S.1. Preparation of materials

Synthesis and characterisation of citrate-stabilised gold nanoparticles (AuNP)

Citrate-stabilised gold nanoparticles were prepared by modification of the method outlined by Slot and Geuze.\textsuperscript{18} An aqueous solution of tetrachloroauric acid trihydrate (1.00 mL of a 1 % wt. aqueous stock solution, 0.026 mmol) in deionised water (79.00 mL) was heated to 60 °C with stirring. To this was added a separate solution heated to 60 °C containing trisodium citrate dihydrate (4.00 mL of a 1 % wt. aqueous stock solution, 0.136 mmol) and tannic acid (0.01 mL of a 1 % wt. aqueous stock solution, 29.4 μmol) in deionised water (3.99 mL) and the combined solution heated at 60 °C until a ruby red colour was observed. This was rapidly brought to the boil and finally cooled to yield a ruby red solution of citrate-stabilised gold nanoparticles.

UV-vis (H2O): \(\lambda/\text{nm} 521.32 \ (6.09\times10^8)\).

DLS (H2O): \(\text{APD}/\text{nm} 18.6 \ (8.7 – 30.6 \text{ nm}, \text{PdI} = 0.156, \text{N} = 3)\).

TEM (100 kV, “holey” carbon film): \(\text{APD}/\text{nm} 16.3 \pm 1.5 \ (10.9 – 20.2 \text{ nm}, \text{N} = 100)\).

Modification and characterisation of multi-walled carbon nanotubes (MWNT)

Multi-walled carbon nanotubes (60.4 mg, Nanocyl #3100) were ground using a pestle and mortar for 15 min to yield a black solid (60.4 mg, 100 % wt.).

IR (KBr): \(\nu/\text{cm}^{-1} 1637\text{s}, 1383\text{m}\).

UV-vis (H2O): \(\lambda/\text{nm} 254.65 \ (54.38 \text{ mL mg}^{-1} \text{ cm}^{-1})\).

TEM (100 kV, “holey” carbon film): \(\text{ATL}/\mu\text{m} 2.04 \pm 0.32 \ (0.6 – 4.6 \mu\text{m}, \text{N} = 20)\); \(\text{ATD}/\text{nm} 9.2 \pm 2.2 \ (5.5 – 13.9 \text{ nm}, \text{N} = 50)\).

Assembly of gold nanoparticle – carbon nanotube composite materials

For each experiment, a known mass of nanotubes (0.01 mg for variation of pH; 0.05 mg for variation of ionic strength) was added to a known volume of gold nanoparticles (3 mL, concentration = 4.90x10^{12} NP mL\textsuperscript{-1} for variation of pH, 5.37x10^{12} NP mL\textsuperscript{-1} for variation of ionic strength). The heterogeneous mixture was sonicated for 30 mins at room temperature, the suspension filtered through a 0.2 μm cellulose acetate syringe filter and the supernatant characterised by UV-vis spectroscopy.
S.2. Characterisation of surface charge by $\zeta$-potential analyses

*Figure S.1.* $\zeta$-potential analysis of AuNP as a function of pH (pH = 2.9, 3.8, 5.1, 6.1, 7.1 and 7.6, right to left).

*Figure S.2.* $\zeta$-potential analysis of MWNT as a function of pH (pH = 2.9, 3.8, 5.1, 6.1, 7.1 and 7.6, right to left).
Figure S.3. ζ-potential analysis of AuNP as a function of ionic strength ($I = 0.0006, 0.003, 0.006, 0.015, 0.03$ and $0.06$ mol dm$^{-3}$, left to right).

Figure S.4. ζ-potential analysis of MWNT as a function of ionic strength ($I = 0.0006, 0.003, 0.006, 0.015, 0.03$ and $0.06$ mol dm$^{-3}$, left to right).
S.3. Calculation of the magnitude of electrostatic forces

Conversion of the recorded ζ-potential into the magnitude of electrostatic forces was determined using a combination of the theories established by Fitzmaurice et al.\textsuperscript{28,29} and Ohshima et al.\textsuperscript{30,31} ζ-potential (in mV) was initially converted into surface charge density (in C m\textsuperscript{-2}) using the equations for distribution of charge around a sphere (AuNP) and a cylinder (MWNT)\textsuperscript{30} of known dimensions respectively. Surface charge density was next converted into total charge per active surface area (in C)\textsuperscript{30,31} – the active surface area of the components were determined by approximating ellipses overlayed on the surface of a sphere (AuNP) and a cylinder (MWNT) respectively. Relative magnitude of electrostatic force (in N) was calculated using Coulomb’s inverse square law – the distance between charges was assumed to be 3 nm as this is the distance where all short-range, attractive interactions tend to zero\textsuperscript{29} and as such this is the position of maximum electrostatic repulsion.