Effects of Aging on the Broadband Luminescence of Semiconductor Nanocrystals

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Luminescence from semiconductor nanocrystals depends on both the cluster surface chemistry and the surrounding environment or matrix. Aging at mild temperatures in the presence of suitable surfactants adsorbed onto the surface of 2 nm CdS nanocrystals is shown to result in a range of photoluminescent (PL) peak energies and widths. These changes in surface structure can be used to create broadband, white light emission from such clusters. The development of white light PL occurs through control of the nanocluster surface and as part of a surface reconstruction process over a period of time, i.e., aging. The onset of light absorption does not change with age. This aging process which results in broadband PL requires months at ambient temperatures. In this work we demonstrate the acceleration of the aging process to create white light over shorter periods of time. There are three approaches to accelerating the aging process. In the first approach, the CdS nanocluster solutions were heated to low temperatures, ~40-65^{o}C, for varying periods of time. This method allows ions on the CdS surface to rearrange to their thermodynamically favored state in a shorter period of time. Figure 1 shows the effects of low temperature controlled heating of the CdS nanoclusters over different time ranges. At elevated temperatures such as 65^{o}C, we observed a significant broadening of the spectra (~34nm). The second approach involves altering and controlling the surface chemistry of the CdS nanoclusters. Different chain length thiols were added to the CdS nanocrystals surface in controlled amounts and under controlled environmental conditions. The ligand binding strength of the thiol to the nanocluster surface is affected by the thiol chain length. This effect changes the rearrangement rate of the surface species. The third approach combines altering the surface chemistry with temperature treatment. Figure 2 shows CdS nanoclusters with hexadecane thiol added to the surface and exposed to elevated temperatures. Not only did the spectra broaden, but luminescence intensity increased. In each approach High Pressure Liquid Chromatography (HPLC), Dynamic Light Scattering (DLS) and Transmission Electron Microscopy (TEM) were used to verify that the nanocrystal size did not change. Another important factor is the control of the Cd:S stoichiometry. This is critical to the PL efficiency. X-ray Fluorescence (XRF) was used to verify the stoichiometry of the clusters. The effect of spectral broadening appears to be due solely to change in the surface structure of the nanocluster.

Acknowledgments:

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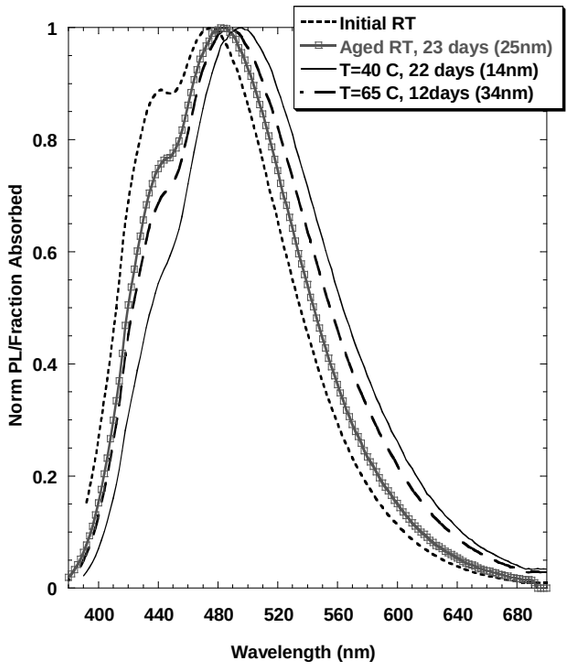

Figure 1. Normalized PL/fraction of light absorbed showing effects of age and temperature on PL spectral broadening.

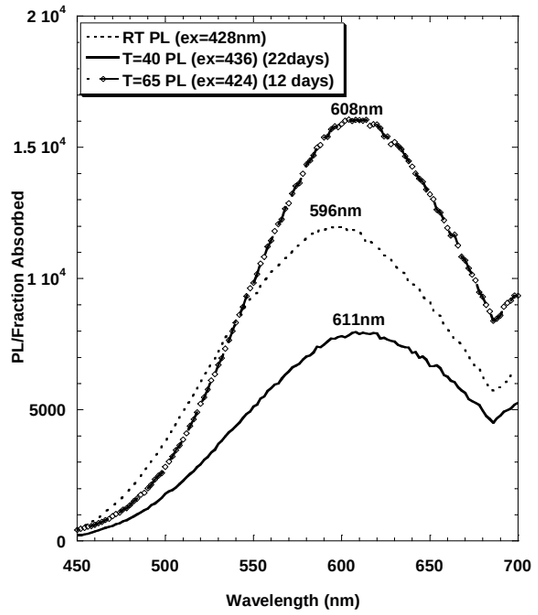

Figure 2. PL/fraction of light absorbed for Thiol treated CdS nanoclusters. Note the red shift and broadening compared to figure 1.