_2}$, MG$_{CD}$, MG$_{CD/N(Me)_2}$. Due to its hydrophobicity, azobenzene when embedded in microgels (MG$_{AZ}$) leads to water expulsion and subsequent shrinking exhibiting lower z-average radius, $<R_a>_z$ than unmodified MG. The suspensions were prepared in MilliQ water with a final concentration of 0.05 mg/mL.
**Figure S3.** Photographs of the centrifuged samples. (a) Parent MG and (b) MG$_{AZ}$, obtained after functionalization of parent MG with AZ-PEG-NH$_2$. The change in the color of the suspensions from pink to orange/peach is a macroscopic proof for a successful linkage.

**Figure S4.** pH-dependent surface ζ-potential values of MG$_{N(Me)2}$. The electrostatic repulsions decrease upon increasing the pH from 3 to 9. The error bars represent the standard deviation from three independent measurements.

**Figure S5.** Calibration curves of: (a) glucose standard solutions and (b) H$_2$O$_2$ solutions.
Figure S6. pH-dependent CLSM images of 1:1 mixture of MGCD/N(Me)₂ and MGAZ/N(Me)₂. Co-assemblies only occur at pH 8.0 and disassemble reversibly at acidic pH. Scale bar, 3 µm. The final concentration of MGs in the suspensions is 0.0064 wt%.

6. References

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