Carbon-gold core-shell structures: Formation of shells consisting of gold nanoparticles

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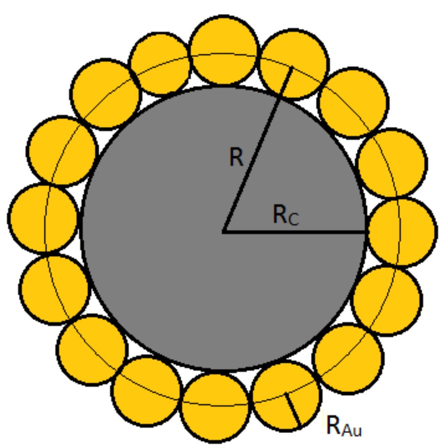
Estimation of Au wt% in the carbon-gold core-shell particles

Calculations are performed for a single carbon sphere having radius of 250 nm and a gold nanoshell having thickness of 50 nm; this shell is assumed to be a monolayer of densely-packed gold nanospheres having radius of 25 nm (**Model I**) or a continuous gold layer having thickness of 50 nm (**Model II**).

Model I: Densely packed monolayer of gold nanoparticles

Radii of a carbon particle (R_C) and gold nanoparticles (R_{Au}) were assumed to be 250 nm and 25 nm accordingly, as determined from the SEM images. The number of gold nanoparticles (N) on the carbon sphere surface (see Scheme S1) is evaluated by dividing the surface (S) of a sphere determined by the centers of gold nanoparticles in the monolayer by the effective cross-section area of a single gold particle (S^e_{Au}). This effective cross-section area is calculated by adjusting the principal cross-section area (S_{Au}) for the unoccupied area between the closed-packed gold spheres (S_{in}). Although this method does not take into account the curvature of the cross-section area, this approximation works well because: (a) the aforementioned curvature is negligible for the particles under consideration ($R_{Au}/R \approx 0.09 rad \approx 5.2°$) and (b) the error made by underestimation of the cross-section area is self-compensated to some extent because of its presence in both the nominator and denominator of the final equation.



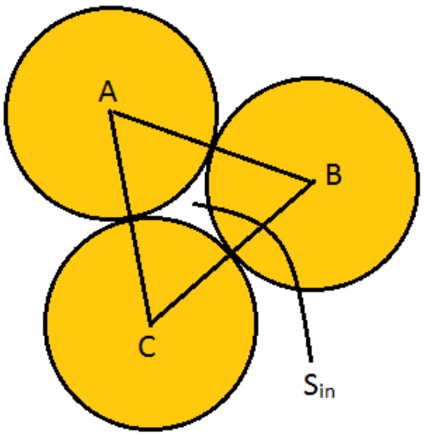
Scheme S1. A cross-section of a carbon particle with gold nanoparticles on the surface.

R_C – radius of a carbon sphere
V_C – volume of a carbon sphere
d_C – density of a carbon sphere
m_C – mass of a carbon sphere
R – radius of a sphere determined by the centers of gold nanoparticles deposited on a carbon sphere
S – surface of a sphere with the radius R
R_{Au} – radius of a gold particle
S_{Au} – cross-section area of a gold particle
S^e_{Au} – effective cross-section area of a gold particle
V_{Au} – volume of a gold particle
d_{Au} – density of a gold particle
m_{Au} – mass of a gold particle

First, the mass m_C of a carbon sphere with a radius $R_C = 250nm$ is calculated using the density $d_C = 1.5\frac{g}{cm^3}$.

$$V_C = \frac{4}{3}\pi R_C{}^3 = \frac{4}{3}\pi(250nm)^3 = 6.545 * 10^7 nm^3$$
$$m_C = V_C * d_C = 6.545 * 10^7 nm^3 * 1.5\frac{g}{cm^3} = 98.17 * 10^{-15} g$$

Then, the mass m_{Au}, the cross-section area S_{Au}, and the effective cross-section area S_{Au}^e of a single gold particle are calculated assuming: $R_{Au} = 25nm$, the density of gold $d_{Au} = 19.3\frac{g}{cm^3}$, and the closed-packed structure of gold particles in the monolayer formed on the carbon sphere surface (see Scheme S2).



