Fabrication of a Combustion-Reacted High-Performance ZnO Electron Transport Layer with Silver Nanowire Electrodes for Organic Solar Cells

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ABSTRACT: Herein, a new methodology for solution-processed ZnO fabrication on Ag nanowire network electrode via combustion reaction is reported, where the amount of heat emitted during combustion was minimized by controlling the reaction temperature to avoid damaging the underlying Ag nanowires. The degree of participation of acetylacetones, which are volatile fuels in the combustion reaction, was found to vary with the reaction temperature, as revealed by thermogravimetric and compositional analyses. An optimized processing temperature of 180 °C was chosen to successfully fabricate a combustion-reacted ZnO and Ag nanowire hybrid electrode with a sheet resistance of 30 Ω/sq and transmittance of 87%. A combustion-reacted ZnO on Ag nanowire hybrid structure was demonstrated as an efficient transparent electrode and electron transport layer for the PTB7-Th-based polymer solar cells. The superior electrical conductivity of combustion-reacted ZnO, compared to that of conventional sol–gel ZnO, increased the external quantum efficiency over the entire absorption range, whereas a unique light scattering effect due to the presence of nanopores in the combustion-derived ZnO further enhanced the external quantum efficiency in the 450–550 nm wavelength range. A power conversion efficiency of 8.48% was demonstrated for the PTB7-Th-based polymer solar cell with the use of a combustion-reacted ZnO/Ag NW hybrid transparent electrode.

KEYWORDS: combustion, solution-processed, zinc oxide, calorific value, organic solar cell, light scattering

1. INTRODUCTION

Organic solar cells (OSCs) have attracted much attention due to not only their high performance but also their low cost, light weight, and flexibility,1–4 making them a promising candidate for replacing conventional inorganic solar cells. Significant advances in power conversion efficiency (PCE) have been made with efficiencies of >10% being reported,5–7 and thus the OSC is advancing toward meeting the industrial power output requirement for flexible device applications. However, conventional transparent conducting oxides paired with OSCs, such as indium tin oxide (ITO), may be unsuitable for flexible solar cells due to their intrinsically brittle nature. Various one-dimensional or two-dimensional materials, such as carbon nanotube, graphene, and metal nanowire (NW), have been introduced as potential replacements for ITO, and among them,5–13 silver nanowire (Ag NW) network has been extensively investigated for flexible transparent electrode application owing to its excellent optical and electrical properties, low cost, solution-based synthesis, as well as mechanical flexibility.

The drawback of Ag NW network application in solar cells, however, is insufficient surface coverage; the Ag NW network covers less than 40% of the entire electrode area, which leaves much of the solar cell without contact with a conductor, which can lead to a decrease in the cell efficiency.12 There have been numerous attempts to overcome this issue, and the use of an auxiliary transparent conductor was found to be suitable. For example, poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) coating on Ag NW can solve the coverage issue to improve the overall conversion efficiency of the device,13–15 but the corrosive nature of PEDOT:PSS can harm the reliability of the Ag NWs. Sputter deposition of ZnO on Ag NW network has also been reported to solve the coverage issue as well as enhance the thermal and chemical stability of the Ag NW electrode,16–18 but the cost associated with the high-vacuum processes remains a challenge. Application of ZnO nanoparticles to Ag NW networks was suggested to solve the coverage issue with low cost, but use of fine particles typically results in low conductivity as well as being less effectiveness in passivation due to the presence of many large-sized pores and

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Thus, the development of solution-processed metal oxides with sufficient optical and electrical properties is of critical interest.

In an inverted structure of OSCs, an electron transport layer (ETL) is coupled with the ITO to serve as a hole blocking layer for enhanced efficiency. ZnO is a widely adopted choice for an ETL due to its high electron mobility as well as its potential use of a low cost, solution-based coating process such as the sol–gel method. When replacing the ITO with Ag NW network, the ZnO overlayer should, therefore, serve as both the ETL and the auxiliary transparent electrode. However, the challenge is to synthesize high-quality ZnO that can function as an auxiliary conductor without elevating the processing temperature beyond 200 °C, above which Ag NWs are known to be thermally unstable. Therefore, there exists a need to develop an alternative method for obtaining a high-quality ZnO layer via solution processing at a lower temperature while ensuring sufficient conductivity for the charge carriers to travel through the ZnO layer before reaching the Ag NW network.

