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DNA template-driven synthesis of HgTe nanoparticles

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Abstract

Nanoparticle (mercury telluride (HgTe))–single-stranded (ss) DNA complex has been synthesized using an electrodeposition technique. As demonstrated by transmission electron microscopic measurements, the morphology of these particles is found to depend upon the ssDNA sequence. The surface feature of HgTe–ssDNA deposit gives an appearance of nanostars with an average size as 1.4 nm and exhibits the single crystalline cubic phase, contrary to the polydispersity and polycrystallinity observed with HgTe nanoparticles alone. The change in infrared absorption band of ssDNA as a consequence of its association with HgTe nanoparticles have been confirmed from Fourier transform infrared spectroscopic measurements. Photoluminescence (PL) at 300 K of these nanostars have yielded a unique single PL of line width 8 nm at wavelength 548.4 nm

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1. Introduction

Semiconductor nanoparticles have been of immense research interest due to their potential application in nanoscale science and technology derived from their size quantization [1–3] and surface effects [4]. Synthesis of nanoparticles by conventional technique has yielded polydispersity which is the major limitation to their use in device applications [5–8]. However, a bottom–up approach can be employed to grow nanostructure semiconductors with a good control over the size. Preparation of nanoparticles by sequence selective DNA is expected to be a good strategy to achieve monodispersity and ordered nanoparticles. Because of the presence of phosphate group, the DNA backbone is negatively charged and behaves as a polyelectrolyte. An interesting property associated with DNA molecule is the electrostatic interaction with positively charged ions [9,10] in the aqueous medium which make them an ideal template to grow nanowires [11,12] and nanoparticles. Thus, such nanoparticles tagging to DNA can be employed for biosensor applications [13–19]. The present report proposes a new and simple strategy for synthesis of HgTe nanocrystals (NCs) using single-stranded DNA (ssDNA) as a template by electrodeposition technique. The unique structural and optical properties, arising from nearly monodispersed and single crystalline HgTe–ssDNA complex (nanostars) are being demonstrated by transmission electron microscopy (TEM) and photoluminescence (PL) measurements, respectively.

2. Experimental

To synthesize the DNA template-mediated HgTe nanoparticles, ssDNA 5′GCAAGCGGTGAACCAGTTGTG3′ was chosen as a typical bio-precursor and the sequence specificity was tested by choosing another ssDNA, poly G(30). The HgTe NCs were synthesized from Hg^{2+}(0.014 M) and Te^{4+}(0.026 M) ions which were made available from their respective salt solutions [18]. The electrolytic bath consisting of ssDNA, Hg^{2+} and Te^{4+} ions was maintained at pH = 0.6 and temperature = 278 K. The

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electrodes were indium tin oxide (ITO) coated conducting glass cathode and platinum anode. Electrodeposition under stirring condition was carried out at 1.5 mA/cm^2 cathodic current density for 2 min. The structure, surface and optical properties of HgTe and HgTe–ssDNA NC systems were evaluated by TEM and PL measurements.

3. Results and discussion

Figs. 1(a) and (b) show the TEM micrograph and selected area electron diffraction (SAD) pattern of HgTe taken at 200 keV. The average size of the HgTe NCs estimated from TEM is 5.35 nm. The inset of Fig. 1(a) is the lattice image of HgTe NCs, which gave the lattice spacing, $d = 0.62$ nm, corresponding to the cubic phase structure of HgTe. The SAD pattern clearly depicts the polycrystallinity with preferred orientation in the sample. On the other hand, the TEM micrograph and the SAD (recorded at 80 keV) of HgTe–ssDNA complex NCs are shown in Figs. 1(c) and (d). The micrograph (Fig. 1(c)) reveals the rounded structures with some protrusion which gives an impression of small stars "nanostars" (as shown by arrows) with an average lateral dimension as 1.4 nm. The SAD of these nanostars shown in Fig. 1(d) depicts the single crystalline cubic phase corresponding to HgTe in contrast to polycrystallinity as observed in HgTe (Fig. 1(b)). Although it is very difficult to distinguish the ssDNA from nanostars because of the large contrast in HgTe compared to ssDNA, the presence of the same with HgTe has been confirmed from Fourier transformed infrared (FTIR) spectroscopic measurement, which is to be discussed later. The overall feature of the micrograph (Fig. 1(c)) reveals as if the ssDNA is wrapped the HgTe NCs. The very fact that nanostars are deposited on the cathode under a cathodic current suggests that the ($Hg^{2+} + Te^{4+}$)–ssDNA complex in the solution is positively charged. Without application of a cathodic current, no deposit was found on the cathode. Then the question arises as to why ssDNA should wrap the counter ions and deposit on the electrode. In order to explain this, we propose the following model.