Scheme S2. A from-top view of the gold-spheres monolayer.

$$S_{Au} = \pi R_{Au}{}^2 = \pi(25nm)^2 = 1.963 * 10^3 nm^2$$
$$V_{Au} = \frac{4}{3}\pi R_{Au}{}^3 = \frac{4}{3}\pi(25nm)^3 = 6.545 * 10^4 nm^3$$
$$m_{Au} = d_{Au} * V_{Au} = 19.3\frac{g}{cm^3} *$$
$$* 6.545 * 10^4 nm^3 = 1.263 * 10^{-15} g$$

$$S_{ABC} = 3 * \frac{1}{6} S_{Au} + S_{in} = \frac{1}{2} S_{Au} + S_{in}$$
$$S_{Au}^e = S_{Au} + 2S_{in} = 2S_{ABC} = 2 * \left(\frac{\sqrt{3}}{4}a^2\right) =$$
$$= 2 * \left(\frac{\sqrt{3}}{4}\left(2R_{gold}\right)^2\right) = 2 * \left(\frac{\sqrt{3}}{4}(2 * 25nm)^2\right) =$$
$$= 2.165 * 10^3 nm^2$$

The surface S of a sphere determined by centers of the gold particles in the monolayer is:

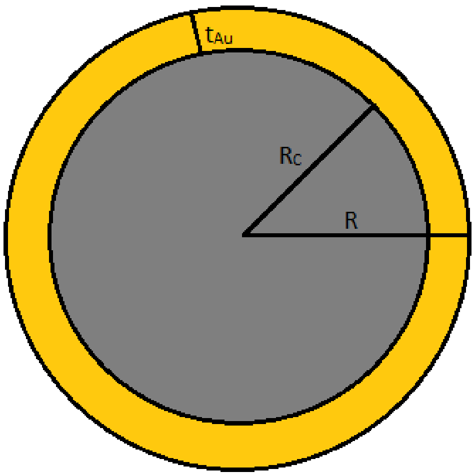
$$S = 4\pi R^2 = 4\pi(275nm)^2 = 0.9503 * 10^6 nm^2$$
where $R = 250nm + 25nm = 275nm$

Finally, the number N of gold particles on the carbon sphere surface is estimated by dividing the S surface area by the effective cross-section surface area S_{Au}^e. The wt% Au in the carbon-gold core-shell particle is calculated by dividing the mass of N gold nanoparticles by the total mass of the carbon-gold core-shell particle

$$N = \frac{S}{S_{Au}^e} = \frac{0.9503*10^6 nm^2}{2.165*10^3 nm^2} = 439$$
$$R\% = \frac{N*m_{Au}}{m_C+N*m_{Au}} = \frac{439*1.263*10^{-15}g}{98.17*10^{-15}g+439*1.263*10^{-15}g} = 85.0\%$$

Model II: Continuous layer of gold

This model assumes a continuous gold layer formed on the surface of a carbon sphere. A radius of the carbon particle (R_C) and the thickness of the gold layer (t_{Au}) are assumed to be 250 nm and 50 nm accordingly, as determined from the SEM images. The theoretical amount of gold is calculated by dividing the mass of a single gold layer by the total mass of a carbon-gold core-shell particle.



Scheme S3. A cross-section of a carbon particle with a continuous gold layer on the surface.

R_C – radius of a carbon sphere
V_C – volume of a carbon sphere
d_C – density of a carbon sphere
m_C – mass of a carbon sphere
R – radius of a carbon-gold core-shell particle
V – volume of a carbon-gold core-shell particle
t_{Au} – thickness of a gold layer
V_{Au} – volume of a gold layer
d_{Au} – density of a gold layer
m_{Au} – mass of a gold layer

First, the mass m_C of a carbon sphere with the radius $R_C = 250nm$ is calculated using the density $d_C = 1.5\frac{g}{cm^3}$.

$$V_C = \frac{4}{3}\pi {R_C}^3 = \frac{4}{3}\pi(250nm)^3 = 6.545 * 10^7 nm^3$$
$$m_C = V_C * d_C = 6.545 * 10^7 nm^3 * 1.5\frac{g}{cm^3} = 98.17 * 10^{-15} g$$

Then, the mass m_{Au} of a continuous gold layer is calculated assuming: $t_{Au} = 50nm$ and the density of gold $d_{Au} = 19.3\frac{g}{cm^3}$. (see Scheme S3).