Among numerous studies in the development of producing high-quality oxide thin films at low temperature, the combustion reaction method was recently highlighted to be effective in achieving high electrical and optical performance. During the combustion-reacted ZnO synthesis of thin film, high-quality ZnO can be fabricated via the combustion reaction at 200 °C and the reaction completes within a short time in comparison with sol–gel methods. Although the reaction temperature is at 200 °C, the localized temperature due to release of the thermal energy is expected to be higher. Thus, if one can lower the thermal energy released during the combustion reaction, the combustion-reacted method could be a useful technique for achieving high-quality ZnO ETLs without damaging the underlying Ag NW network.

In this study, the fabrication of high-quality ZnO was demonstrated by controlling the amount of heat emitted during the combustion reaction, which allowed for a high-quality ZnO layer and also prevented thermal damage on the underlying Ag NWs. Using the developed combustion reaction method, the ZnO/Ag NW hybrid electrode was successfully fabricated and integrated into PTB7-Th:PC70BM-based polymer solar cell, which demonstrated an improved efficiency in comparison to that with conventional sol–gel ZnO/ITO or sol–gel ZnO/Ag NW electrodes. Furthermore, we report a unique optical behavior in which the combustion-reacted ZnO electrode in presence of nanopores improved light scattering, resulting in enhancement of the quantum efficiency.

2. EXPERIMENTAL SECTION

2.1. Fabrication of Silver Nanowire Transparent Electrode and ZnO ETL. Silver nanowire solution (Nanopyxis, Korea) is air-sprayed on a precleaned glass substrate to obtain a sheet resistance value of 25 Ω/sq. After spraying, Ag NW network is annealed at 200 °C in air for 10 min, followed by acetone wash for 1 min. For sol–gel ZnO coating, zinc acetate (Sigma-Aldrich) is dissolved in 2-methoxyethanol (Sigma-Aldrich) at a concentration of 0.4 M. Monoethanolamine (Sigma-Aldrich) is also dissolved in zinc acetate solution with a 1:1 ratio for zinc precursor. After aging for 24 h in room temperature, the solution is spin-coated on ITO substrate or on top of Ag NW network at 6000 rpm, followed by annealing at 200 °C for 1 h. For combustion ZnO, a 1:1 ratio of zinc nitrate (Sigma-Aldrich) and zinc acetylacetonate (Sigma-Aldrich) is dissolved in 2-methoxyethanol at a Zn concentration of 0.2 M. Undissolved large particle residues are removed by 0.45 μm syringe filter, and the solution is diluted to 0.05 M. The solution for combustion-reacted ZnO is spin-coated on top of Ag NW at 2500 rpm for 30 s, followed by annealing at 180 °C for 30 s for each coating and annealing step. Eight times of coating with 0.05 M solution is conducted for 50 nm thickness of combustion-reacted ZnO.

2.2. Device Fabrication. PTB7-Th:PC70BM (1-materials:Nano-C) at a weight ratio of 1:1.5 was dissolved in chlorobenzene/1,8-diiodooctane (97:3); it was spin-coated on the prepared electrode (ZnO on ITO or Ag NW) at 2000 rpm speed for 30 s, and methanol was immediately spin-casted on the PTB7-Th:PC70BM layer. Thereafter, substrates were loaded into a vacuum chamber (pressure < 1.0×10⁻⁷ Torr). Finally, a 10 nm layer of MoO₃ and 150 nm layer of silver electrode were sequentially deposited by thermal evaporation. Other devices were fabricated with the same method on Ag NW/ZnO hybrid electrodes instead of ITO. For flexible devices, exactly the same materials and method are used except for replacing Willow glass (Corning).

