Supporting Information

1. Materials

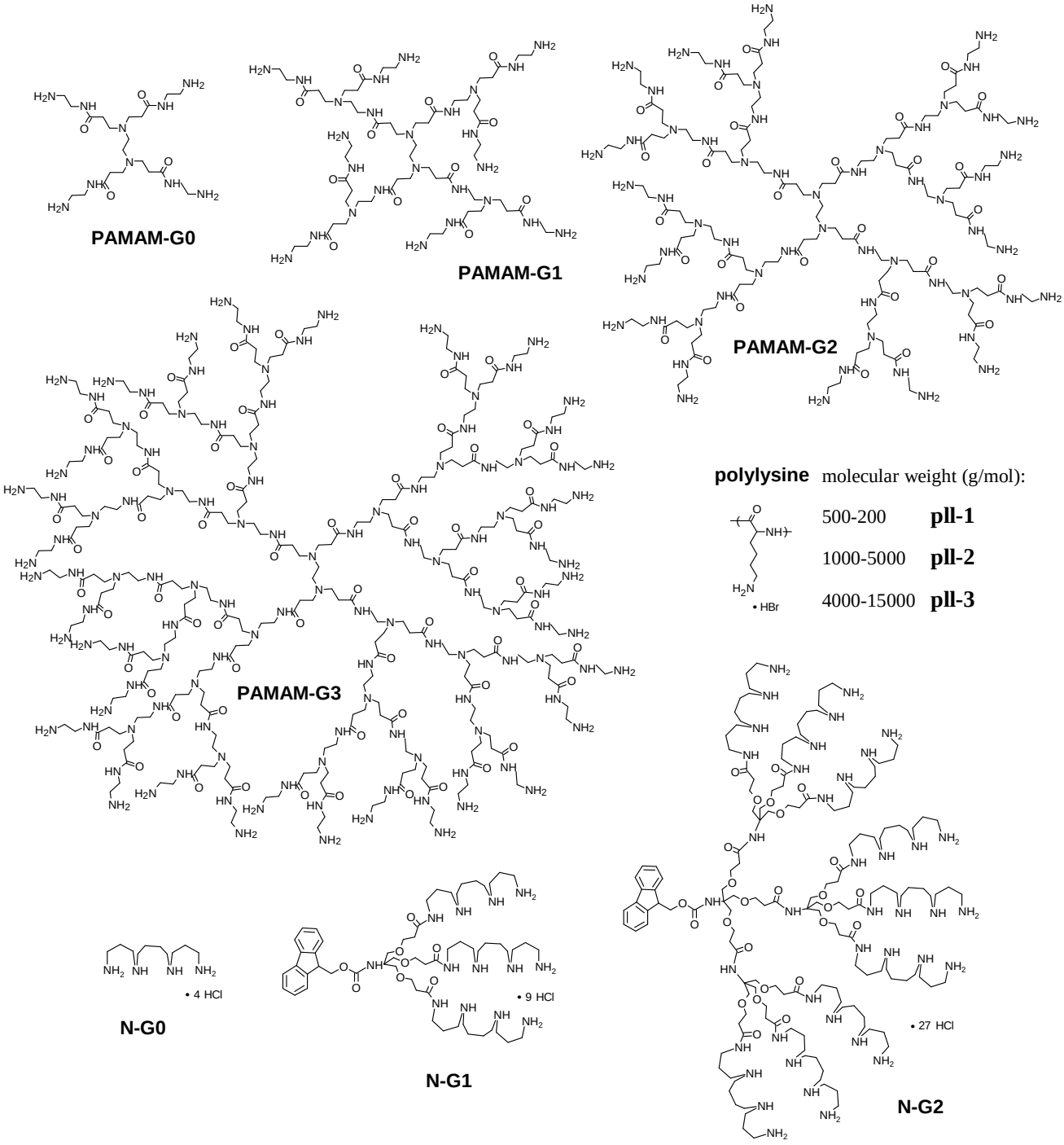

Figure S1. Structures of the different polymers studied in this work.

