

## Description of Additional Supplementary Files

### File name: Supplementary Movie 1

Description: PolyP-Mg<sup>2+</sup> condensates form spherical droplets that fuse over time. (1mg/mL polyP, 10% polyP-AF647, 100 mM MgCl<sub>2</sub>, 50mM HEPES, pH 7.5; scale bar = 10μm).

### File name: Supplementary Movie 2

Description: PolyP-Mg<sup>2+</sup> condensates form vacuoles under non-equilibrium conditions that fuse with time (1mg/mL polyP, 300 mM MgCl<sub>2</sub>, 50mM HEPES, pH 7.5; scale bar = 10μm).

### File name: Supplementary Movie 3

Description: PolyP-Mg<sup>2+</sup>-DNA condensates form spherical polyP-Mg<sup>2+</sup> condensates (polyP-AF647, blue) surrounded by a DNA shell (1μM YOYO-1, yellow) (1mg/mL polyP, 10μg/mL pUC19, 100 mM MgCl<sub>2</sub>, 50mM HEPES, pH 7.5; scale bar = 10μm).

### File name: Supplementary Movie 4

Description: Visualization of polyP-Mg<sup>2+</sup> condensate on cryo-EM grid. Top-down and side views of the 3D rendering shown in Figure 3h. PolyP is shown in dark red, the polyP dense edge is shown in light red, the carbon/polyP interface is indicated by a green bracket, and DNAs are shown in cyan.

### File name: Supplementary Movie 5

Description: Visualization of polyP-Mg<sup>2+</sup>-pUC19 condensate on cryo-EM grid. Top-down and side views of the 3D rendering shown in Figure 3i. PolyP is shown in dark red, the dense edge+DNA is shown in yellow, the carbon/polyP interface is indicated by a green bracket, and DNAs are shown in cyan.

### File name: Supplementary Movie 6

Description: Visualization of polyP-Mg<sup>2+</sup>-pUC19(10X) condensate on cryo-EM grid. Top-down and side views of the 3D rendering shown in Figure 3j. PolyP is shown in dark red, the dense edge+DNA is shown yellow, the carbon/polyP interface is indicated by a green bracket, and DNAs are shown in cyan.

### File name: Supplementary Movie 7

Description: Visualization of polyP-Mg<sup>2+</sup>-15kb condensate on cryo-EM grid. Top-down and side views of the 3D rendering shown in Figure 3k. polyP is shown in dark red, the dense edge+DNA is shown in yellow, the carbon/polyP interface is indicated by a green bracket, and DNAs are shown in cyan.

### File name: Supplementary Movie 8

Description: Addition of 15kb plasmids, results in smaller DNA-shelled condensates (polyP-AF647: blue, 1μM YOYO-1: yellow) (1mg/mL polyP, 10μg/mL pUC19, 100 mM MgCl<sub>2</sub>, 50mM HEPES, pH 7.5; scale bar = 10μm).

### File name: Supplementary Movie 9

Description: When T4 DNA was added, grape-like clusters of condensates emerged. These clusters moved together as a unit, but did not fuse on the timescale observed.