

## Supplementary data

### Synthesis of TMA<sub>n</sub>AuNP:

Dodecanethiol (C<sub>12</sub>) functionalized gold nanoparticles (AuNP), NNN trimethyl(11-mercaptoundecyl) ammonium chloride ligands, and trimethylammonium (TMA) C<sub>12</sub> functionalized gold nanoparticles (TMA<sub>n</sub>AuNP, n denotes the number of TMA ligands on a nanoparticle) were synthesized using the available procedures.<sup>15-17</sup> Different reaction times (24-hour or 51-hour) were used in the ligand exchange reactions to make TMA<sub>n</sub>AuNP, which yielded different n values. NMR and UV-VIS were used to verify the reaction intermediates and products.

### General description of the electrophoresis experiments:

1.2% and 0.8% Agarose E-gels (Invitrogen) and Agarose gels prepared in the lab were used to detect supercoiled DNA, gold nanoparticles, and their complexes. The running conditions were between 50 or 60 V for 45 min (E-gels) or 3 hours (self-poured gels). Gels were inspected with a transilluminator (ChemiDoc XRS, Bio-Rad).

### Nanoparticle inspections and radiation testing conditions:

Transmission electron microscope (TEM, Philips CM-12) was used to examine the nanoparticle samples. X-ray radiation tests were performed at the UC Davis Cancer Center (RS2000, Radsource, operated at 100 keV). The maximum dosage was determined by radiating free scDNA, normally at 0.5 Gy/min radiation flux. 0 to 16 minutes of radiation assays were used, with the 16-min radiation (8 Gy total dosage) being able to completely convert scDNA into relaxed scDNA.

Lane assignments for the gel electrophoresis experiments studying the mobility of the scDNA-AuNP complexes, as shown in Figure 2:

In these gels (in 0.5× TBE buffer), two parallel columns of wells, 6 mm (w) × 2 mm (thickness) × 3 mm (depth) in dimension and labeled as 1A through 6A and 1B through 6B were prepared. The locations of scDNA were detected by adding ethidium bromide to the gels and viewed on the transilluminator. 1.65 μg 51-hour TMA<sub>n</sub>AuNP in 10 μL Milli-Q (MQ) water was added into wells 2A, 3A, 5A, and 6A. 1 μg scDNA in 8 μL MQ water was added into wells 1A, 4B, 5B, and 6B. A 1:1000 ratio mixture of scDNA (1 μg scDNA in 8 μL MQ water) to TMA<sub>n</sub>AuNP (16.5 μg 51-hour AuNP in 10 μL MQ water) was added into well 2A. Wells 1B, 2B, 3B, and 4A were left empty.

Lane assignments for the radiation testing described in Figure 3:

Three pairs of 6-set samples were prepared. 200-ng scDNA was used for preparing each of the 36 samples. 1-μg 24-hour TMA<sub>n</sub>AuNP was then added to each of the scDNA solutions to prepare the 18 scDNA-TMA<sub>n</sub>AuNP samples. Each pair of samples were identically prepared and radiated. For each pair of the 6-set free scDNA and scDNA-TMA<sub>n</sub>AuNP samples, they were injected into PCR tubes with the tabs removed and covered with 3-μm Mylar films, and were radiated for 0, 1, 2, 4, 8 and 16 minutes respectively. The experimental conditions such as preparation times, exact radiation times, and waiting times before and after the radiation varied slightly for these three pairs of samples. The samples were then loaded into the wells of the E-gels for quantitative analysis.