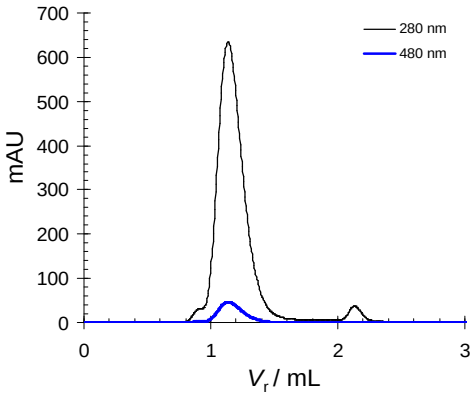

Figure S2. FPLC chromatogram of the Prussian blue particles in capsids after irradiation and gel column. The main peak with V_r = 1.15 mL corresponds to the elution volume of the native virus. FPLC fractions show a distinct blue colour between V_r 1.0–1.4 mL indicating the presence of Prussian blue.

2. Dynamic light scattering at 10 mM NaCl

DLS measurements were carried out as described in the experimental section. The c_{50} values obtained from the DLS are presented here as the "charge concentration" presented in Table S1

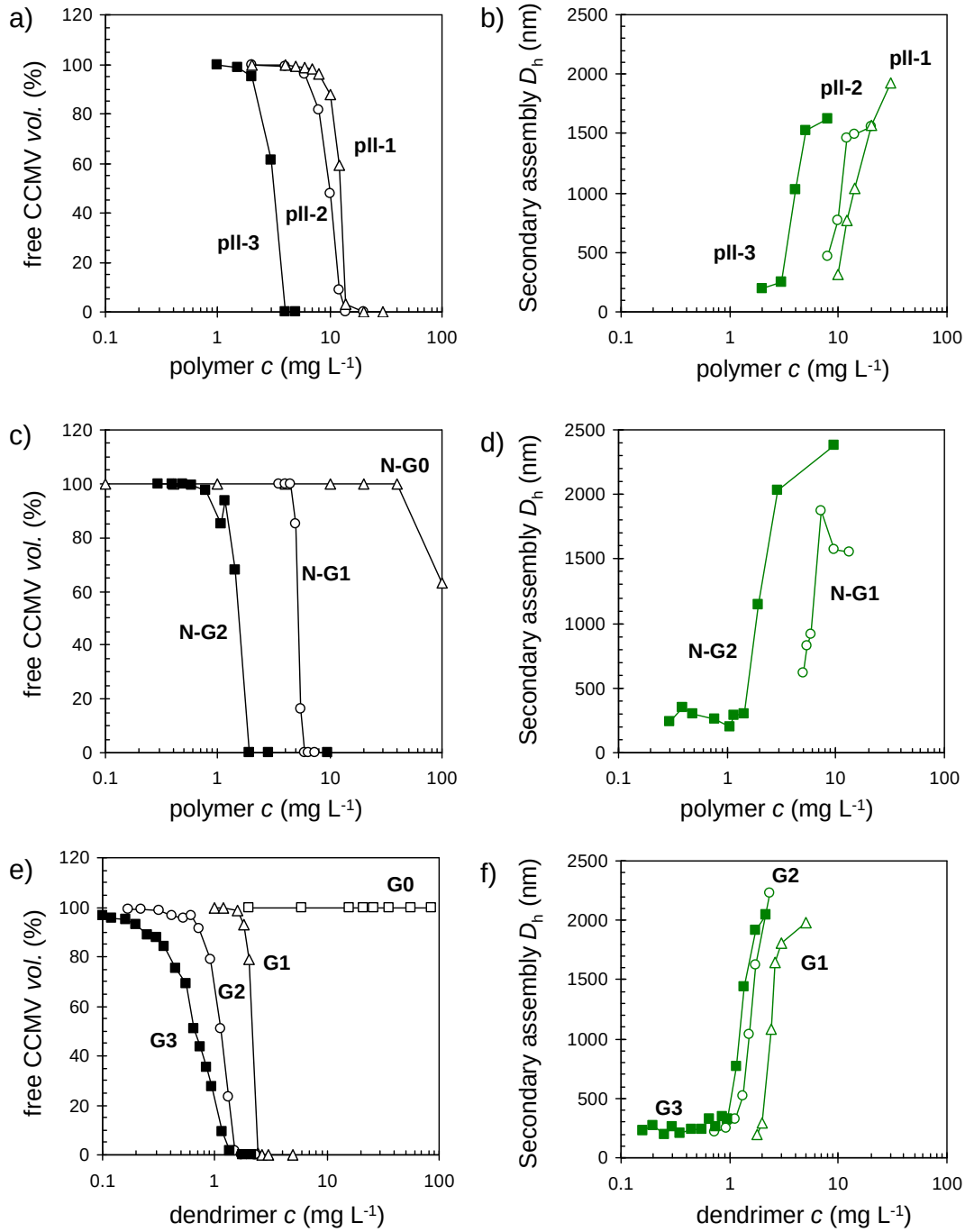

Figure S3. DLS data for the titrations of CCMV with a,b) poly(L-lysine), c,d) Newkome-dendrons and e,f) PAMAM dendrimers at 10 mM NaCl concentration. Figures show a decrease in the volume averaged scattering intensity of the free virus (left) and formation of a larger secondary assemblies (right).