2.3. Characterization. Field emission scanning electron microscopy (FE-SEM, Magellan400, FEI company) was used for characterization of the fabricated hybrid electrode morphology. Thermogravimetric analysis (TGA) (TG209 F1 Libra, Netzsch) is used to measure the quantity of emitted heat during the combustion reaction. Sheet resistance values are obtained by a four-point measurement system (FPP-2000A, Dasoleng, Korea), and diffusive transmittance and haze were measured using a UV–vis spectrometer (UV-3600 plus; Shimadzu). X-ray photoelectron spectroscopy (XPS) analysis is performed using the Sigma Probe system (Thermo VG Scientific), with a base pressure of 1 × 10⁻⁸ Torr using Al Kα sources. TOF-SIMSS (ION-TOF GmbH) is used with Cs⁺ ion beam at 0.5 keV energy. A D/Max-2500 X-ray diffractometer (Rigaku) is utilized to determine the phases and crystallinity of the ZnO thin film. Transmission electron microscopy (TEM) analysis for detailed microstructural analysis is performed using Titan cubed G2 60-300 (FEI company), with an acceleration voltage of 80 kV. Atomic force microscopy measurement is used to evaluate the surface roughness of ZnO thin film byXE-100 (Park systems) in tapping mode. For measuring electrical conductivity of doped/undoped ZnO, Keithley 4200SCS (Tektronix) is used with the four-point van der Pauw method and two-point I−V measurement. The current density–voltage (J−V) characteristics are measured by a solar simulator (K3000 LAB55 System; McScience Inc.) under 100 mW cm⁻² with the shadow mask that has a size of 0.0625 cm². The quantum efficiency and absorption for calculating internal quantum efficiency are operated by a spectral measurement system (K3100 IQX, McScience Inc.).
up to 10 min of annealing at 200 °C in air without an abrupt increase in resistance. Spray-coated Ag NWs were annealed at 200 °C to form a network with fused in junctions, which results in optimized electrical optical properties ($T = 95\%$, $R = 20 \Omega/\text{sq}$). ZnO solution for the combustion reaction was then coated repeatedly at varying temperatures of 200, 180, and 160 °C on top of the Ag NWs. The overall sheet resistance evolution after each ZnO coating is shown in Figure 1a. The combustion reaction carried out at 180 and 160 °C showed a slower increase in sheet resistance as the number of cycles increased and eventually saturated at 25 and 22 $\Omega/\text{sq}$, respectively. For the combustion reaction temperature of 200 °C, the sheet resistance of the electrode increased more rapidly with each coating due to the excessive heat that caused the underlying Ag NW to ball-up to result in disconnection, which is a commonly observed thermal instability behavior in Ag NWs (Figure 1c,d). More damage on the Ag NW network at higher processing temperatures indicated that larger heat emission occurred during the combustion reaction. Therefore, the amount of heat emission is dependent on the annealing temperature and the mechanism that caused such dependence was further investigated.

In a combustion reaction, the amount of heat released is in theory determined by the enthalpy difference between the reactants vs the products and thus should not vary with the reaction temperature. To clarify this mismatch between theory and experimental results, solution for the combustion-reacted ZnO was thermogravimetrically analyzed to compare the calorific value with respect to the reaction temperature. It should be noted that the TGA analysis condition differs from the actual coating and annealing process; the combustion reaction of ZnO is carried out by direct exposure to the preheated hot plate, whereas the TGA analysis involves heating the solution from room temperature to the set point at a relatively slow heating rate. TGA analysis was conducted at heating rates of 2, 5, and 10 °C/min. Interestingly, the amount of emitted thermal energy and temperature at which the reaction is initiated during the combustion reaction are greater at higher heating rates, as shown in Figure 1b. It can be deduced that the release of thermal energy can be suppressed when the combustion reaction is carried out at a low temperature, which would thus limit any damage on the underlying Ag NW network after combustion ZnO coating, as shown in Figure 1a,b.

