476h Enhanced Infusion of Gold Nanocrystals into Mesoporous Silica with Supercritical Carbon Dioxide

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Gold nanocrystal dispersions in toluene-CO2 mixtures were infused into cylindrical pores in mesoporous silica to achieve high loadings over 2 wt% in 24 hours. The nanocrystals were highly dispersed according to transmission electron microscopy and the loadings approached equilibrium. In contrast, the loadings were small for infusion with pure toluene or toluene mixed with an antisolvent, methanol. The differences in loading were correlated with the long-ranged van der Waals forces between the gold and silica through the intervening solvent. These van der Waals forces became stronger as CO2 was added to toluene, as a consequence of a reduction in the Hamaker constant of the mixed intervening solvent, resulting in stronger nanocrystal adsorption. The decoupling of the nanocrystal synthesis step and the infusion step, leads to exquisite control of the nanocrystal size, morphology and dispersibility within the pores. The simplicity of the method allows for the facile production of nanocrystal/silica composites for applications such as catalysis and optoelectronics.