Table S1. Virus binding data extracted from DLS measurements and normalized to the nominal charge concentration.

compound	M_w	nominal charge (ε)	ε_{50} [mM] [10 mM NaCl]	ε_{50} [mM] [150 mM NaCl]
pll-1	1250	8+	80.0	549
pll-2	3000	18+	58.8	477
pll-3	9500	58+	20.1	13.4
N-G0	348.2	4+	>1149	>1149
N-G1	1112.5	9+	46.1	>809
N-G2	3176.4	27+	13.6	92.7
PAMAM-G0	516.7	4+	>774	>774
PAMAM-G1	1429.9	8+	12.3	>559
PAMAM-G2	3256.2	16+	5.4	32.9
PAMAM-G3	6908.8	32+	3.1	3.9

ε_{50} represents the "concentration of positive charges" required to assemble 50% of the free CCMV as observed by volume averaged scattering intensity and is calculated as $\varepsilon_{50} = \frac{\varepsilon \times c_{50}}{M_w}$

Charge normalized values are clearly not linearly dependent on the number charges at 150 mM NaCl. For example the ε_{50} values for PAMAM dendrimers reduce by approximately an order of a magnitude when the number of charges is doubled indicating that the gained affinity increase is more than a simple sum of the binding ligands.

3. Stability of the virus-polymer complexes over time

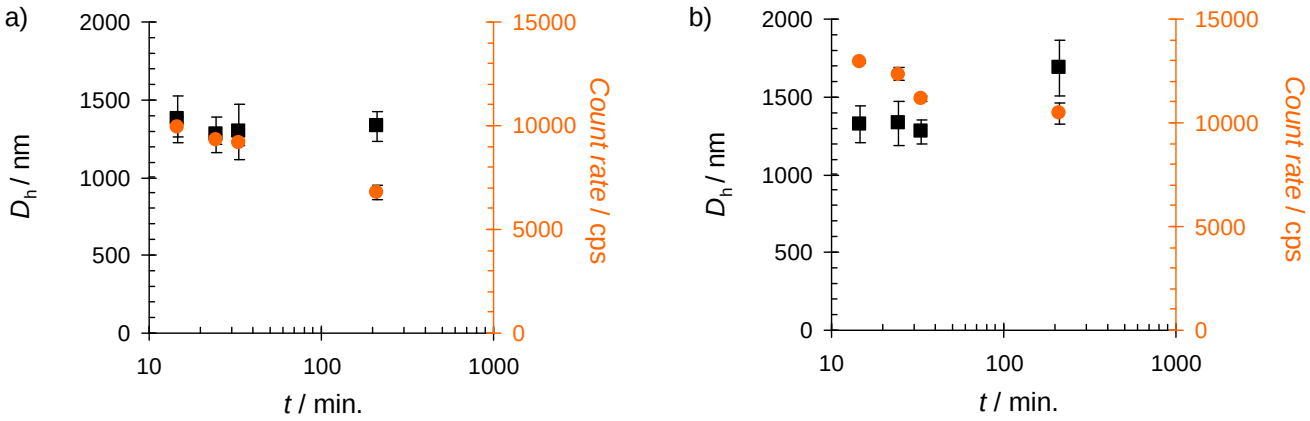

Figure S4. Changes in the volume-average hydrodynamic diameter (primary axis, black squares) and derived count rate (secondary axis, orange spheres) over time after complexing the CCMV (45 mg L^{-1}) with (a) **PAMAM-G3** (2 mg L^{-1}) or (b) **pll-3** (5 mg L^{-1}) at the presence of 150 mM NaCl. Complexes form rapidly in approximately 10 minutes and are clearly stable up to 200 min.

