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Radiolytical Synthesis and Mechanism of Gold Nanoparticle Formation

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Gold nanoparticles (AuNPs) are widely used in analytical chemistry, biomedicine and catalysis. Reducing agent is very important parameter in the synthesis of AuNPs, however, hypothesis of this study is that AuNPs could be synthesized without using reducing agents, i.e. that due to very low affinity of gold for oxygen AuNPs could be synthesized under highly oxidizing conditions.

AuNPs were synthesized in a microemulsion water/TritonX-100/1-pentanol/cyclohexane using various reducing agents: (i) strong chemical reducing agent (NaBH₄), (ii) γ -irradiation under moderately strong reducing/oxidizing conditions, and (iii) synthesis under highly oxidizing conditions (with addition of NaOH aqueous solution). All were performed at room temperature (RT). The size, size distribution, aggregation and stability of AuNPs in the microemulsions depend on the strength of reducing agent. When a strong chemical reducing agent NaBH₄ was used AuNPs are not well-stabilized and rather polydisperse. Rather small and monodisperse AuNPs were obtained using γ -irradiation (30 kGy; 8 kGy/h). γ -irradiation was able to produce AuNPs in air-saturated microemulsions under highly oxidizing conditions. Smaller AuNPs were obtained by irradiation in the presence of N₂ in comparison to the air. The γ -irradiation of nitrogen-saturated microemulsion at acidic pH produced AuNPs about 10 nm, which aggregated under isolation by centrifugation. The microemulsion stirred at RT and at pH<7 under oxidizing conditions did not produce AuNPs, while at pH>7 (stronger oxidizing conditions) well-dispersed 12 nm AuNPs were formed. Synthesis of AuNPs in 1-pentanol by adding NaOH aqueous solution at RT without using microemulsions and irradiation confirmed that oxidation of alcohols was responsible for AuNPs formation. Based on these findings we propose the base-catalyzed alcohol oxidation at RT as a new, simple and versatile synthesis route for obtaining gold nanoparticles.

In order to further exploit the oxidation of organic molecules for AuNPs synthesis we studied the radiolytical synthesis of AuNPs in the presence of citrate. The γ -irradiation of Au(III)/citrate precursor solutions produced well-dispersed and highly concentrated gold colloids in the presence of dissolved oxygen, without adding any reducing or stabilizing agents. AuNP size can be controlled by saturating gases (air or nitrogen) present in the precursor solution. AuNPs synthesized in the presence of air (10 nm) were approximately two times larger than AuNPs synthesized in the presence of nitrogen (5 nm), as determined by UV-Vis spectroscopy. An easy radiolytical reduction of Au(III)/citrate precursor solution in the presence of dissolved oxygen could be explained by enhanced radiolytical oxidation/decarboxylation of citrate to dicarboxyacetone, acetone and other products. Thus, we confirmed that classical approach of using a reducing agent to synthesize AuNPs is not a determining factor, since diametrically different approach can be used, namely in stimulating the oxidation of organic molecules close to gold ions. However, the mechanism of oxidation of organic molecules during the synthesis is not clear and due to these reasons we introduced NMR spectroscopy in order to study the oxidation products of alcohol and citrate ions during the AuNPs synthesis.

Country/Organization invited to participate

Croatia

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