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In-situ EXAFS study of initial formation processes of CdSe nanocrystals

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Introduction

Understanding the details of the nucleation and growth processes is essential for growing nanocrystals with desired size and optoelectronic properties. In an initial stage of a reaction to produce CdSe nanocrystals, typically within tens of seconds, the dominant process is nucleation which is also accompanied by a slow growth process. To date it is still rather challenging to probe the quick nucleation process.

In the present study, *in-situ* extended x-ray absorption fine structure (EXAFS) technique is employed for studying the CdSe nanocrystals nucleation process by using a microreactor. The basic idea is to let the reaction solution flow along a microreactor channel at a constant flow rate and to measure the EXAFS spectra at different positions after the start of reaction.

Experimental

Three stock solutions, Se source, Cd source and amine solution as surfactant were prepared. These three stock solutions were mixed in the microfluidic mixer connected to the heated 70-mm-long reactor channel (Kapton tube). The heating unit was kept at a temperature of 240 °C. *In-situ* EXAFS spectra at Se *K*-edge were performed at beamline BL13B1 of Photon Factory. The microreactor was mounted on a high precision XY stage on a goniometer (Huber 410) controlled by stepping motors. Fluorescence x-ray was detected by a compact silicon drift diode detector. The EXAFS data at four points labeled as *A*, *B*, *C*, and *D* along the channel were collected. A schematic representation of the *in-situ* microreactor EXAFS system is shown in Fig. 1.

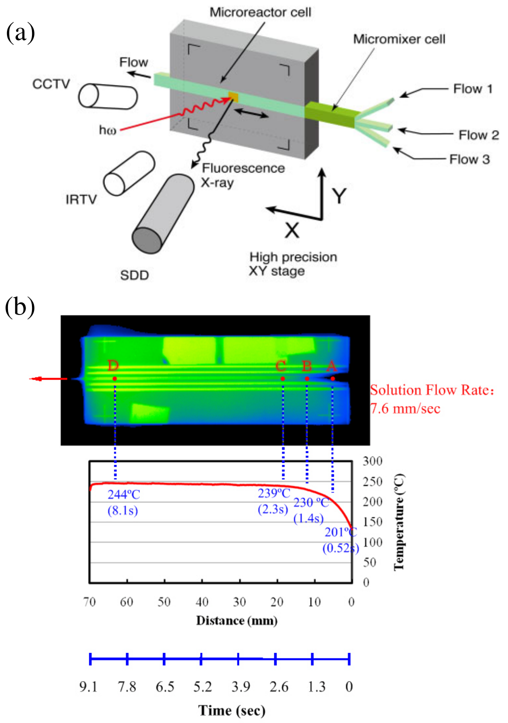

Fig. 1 (a) Schematic presentation of the experimental setup. (b) The positions and temperatures of four points along the microreactor channel as measured.

Results and Discussions

Figure 2 shows the Fourier transforms (FT) of the $k^2\chi(k)$ functions at points *A* through *D*. A prominent peak (R=1.6 Å) for the starting TOP-Se is ascribed to the Se-P bond. For the CdSe powder data, the strong peak at 2.5 Å is attributed to the nearest Se-Cd bond. At point *A* the FT exhibits no Se-Cd signal, indicating that the reaction barely started at 0.52 s. At point *B*, a weak Se-Cd peak appears at 2.5 Å. This Se-Cd peak further increases in intensity at point *C*, and reaches a maximum at point *D*, accompanied by an obvious reduction of the Se-P peak intensity. Because of the limited heating time (<8.1 s) of reactants in the microreactor, the Se-Cd bond can mostly originate from the initial CdSe nuclei formed from the TOP-Se solution. The strong position-dependence indicates a rapid increase of the reaction yield to CdSe nuclei along the microreactor channel.[1]

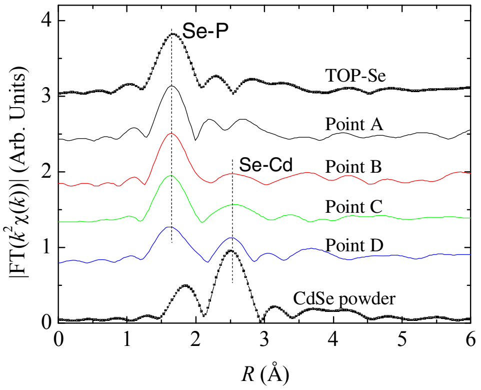

Fig. 2 The Fourier transform of the Se *K*-edge $k^2\chi(k)$ EXAFS functions at four points *A*, *B*, *C*, and *D*.

References

[1] M. Uehara et al., Appl. Phys. Lett. 94, 063104 (2009).

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