Hierarchical Assembly of Colloidal Lead Chalcogenide Nanocrystals for High Carrier Accumulation in Electrical Double Layers Capacitor

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The solution-processable semiconductor colloidal nanocrystals (NCs) offers attractive properties beyond their bulk which is attributed to the size-dependent band-gap due to the quantum confinement effect of the electron wave function. This intriguing feature makes the NCs materials suitable for optoelectronic application related to the energy harvesting (i.e. photovoltaics cell), light-emitting-display, photodetectors as well as the thermoelectric devices application.[1] From the other point of view, the finite density of state (discrete energy level) of NCs materials can emerge the formation of quantum capacitance that has a much higher value compared to the bulk capacitance. Moreover, the large surface area-to-volume ratio on NCs materials can multiply the value of capacitance on the NCs assembly itself. By choosing the compatible electrolyte (i.e. ionic liquid), the high carrier accumulation on the interface of ionic liquid/NCs can be achieved as the electric double layer (EDL) formed.

Here we demonstrate the measurement of the carrier accumulation on the controllable crosslinked assembly of colloidal lead sulfide (PbS) NCs. The morphology of the PbS NCs assembly was controlled by different deposition methods which provide a superlattice to the hierarchical porous structure of the NCs assembly by liquid/air interfacial assembly and dip-coating method, respectively. The carrier accumulation at the interface of ionic-liquid/NCs assembly was firstly evaluated by electrolyte-gated field effect transistor (FET) through the displacement current measurement (DCM).[2] To confirm the result of the carrier accumulation from DCM, we compare to the larger area of measurement by applying the EDL capacitor structure to examine the whole capacitance value of the assembly. The total capacitance was evaluated by the cyclic voltammetry measurement. As a result, the high recorded areal capacitance up to ~40 μF/cm^2 (at low voltage sweep rate, 10 mV/s) can be achieved from the porous structure assembly due to the larger surface area compare to the compact NCs superlattice assembly, in contrast. Moreover, this finding[3] shows that the NCs materials have a comparable capacitance value to the exist supercapacitor from another group of semiconductors.[4] Therefore, the dip-coating technique can be applied for other NCs materials in order to control the hierarchical porous structure for obtaining higher carrier density for the near future.

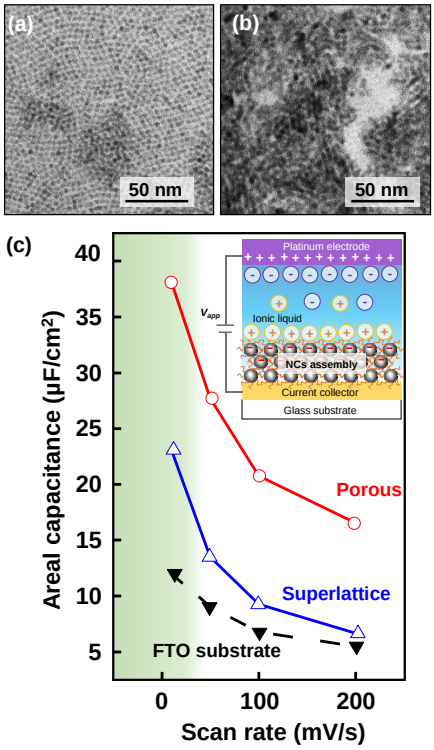

Figure 1. TEM images of PbS NCs (a) superlattice assembly and (b) hierarchical porous structure, (c) areal capacitance of PbS NCs EDL capacitor by using [EMIM][TFSI] ionic liquid. The comparison of capacitance value between superlattice and porous structure of assembly.

Refs.:[1] C. R. Kagan, C. B. Murray. *Nat. nanotech.* 10.12: 1013-1026 (2015), [2] Bisri et al. *Adv. Mater.* 25.31 (2013): 4309-4314, [3] R. D. Septianto, S. Z. Bisri et al. (2019) *submitted*, [4] Peng et al. *Adv. Energy Mater.* 8.9 (2018): 1702179