Supplementary Information:

## **S1: Preparation of DNA nanoparticles**

Condensates were prepared by adding DNA to polycation solution in NaCl to a final concentration of 1 ng/µl DNA, 1.36 mM PEIm/PEI and 50 µM NaCl (Fig.S1). The nominal N/P ratio (number of PEI amines available per DNA phosphate.) of the resulting solution was greater than 10. However, only ~90% of the PEI nitrogens and ~70% of the PEIm nitrogens are protonated and charged at pH 7 <sup>1.4</sup>. Consequently the polymer cation to DNA anion ratio in the condensate solution is somewhat smaller. The nanoparticle solution was incubated for at least 12 hours before characterization studies. The condensate solution does not contain buffer and has low salt content. This makes it possible to to image nanoparticles in dry air with Atomic Force Microscopy, without prior washing of the imaging surface. The washing step may affect the nanoparticle size distribution.

## 12.5 kb plasmid



**Fig. S1**. (Above) DNA plasmid adsorbed on the AFM surface. (Below) AFM phase image (in air) of nanoparticles made from 12.5 kb DNA after 12.5 hours incubation with PEIm.





**Fig. S2.** [A] DLS correlation curve of DNA/PEI nanoparticles (hydrodynamic diameter = 56.9nm), [B] maximum height distribution of DNA/PEI and DNA/PEIm nanoparticles.

The figure above shows a typical DLS autocorrelation curve for DNA/PEI nanoparticles, which indicates broad size distribution with a mean hydrodynamic diameter of 56.9 nm (Fig. S2a). The data was collected with a Zetasizer ZS (Malvern Instruments) at 173° scattering angle. The mean area diameter of these particles was about 85nm indicating that a large population of the particles spread during drying. A rough estimate of the water content of 77% can be made by subtracting the dried volume (calculated from the average volume-equivalent diameter of 35nm from AFM imaging) from the swollen particle volume (estimated from the DLS hydrodynamic diameter). The water fraction is lower than the 90% water fraction estimated for DNA-PEIm nanoparticles. The latter was estimated from their diameters estimated from AFM images. Fig. S2b shows the height distributions of the DNA/PEI and DNA/PEIm nanoparticles.

## **<u>S3. AFM imaging of branched PEI nanoparticles adsorbed in fluid environment.</u>**

Branched PEI (Polysciences Inc, 25kDa) nanoparticles were prepared by the same procedure as PEI and PEIm nanoparticles, but these particles did not hold their shape when adsorbed on a surface in solution.



Fig. S3. AFM images of swollen DNA/PEI-branched nanoparticles in fluid.





Fig. S4. Indentation force curve of DNA/PEIm nanoparticles.

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