Surface Controlled Luminescent CdS Nanocrystals

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Introduction

Multi-colored emissions from semiconductor nanocrystals can be applied for biological labeling, quantum electronics and luminescent devices. The color tunability is often dependent upon the particle size relating to quantum size effect. Accordingly, size controlled synthetic methods by altering starting materials and experimental conditions have intensively been studied. We have recently developed a facile method to prepare nanocrystals highly emissive CdS under mild experimental conditions¹. The enhanced emission must originate from surface modification of the nanocrystals as the surface states are thought to trap photo-generated charges, leading dominantly to non-radiative charge recombination. However, the details of the emission mechanism were not fully understood. We thus present our recent investigations on parameters controlling the emission intensity and the surface structures.

Experimental

Highly luminescent CdS nanocrystals were prepared as we reported previously¹. Briefly, 0.2 mM Cd(ClO₄)₂ was reacted with equimolar H₂S gas or Na₂S solution in the presence of 0.1 mM (NaPO₃)₆ as a stabilizer, resulting in clear yellow colored CdS nanocrystals. The resultant solution emits weakly, originating probably from the recombination at the surface states. Surface modification of the crystals was carried out by adding an appropriate alkaline solution adjusting the solution pH.

Results and discussion

Our recent paper demonstrated that addition of NaOH into a CdS aqueous solution enhanced the emission intensity, however it was dependent upon the solution pH. At pH = 11, the highest intensity was achieved. As this enhancement was carefully investigated, we found it actually increases day by day when it is just left in the room. Figure 1 shows an example of the emission spectral change. After 21 days, the emission reaches maximum and is maintained steadily for several tens of days. Further investigation lead to the point that this increase was in fact accelerated by light irradiation as shown in Figure 2. In this way, the emission gains the maximum at most 5 days after the NaOH treatment.

Our preparation method of the luminescent CdS can simply be applied to obtain multi-colored nanocrystals by controlling a particle size. The size control can readily be achieved by using the size-selective photo-etching technique^{2,3}, which is capable of producing a desired particle size. A series of emission spectra obtained with etching wavelengths of 390, 430, 457, 488 and 500 nm are shown in Figure 3. Narrow emission bands confirm the mono-dispersed particle size.

In the presentation, details of experimental procedures and mechanism of the emission intensity growth will be discussed.

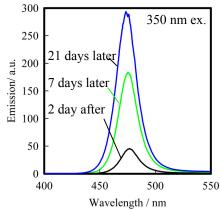


Figure 1. Emission spectra obtained after NaOH addition.

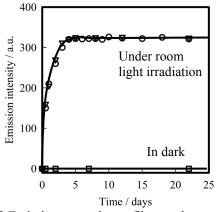


Figure 2.Emission growth profiles under room light irradiation (\bigcirc : first batch, \bigtriangledown : second batch) and in dark (\Box).

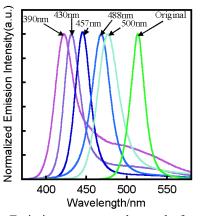


Figure 3. Emission spectra observed for the size controlled CdS nanocrystals achieved by the photoetching. The etching light wavelengths are shown above.

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