$$R = 250nm + 50nm = 300nm$$
$$V_{Au} = V - V_C = \frac{4}{3}\pi R^3 - \frac{4}{3}\pi {R_C}^3 = \frac{4}{3}\pi\left(R^3 - {R_C}^3\right) = \frac{4}{3}\pi((300nm)^3 - (250nm)^3) =$$
$$= 4.765 * 10^7 nm^3$$
$$m_{Au} = V_{Au} * d_{Au} = 4.765 * 10^7 nm^3 * 19.3\frac{g}{cm^3} = 9.196 * 10^{-13} g$$

Finally, the wt% Au is calculated by dividing the mass of a continuous gold layer by the mass of the carbon-gold core-shell particle.

$$R\% = \frac{m_{Au}}{m_C + m_{Au}} = \frac{9.196*10^{-13}g}{98.17*10^{-15}g + 9.196*10^{-13}g} = 90.4\%$$

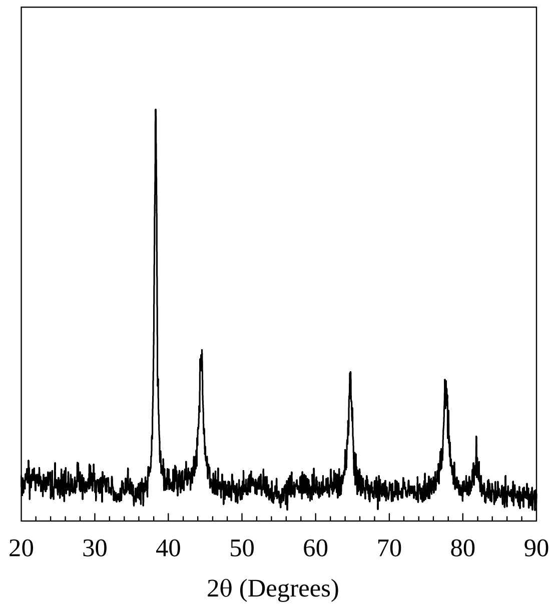


Figure S1. A wide-angle XRD pattern of the carbon-gold core-shell particles recorded on a powder X-ray diffraction HZG-4 instrument (VEB Freiburg Prazisionmechanik, Germany) using Cu Kα radiation. The five reflection peaks (from left) (111), (200), (220), (311) and (222) are identified according to the JCPDS card number 03-065-2870 for gold crystals possessing a cubic structure with *Fm3m* symmetry.

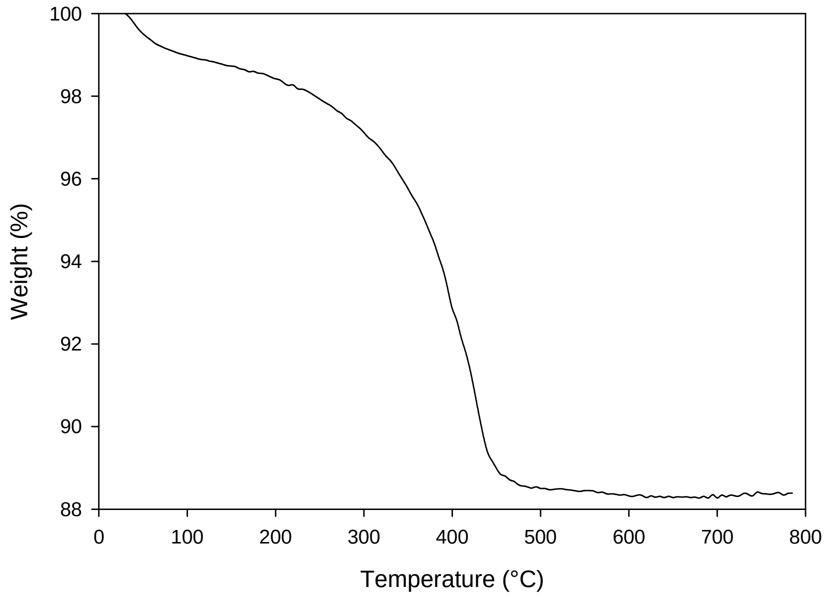


Figure S2. A thermogravimetric (TG) profile of the carbon-gold core-shell particles recorded using TA Instruments TGA Q500 (New Castle, DE, USA) with 10°C/min heating rate in flowing air in 20 – 800°C range. The amount of deposited Au nanoparticles was estimated as the residue at 700^{o}C in relation to the sample mass at 150^{o}C; this residue is 89.4%.