

Preparation of Uncapped $\text{CdSe}_x\text{Te}_{1-x}$ Nanocrystals with Strong Near-IR Tunable Absorption

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Semiconducting nanocrystals with near-infrared (NIR) photosensitivity are appealing materials for application as photodetectors and in medical diagnostics. Herein, we report the preparation of composition-tunable, uncapped $\text{CdSe}_x\text{Te}_{1-x}$ ($x = 0$ to 1) nanocrystals by simple mechanical alloying. The resulting ternary $\text{CdSe}_x\text{Te}_{1-x}$ ($x = 0.25, 0.5, 0.75$) nanocrystals with average sizes smaller than 10 nm have zincblende crystal structure, instead of the wurtzite structure commonly obtained by wet chemical routes, and show strong NIR absorption even beyond 1400 nm. While a linear relationship between the lattice parameter and the chemical composition (Se/Te ratio) is observed, indicating the formation of homogeneous alloys, the bandgap energy of the three ternary samples is found to be substantially lower than that of binary CdSe or CdTe nanocrystals, and lower than any ternary CdSeTe reported so far. Existence of a small number of tellurium metal defects in the $\text{CdSe}_x\text{Te}_{1-x}$ ($x = 0.25, 0.5, 0.75$) nanocrystals is confirmed by x-ray diffraction and Raman spectroscopy. Both the optical bowing effect and tellurium metal-induced defects of the mechanically alloyed samples are believed to cause the strong NIR photosensitivity.

Key words: Semiconducting nanocrystals, mechanical alloy, near-IR absorption, cadmium selenide telluride

INTRODUCTION

Semiconducting nanocrystals (NCs) or quantum dots (QDs) exhibit attractive size- and composition-dependent optical properties which make them promising materials for application in bioimaging,^{1–3} solar energy conversion,^{4–7} photocatalysis,^{8,9} and light-emitting diodes.^{10–12} Although a large number of semiconducting NCs have been reported with absorption and emission wavelength ranges covering the ultraviolet (UV) to visible spectrum,^{13–17} less attention has been paid to the near-infrared (NIR) range.^{18–22} The NIR photosensitivity of QDs is an appealing and desired feature for

applications in biological imaging/labeling.^{21,22} It is known that light within the biological NIR window (650 nm to 900 nm), when compared with UV–visible (UV–Vis) light, can penetrate deeper through biological tissues due to less absorption and scattering by tissue constituents.^{23–25} NCs with NIR absorption/emission can thus potentially be used as bio-detectors or fluorescent probes in biological and medical examinations and diagnostics. While there have been a few attempts to fabricate various pure and alloyed NIR-absorbing/emitting NCs such as PbSe QDs,^{26,27} HgS QDs,²⁸ M_xWO_3 ($\text{M} = \text{Na}, \text{Cs}, \text{Tl}, \text{Rb}$) nanoparticles,²⁹ and $\text{InAs}_x\text{P}_{1-x}/\text{InP}/\text{ZnSe}$ alloyed core–shell QDs,³ most attention has focused on cadmium telluride (CdTe) and its alloyed compounds. CdTe has a bandgap of only 1.44 eV, which is considerably lower than that of CdS (2.42 eV) or

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