**Supplementary Information** 

# Stepwise DNA Condensation by a Histone-mimic Peptide-coated $M_{12}L_{24}$ Spherical Complex

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## General

AFM images on a mica plate (PELCO® Mica Discs with a diameter of 9.9 mm purchased from Ted Pella, Inc.) were collected on an Asylum Research MFP-3D-SA-J microscope. DLS experiments were conducted using a Dynapro Model-MSXTC (Protein Solutions, Inc.) equipped with a 58 mW laser and a wavelength of 825.9 nm. Plasmid pBR322 DNA (4363 bps) was purchased from Takara Bio, Inc. as an aqueous solution (500 ng· $\mu$ L<sup>-1</sup>) containing 10 mM of Tris·HCl (pH 8.0) and 1 mM of EDTA.

### **AFM Measurements**

A DMSO solution of sphere 1 was diluted to a target concentration with Milli-Q water, and was added to an aqueous solution of plasmid pBR322 DNA with a final DNA concentration of 0.75 ng· $\mu$ L<sup>-1</sup>. Then 10  $\mu$ L of the resultant mixture was applied to freshly cleaved mica surface, and incubated at room temperature for ca. 10 min. After the sample solution was concentrated by natural drying until no droplets could be seen, the mica plate was rinsed with water (50  $\mu$ L × 4), and dried up with N<sub>2</sub> flow and following evacuation.

The microscope was operated in AC mode on ambient conditions at room temperature, using silicon cantilevers (OMCL-AC200TS-C3, Olympus) with resonance frequencies of approximately 150 kHz, a force constant of ca. 9 N/m, a tip height of 16 nm, and mean tip radii of 7 nm. The images were obtained at scan rate of 0.8 Hz over a 2000 nm × 2000 nm scan area with 512 points × 256 lines with a scan angle of 90°. Obtained images were analyzed on IGOR Pro software ver 6.1.2.1 (WaveMetrics, Inc.).

## **DLS Measurements**

A DMSO solution of sphere **1** was diluted to a target concentration with Milli-Q water, and was added to an aqueous solution of plasmid pBR322 DNA with a final DNA concentration of 0.75 ng· $\mu$ L<sup>-1</sup>. Then 12  $\mu$ L of the resultant mixture was transferred into a quartz cuvette with an optical length of 1.5 mm, and the scattered light was detected at 25 °C at an angle of 90° to the incident beam. Data analysis was performed using a Dynamic version 5 software package (Protein Solutions, Inc.).



## **AFM Image**

Figure S1. (a) Representative AFM image of sphere 1 on a mica surface (image size is  $2000 \text{ nm} \times 2000 \text{ nm}$ ). (b) Cross-section profile along the red line in figure (a).