4. Small Angle X-ray Scattering (SAXS)

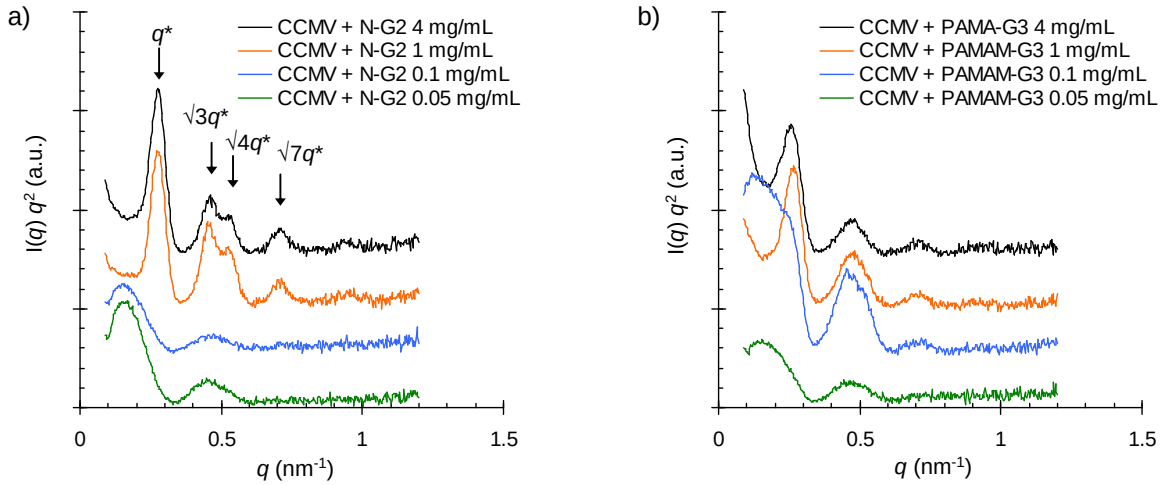

Figure S5. SAXS scattering curves of CCMV-polymer complexes. a) CCMV complexed with an increasing amount of **N-G2**. At low concentrations of the polymer (0.05 and 0.1 mg mL^{-1}) hexagonal packing is not observed. However, when the dendron concentration reaches 1 mg mL^{-1} a hexagonal arrangement (peak position ratios $q^n/q^* \approx \sqrt{1}:\sqrt{3}:\sqrt{4}:\sqrt{7}$) can clearly be observed. b) CCMV-**PAMAM-G3** complexes at the same polymer concentration do not show clear hexagonal packing.

5. Assembly of the Prussian blue-loaded capsids

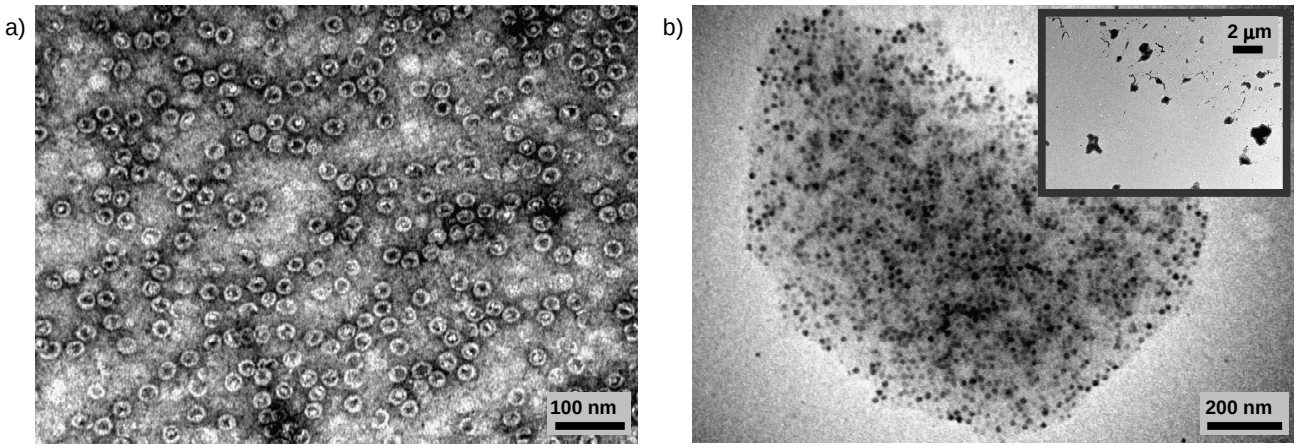

Figure S6. Full-size TEM images of Figure 7b,c (main text). a) Free PB-loaded capsids stained with uranyl acetate. b) PB-loaded capsids complexed with **PAMAM-G3**. No staining applied and consequently the electron-dense iron cores of the particles are clearly visible. Low-magnification image (inset) shows the micrometer–sized complexes.