

Abstract

The unavailability of transparent hole-transporting inorganic semiconductors that can be solution-processed, has good stability, and shows excellent carrier transport characteristics remains one of the challenges in the field of organic electronics. This work aims to develop such semiconductors by focusing on coordination compounds based on the thiocyanate ligand. Our earlier investigations have shown that copper(I) thiocyanate (CuSCN) is a promising candidate as it shows hole mobility on average of $0.01 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and has a large optical band gap of 3.9 eV.

For the first part of the work, we expand the thiocyanate-based semiconductor library by showing that tin(II) thiocyanate [$\text{Sn}(\text{NCS})_2$] is also a wide band gap semiconductor with promising electronic structure for hole transport. We synthesized the compound and comprehensively characterized it with a wide range of experimental techniques as well as studied theoretically with density functional theory (DFT). We can reveal the low-dimensional nature of the compound through in-depth analysis of the single crystal X-ray data. $\text{Sn}(\text{NCS})_2$ is found to have a 1D polymeric chain that further form 1D ribbons and 2D sheets. The DFT calculations show that $\text{Sn}(\text{NCS})_2$ has a wide band gap, a feature which is backed up by the experimental absorption spectrum that shows the optical band gap of 3.4 eV. The analysis of the density of states also shows that the Sn 5s electrons contribute to the states at the top of the valence band. These states are expected to favor good carrier transport. Furthermore, we explore the application of $\text{Sn}(\text{NCS})_2$ as a hole transport layer in organic solar cells. Initial results of unoptimized systems are highly promising with solar cells showing efficiencies in the range of 4-6%. We expect that further optimizing of the structure and processing would enhance the efficiency further.

We also studied the effects of treating CuSCN thin films with anti-solvents on the film morphology and device applications. By treating CuSCN with tetrahydrofuran, *p*-type thin-film transistors based on CuSCN as the semiconducting channel show improved characteristics of higher on current and increased field-effect hole mobility from 0.01 to $0.05 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. On the other hand, when CuSCN is deposited on glass/ITO substrates for organic solar cell device, treating CuSCN with isopropyl alcohol is found to increase the device power conversion efficiency from 8.35% to 8.93%. We have demonstrated that the simple anti-solvent treatment is a versatile and easily applicable method for improving the performance of CuSCN-based devices.

Through this project, we have shown that the 'coordination polymer semiconductors' are a novel family of compounds that have promising electronic properties. They can be solution-processed, and further modifications can be done based on coordination chemistry. Different metals and ligands await the exploration, and this the subject of our next project.