3.2. Mechanism of Heat Emission Loss. The difference in calorific value in relation to the heating rate can be attributed to the different degrees of reactant participation during the combustion reaction at different reaction temperatures. In the combustion reaction solution, nitrate serves as an oxidant and acetylacetonate acts as a fuel for driving combustion. Acetylacetonate is highly volatile in the form of acetylacetone and can evaporate into air before and during the combustion reaction, and this loss of combustible fuel results in a decrease in the actual released heat and causes a larger amount of unreacted nitrate to remain after the reaction terminates.

The amount of nitrate that serves as an oxidant in the combustion reaction was analyzed as a function of different processing temperatures using time-of-flight secondary ion mass spectroscopy (TOF-SIMS). Higher nitrate intensities were detected in samples annealed at lower temperatures and these results are consistent with XPS results, as shown in Figure 2 and Table S1. Considering that all N species originated from
the nitrate, it can be explained that varied caloric values at each temperature result from the different degrees of participation of fuel (acetylacetonate) and oxidant (nitrate). Another interesting point confirmed by TOF-SIMS analysis is that the proportion of these N species was much higher near the surface, which constantly decreased with an increase in depth. Such gradient in concentration can be explained considering that each repeated annealing step may lead to the newly deposited solution to provide the necessary fuel to react with the remaining nitrate reactant in the underlying layer. The fact that the trace content of C does not change with respect to the annealing temperature also indicates that the varied degrees of participation of fuel are represented in the form of evaporation, not remaining as unreacted C species in ZnO.

3.3. Influence of Varied Calorific Values on the Quality of ZnO. The microstructure and composition of the fabricated combustion-reacted ZnO thin film are heavily dependent on the calorific value, which in turn is dependent on the annealing temperature. As shown in the cross-sectional SEM images of combustion-reacted ZnO thin films at various annealing temperatures in Figure 3a, a clear relationship between the annealing temperature and microstructure of the thin film can be observed. At low annealing temperatures, the ZnO thin film is composed of irregular-shaped nanoparticles with some porosity rather than being a dense thin film. At higher annealing temperatures, a denser arrangement of spherical grains with a low porosity that resembles a dense thin film is formed. Besides, there is small difference between the 190 and 200 °C annealed samples and it means that the effect of heat emission must be similar in both cases of the 190 and 200 °C annealed samples such that a similar compact microstructure with low density of pores was found in both. A further magnified TEM image in Figure 3b shows the presence of nanosized pores inside the combustion-reacted ZnO thin film annealed at 200 °C, and these nanopores are a resultant of trapped gas during the rapid combustion reaction.23

Mostly, one of the critical factors determining the quality of solution-processed metal oxide is the amount of phase conversion from hydroxide (M−OH) to oxide (M−O). Variation in the quantity of emitted heat was also shown to cause different degrees of oxide formation, as revealed by XPS and TOF-SIMS composition analysis. Figure 4 shows the XPS spectra of O 1s of combustion-reacted ZnO thin films at various temperatures. The percentage of metal oxide bonding ($E_b = 528.2$ eV) in comparison to the metal hydroxide bonding ($E_b = 530$ eV) showed that combustion-reacted ZnO annealed
at 170 °C has 78.6% Zn−O bonds, whereas the ZnO annealed at 200 °C has almost 80% Zn−O bonds.21,25 TOF-SIMS analysis provided similar results, indicating the intensity ratio between 68ZnO and 68ZnOH to be 8.33:1 and 2.56:1 at annealing temperatures of 200 and 170 °C, respectively, indicating that a greater fraction of reactants form ZnO at a higher annealing temperature (Figure S1). Thus, both XPS and TOF-SIMS compositional analyses confirmed that the difference in calorific values resulting from different annealing temperatures influenced the degree of the dehydration reaction, which is critical for the hydroxide-to-oxide phase transformation.