As per the literature [20], within a cell (aqueous medium), DNA (PO_4^{2-}) interacts with the protein (positively charged) and is self-organized after charge neutralization followed by counter ion condensation [19]. In an analogy of the above, in the present case, ssDNA interacts with the Hg^{2+} and Te^{4+} ions and undergoes a charge condensation process which leads to over-neutralization effect due to the correlation in mobile ion positions arising from high ionic concentration and valence in the presence of a strong surface field [21]. As a result, the ($Hg^{2+} + Te^{4+}$)–ssDNA complex exibits net positive charge due to the charge reversal process [22]. The ssDNA has 21 nitrogenous bases comprising A, T, G and C and each has different oxidation potentials [23]. Hence, the overall feature across the ssDNA length results in a fluctuating oxidation potential. Thus, in order to maximize the entropy or minimize the free energy of the system, the ssDNA has to wrap/bend around the counter ions so as to increase the number of conformation. As the ($Hg^{2+} + Te^{4+}$)–ssDNA complex is now positively charged, upon impression of a cathodic current, it is dragged to the cathode [24]. After ionic discharge at the cathode, the HgTe–ssDNA complex NCs are deposited on its surface.

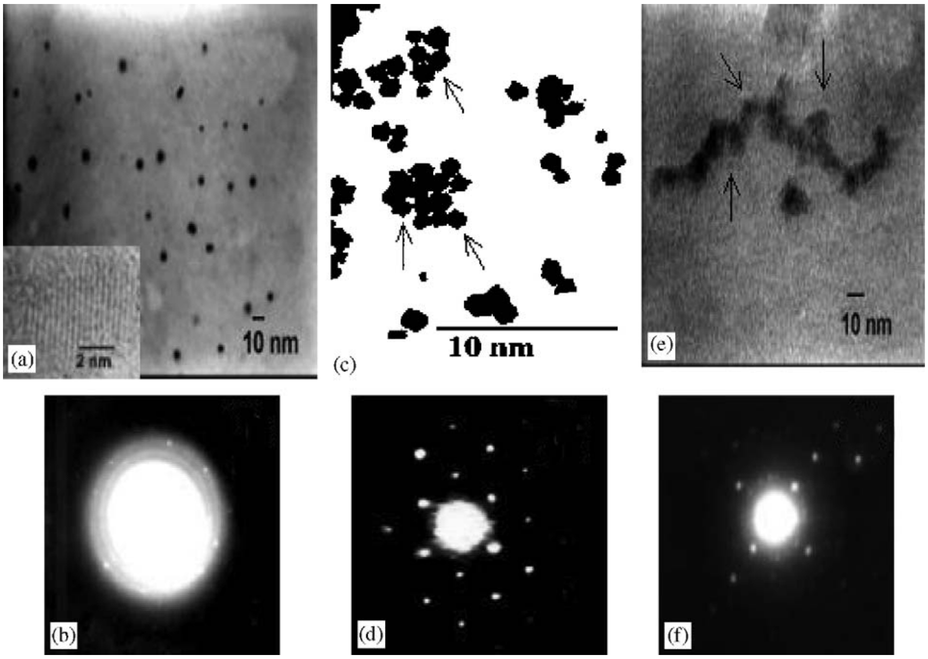

Fig. 1. TEM micrograph of HgTe nanoparticles (a). The inset shows the high-resolution lattice image and (b) is the corresponding electron diffraction pattern. (c) is the TEM micrograph of HgTe–ssDNA complex nanostars and (d) is the corresponding electron diffraction pattern. (e) is the TEM micrograph of HgTe–poly G (30) and (f) is the corresponding electron diffraction pattern.