In-situ XAS Study on the Initial Nucleation and Growth Processes of CdSe Nanocrystals

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The initial nucleation and growth processes of CdSe nanocrystals are studied by in-situ extended X-ray absorption fine structure (EXAFS) combined with UV-vis absorbance and photoluminescence spectroscopy. We measured the Se K-edge EXAFS spectra for CdSe nanocrystals (NCs) along a microfluidic cell (reactor) which indicates time-dependent growth kinetics of bond formation that are consistent with that of UV-vis spectroscopy. It was found that initial nucleation burst is associated with a rapid particle size increase (NC growth stage). We show that Cd-Se bond formation kinetics obtained from the Se-Cd bond creation and Se-P bond breaking based on the EXAFS analysis are consistent although XANES results are sensitive to validity of reference. The role of dodecylamine (DDA) as a surfactant in the initial nucleation and growth was also studied. We found that the initial nucleation stage is significantly accelerated by the presence of DDA, i.e., DDA increases the average particle size while the growth rate of the NCs is suppressed. The results may indicate two competitive roles of DDA in the current system, namely, activating reagent of the precursors in the initial nucleation stage and passivating ligand of the NCs during the subsequent growth process that suppresses growth into larger size. The results demonstrate that by optimizing the DDA concentration, both particle size and density can be tailored.

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