Although combustion-reacted ZnO annealed at a lower temperature showed less Zn−O bond formation with more impurities than those annealed at high temperatures, its quality or performance such as the conductivity were still significantly improved over that of the conventional sol−gel ZnO, as shown in Figure 5. Eight times coated and annealed ZnO thin films fabricated by both the combustion reaction and sol−gel methods were evaluated by XPS and X-ray diffraction (XRD). In XPS analysis, as shown in Figure 5a, the sol−gel ZnO thin film annealed at 200 °C showed only 60% Zn−O bonding, which is consistent with previous reports,18 whereas the combustion-reacted ZnO thin film showed almost 80% Zn−O bonding. The differences between combustion ZnO and conventional sol−gel ZnO extend to the crystallinity of the fabricated thin film. Glazed incidence angle X-ray diffraction analysis was conducted that showed a higher degree of crystallinity for the combustion-reacted ZnO annealed at 200 °C than that for the sol−gel ZnO (Figure 5b).

It can be easily expected that the quality difference between the combustion-reacted ZnO and sol−gel ZnO causes changes in conductivity. Both Al-doped and undoped ZnO thin film fabricated by two distinct methods were compared. The change in the conductivity of Al-doped ZnO thin films produced by the combustion reaction with respect to the annealing temperature is plotted in Figure 5c. Al-doped combustion-reacted ZnO showed an enhanced conductivity at higher annealing temperatures due to the low porosity, reduction in the residual N content, and increased oxide/hydroxide ratio. Especially, it is well known that presence of impurities is quite a critical factor for determining the electrical properties of semiconductors. Residual N elements which are major impurities in combustion-reacted ZnO can act as an acceptor in semiconductor oxide by substituting oxygen sites and function as a dopant compensator in the intrinsic or n-type Al-doped ZnO semiconductor, thereby reducing the doping effect.26−28 Furthermore, NOx can be chemisorbed on the surface, which causes a decrease in conductivity by creating defect sites.29−31 A lateral conductivity of 0.217 S/cm at an annealing temperature of 200 °C was obtained for the combustion-reacted Al-doped ZnO thin film, which is higher than that of (1.5 × 10−3 S/cm) sol−gel Al-doped ZnO obtained at 250 °C marked as the dotted line in Figure 5c. For the undoped ZnO case, combustion-reacted ZnO showed resistivity of 2.8 × 103 Ω cm in a dark condition, whereas sol−gel ZnO showed 1.9 × 105 Ω cm, as shown in Figure 5d. Therefore, it can be said that both the combustion-reacted Al-doped or undoped ZnO has relatively high conductivity compared to that of doped or undoped sol−gel ZnO.
3.4. Unique Properties of Combustion-Reacted ZnO and Application to Organic Solar Cell. A PTB7-Th:PC$_{70}$BM-based organic solar cell (OSC) has a high power conversion efficiency that can be made flexible with an
The average PCEs of 10 cells.

An additional increase in EQE in the range of 450–550 nm is attributed to the increased absorption from the light scattering effect of combustion-reacted ZnO, which has nanopores as explained below. The combustion reaction occurs in a shorter timeframe with more release of combusted gases compared to that in the sol–gel reaction. As a result, nanosized pores are formed within the ZnO layer, as confirmed by SEM and TEM images shown in Figures 1c,d and 3. A higher degree of porosity equates to greater density of air and ZnO interfaces within the ETL, where light is highly scattered, as shown in Figures 6c,d, S3 and S5. In the case of the 50 nm thick ZnO thin film, the absolute haze value was not significant; however, this became pronounced in the case of the hybrid with Ag NW network. As shown in Figure 6c, the combustion-reacted ZnO/Ag NW electrode shows a higher haze value compared to that of the sol–gel ZnO/Ag NW, indicating that combustion-reacted ZnO amplifies the light scattering effect of the Ag NW network. This light scattering effect can be explained by Rayleigh scattering as the pore size in the combustion-reacted ZnO thin film is about a few tens of nanometers, as shown in Figure 3, which is less than 1/10 of the wavelength of visible light.55–57 Because of the difference in the refractive index between ZnO and air, strong scattering takes place and this effect becomes more pronounced in the short wavelength region. Haze tends to become higher in the short wavelength region, and it results in an increase of EQE enhancement values from 550 nm to shorter wavelength for solar cell devices. However, below the 450 nm, the absorption of light by ZnO becomes dominant even though much higher scattering effects are exhibited. Therefore, enhanced light absorption of the active layer by combustion ZnO’s scattering effect becomes negligible below 450 nm. Through these light scattering and absorption effects by combustion ZnO, EQE enhancements are pronounced in the range of 450–550 nm.

