Studies of nano-carbon hole transport layer for high performance photovoltaic devices

by

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Low-dimensional nano-carbon materials including the form of zero-dimensional (0D) fullerenes, 1D nanotubes, and 2D graphene with extraordinary electronic and optoelectronic properties, provide a large impact on the research field of nanotechnology. The extreme electron confinement in the low-dimensional nano-carbon materials endows them with unique electronic structure and properties, which have been extensively studied from the viewpoint of fundamental studies and device applications.

Carbon nanotube is a very important member of low-dimensional carbon materials, because it has the superior characteristic of high carrier mobility, high specific surface area, high mechanical properties, and tunable bandgap. The fundamental researches on the optoelectronic applications have been extensively studied in the carbon nanotubes. Indeed, the photocurrent has been achieved in single-walled carbon nanotubes. The carbon nanotubes also show high electronic conductivity and have been used as the electrode and carrier transport layer in the opto-electronic devices. Furthermore, the carbon nanotubes have excellent elastic properties, which are very suitable for the flexible and portable optoelectronic devices.

In this thesis, I applied the carbon nanotube for the photovoltaic solar cell devices. I conducted the fundamental studies of photovoltaics of carbon nanotube solar cells with the combination of well-defined materials (crystalline silicon) and fabricated the carbon nanotube/Si heterojunction solar cells. In the carbon nanotube/Si heterojunction solar cells, I could not detect the obvious photocurrent contributions from carbon nanotubes, which suggests that the carbon nanotube work as a hole extraction/transport layer. Therefore, I found that the electrical conductivity of carbon nanotubes becomes very important for improving the photovoltaic performance of the solar devices, because the carbon nanotube also works as a semi-transparent electrode in the devices.

First, I have started the studies of carbon nanotube/Si heterojunction solar cells using the sprayed single-walled carbon nanotube film, and studied *p*-carbon nanotube/*n*-Si heterojunction solar cells for understanding the series resistance effects and the photovoltaic mechanism of device. From the model and calculation, the resistance is recognized as the main culprit of the relatively low performance in solar cells. The enhancement of the conversion efficiency was realized by HNO_3 doping which decreased in the series resistance of the carbon nanotube network films and increased in the *p*–*n* junction density.

As a next step, I used high quality carbon nanotube thin film fabricated by the floating catalytic chemical vapor deposition method for further improvement of photovoltaic performance. The sheet resistance of carbon nanotube fabricated by this method is about 4 kΩ/sq with a transparency of 91% at 550 nm, which is much lower than sprayed nanotube film with the same transparency. The carbon nanotube/Si heterojunction solar cell using this high quality carbon nanotube films shows a photovoltaic conversion efficiency of 12% without post-processing. In addition, the high reproducibility of the photovoltaic performance of this type of device was clearly demonstrated, suggesting the superior potential of carbon nanotube solar cells.

I also investigated the photovoltaic performance of carbon nanotube-based solar cells using environmentally friendly, durable, and inexpensive materials such as MoO_x and ZnO, which work as antireflection layers for increasing the optical absorption, carrier dopants for a reduction of the series resistance, and effective carrier transport layers for the reduction of the Schottky barrier. The photovoltaic performance of *p*-carbon/*n*-Si and *n*-carbon/*p*-Si heterojunction solar cells was improved using multifunctional layers of MoO_x and ZnO, which resulted in very high photovoltaic conversion efficiency values of 17.0% and 4.0%, respectively. Finally, the photovoltaic performance of organo-lead iodide perovskite solar cells with efficient carbon nanotube/graphene oxide hole-transport layers was studied. A perovskite/carbon nanotubes/graphene oxide solar cell with a protecting layer of polymer exhibited a high photovoltaic conversion efficiency of 13.3%, which was comparable to that of a perovskite solar cell containing a well-sued standard organic hole-transport layer. Moreover, it was demonstrated that the photovoltaic conversion efficiency value for the perovskite/carbon nanotube/graphene oxide/polymer decreased only slightly compared to that of the standard perovskite solar cell using organic hole transport layer in air condition. With their respective efficient carrier extraction/transport and electron-blocking properties, the combination of carbon nanotube and graphene oxide in the perovskite solar cell contributes to a reduction in carrier recombination losses and an increase in the open-circuit voltage, resulting in a high photovoltaic conversion efficiency.

These findings regarding the performance of carbon nanotube films as an efficient carrier transport layer shed new light on device physics and suggest that these materials have significant potential for use in optoelectronic applications.

Published papers

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[3] Feijiu Wang, D. Kozawa, Y. Miyauchi, K. Hiraoka, S. Mouri, Y. Ohno, K. Matsuda, Considerably Improved Photovoltaic Performance of Carbon Nanotube-based Solar Cells using Metal Oxide Layers. *Nature Communications*, 2015, 6, 6305.

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