Application of pre-dyeing method to flexible dye-sensitized solar cell based on zinc oxide nanoparticle

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学 位 論 文 要 旨

Abstract of Doctoral Thesis

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論文題目：

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酸化亜鉛ナノ粒子に基づくフレキシブル色素増感太陽電池における先染法の応用

論文要旨：

Abstract：

With the global warming and the increasing demands for energy, the necessity of stemming increases in carbon dioxide emissions. Thus, it has become quite essential to develop renewable energy sources and to develop non-polluting, clean sources of energy are becoming increasingly crucial. Solar energy holds the possibility of can solve these energy problems. However, the relatively high production costs of ordinary solar cells have limited their widespread commercialization. But the original paper by O'Regan and Grätzel in 1991 reported dye-sensitized solar cells (DSSC) are one of the next-generation solar cells because of their relatively easy fabrication procedures, high energy conversion efficiencies (~15%) and low production cost. Especially, with the increasing of the requirement for the portable and convenient applications, flexible substrates have been extensively investigated to be used for the solar cells. Flexible solar cells based on plastic, paper or metal foil possess more advantages than the glass-based ones. In particular, flexible DSSCs using thin and light-weight conducting plastic substrates have attracted much interest for their potential of mass production by the roll-to-roll process. In the traditional preparation of the mesoporous photoelectrode of the DSSCs, a thermal treatment higher than 450ºC is necessary, with which the photoelectrode can have a good adhesion between the particle-particle and particle-substrate. However, the plastic substrate can only bear the temperature lower than 150ºC. In this study, we developed a simple method in which dyeing process is applied to ZnO nanoparticle then pre-dyed ZnO (pd-ZnO) paste is applied to transparent electrode substrate to form colored nanoporous ZnO photoelectrode. We call this process as pre-dyeing method. By employing this method, time-consuming dyeing process can be removed from roll-to-roll process. However, the efficiency of DSSC made by pre-dyeing method was much lower than traditional method. It is conceivable that the whole surface of ZnO particle could be covered with dye, resulting in deterioration of the contact between the ZnO particles and reduction of the electrical conductivity. In order to improve lower porosity and bad interparticle connectivity, hot-press method was applied to photoelectrode on plastic substrate (ITO/PEN). This is regarded to improve the contact between the particles by pressing, but at the same time, the pores of the porous thin film are buried, the surface area decreases, lead to the amount of dye adsorption decreases.

After hot-press treatment, adhesion of pd-ZnO film to the flexible substrate can be improved, and the compact layer becomes more homogeneous. It was possible to prepare a dye-sensitized solar cell with a conversion efficiency of 1.97% by adjusting the proper quantities of dye to the zinc oxide and subjecting it to a hot press treatment using EosinY dye. In order to reach higher efficiency, D149 dye was used which has broader absorption wavelength range than EosinY dye. The solar-to-electric conversion efficiency of 4.56% under 1 sun illumination was achieved by utilizing the hot-press technique on the low-temperature preparation of nanostructured ZnO films.

Absorption of photon in wide wavelength region is an important requirement for the enhancement of photoconversion efficiency of DSSCs. Some researchers reported that a mixture of two kinds of dye which has different wavelength absorption show the better performance to DSSCs by virtue of the absorbing the light of wider wavelength range. By employing pre-dyeing method, simple mixture of pd-ZnO powders adsorbed different dye was prepared and applied to DSSCs. Three kinds of dyes D131, D358 and EosinY show yellow, purple and pink color, respectively. Mixed pd-ZnO paste was easily prepared by simple mixed two kinds of pd-ZnO powder in 1:1 ratio. Efficiency improvements are observed in three kinds of DSSC made of pd-ZnO mixed film. For example, DSSC based on mixture of D131 and Eosin Y pd-ZnO shows appreciable increase of IPCE at 400 nm wavelength region compared with single Eosin Y pd-ZnO based DSSC. The photoenergy conversion efficiency of mixed pd-ZnO based DSSCs are higher than that of single pd-ZnO based DSSCs except for the case of D358 and Eosin Y combination because light absorption wavelength ranges of D358 and Eosin Y are overlapped each other. In our successive investigation for various dye combination, the best results was obtained by purple D149 and yellow D131 combination. The photovoltaic parameters of the best DSSC were Jsc = 9.73 mA cm^{-2}, Voc = 0.71 V, FF = 0.68 and η =4.56%. It is notable that not only Jsc improvement but also higher Voc contribute the improvement of efficiency. Although the reason of this Voc improvement is unclear yet, it is suggested that multiple dye combination using pre-dyeing method has some potential for improving not only Jsc by expanding absorption wavelength range but also other photovoltaic parameters such as Voc. Wide range wavelength light absorption by virtue of dye combination can contribute the improvement of short circuit current. To the best of our knowledge, this is first successful example of colored paint (pd-ZnO paste) based DSSCs. Pre-dyeing method is promising especially for the production of flexible DSSCs made by roll-to-roll process.