To demonstrate the improved light scattering ability, a red laser beam was directed through the side of the glass substrate with ZnO and is shown in Figure 6d–f. The incident laser beam passing through the ZnO/glass structure can be clearly observed due to the intense light scattering effect of the combustion-reacted ZnO thin film, whereas it was not observed in the sol–gel ZnO thin film. Higher intensity of light was observed in combustion-reacted ZnO/Ag NW hybrid electrode due to the amplified light scattering effect.

Table 1. Device Performance Parameters of Fabricated PTB7-Th Organic Solar Cell with ITO/ZnO (sol–gel), Ag NW/ZnO (Combustion), and Ag NW/ZnO (Sol–Gel)

<table>
<thead>
<tr>
<th>cathode</th>
<th>Voc [V]</th>
<th>Jsc (EQE) [mA/cm²]</th>
<th>FF</th>
<th>PCEabs [%]</th>
<th>PCEmax [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>reference ITO</td>
<td>0.79 ± 0.00</td>
<td>16.26 ± 0.06 (16.86)</td>
<td>0.62 ± 0.01</td>
<td>7.93 ± 0.10</td>
<td>7.98</td>
</tr>
<tr>
<td>Ag NW + sol–gel ZnO</td>
<td>0.80 ± 0.00</td>
<td>15.62 ± 0.18 (15.41)</td>
<td>0.63 ± 0.02</td>
<td>7.70 ± 0.19</td>
<td>7.91</td>
</tr>
<tr>
<td>Ag NW + combustion ZnO</td>
<td>0.80 ± 0.00</td>
<td>16.48 ± 0.17 (17.00)</td>
<td>0.64 ± 0.01</td>
<td>8.21 ± 0.22</td>
<td>8.48</td>
</tr>
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The average PCEs of 10 cells.

References:
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Having developed a combustion-reacted ZnO thin film of high quality, conductivity, and crystallinity with optimized process conditions for OSC applications, we fabricated a flexible solar cell using the combustion-reacted ZnO/Ag NW hybrid electrode. The demonstration takes full advantage of the flexibility of the Ag NW network. A PTB7-Th:PC_70 BM-based organic solar cell with the combustion-reacted ZnO/Ag NW hybrid electrode structure was successfully fabricated on a flexible Willow glass with a PCE of 7.30%, as shown in Figure S6a,b.

4. CONCLUSIONS

In this study, a new methodology of fabrication of solution-processed ZnO thin film via combustion reaction with minimal damage to the underlying Ag NWs was developed. Heat emission during the combustion reaction was shown to be controlled by varying the annealing temperature; the mechanism was determined to be due to more loss of volatile fuel in the combustion reaction at lower temperature that hence results in unreacted nitrate and thus less heat emission. The optimized combustion-reacted ZnO thin film on Ag NW electrode was applied to PTB7-Th-based organic solar cell to confirm a much improved power conversion efficiency value of 8.48% compared to that of the conventional ITO-based device. The improvement is attributed to the high conductivity and high light scattering ability of combustion-reacted ZnO. The advantages of the combustion-reacted ZnO fabrication method includes cost effectiveness and scalability while allowing for all solution-processed synthesis that is expected to have a significant impact in various flexible optoelectronic devices, especially in the field of organic solar cells.

REFERENCES


ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.7b17148.

Additional TOF-SIMS data for the combustion-reacted ZnO thin film; optical and electrical properties for fabricated electrodes and flat and curved solar cells; experimental setup for the laser incident experiment and surface roughness measurement for ZnO films (PDF)

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Notes

The authors declare no competing financial interest.

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