COMMUNICATION

Optically enhanced semi-transparent organic solar cells through hybrid metal/nanoparticle/dielectric nanostructure

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Abstract
Semi-transparent organic solar cells (st-OSCs) that hold high recovery of efficiency with sunlight illumination from both electrodes are of great interest in different applications. While the issues for improving the recovery of efficiency from top-illumination are very limited studied, we propose a hybrid optical nanostructure metal/nanoparticle/dielectric (M/NP/D) to achieve high recovery of efficiency. The M/NP/D nanostructure consists of high index and low-loss nanoparticles (NPs) (here we use Si NPs scatter instead of metal NPs) and index matching material (here we use Tris(8-hydroxyquinolinate) aluminum-Alq3) on ultra-thin Ag film, i.e. Ag/Si NPs/Alq3 as the top hybrid electrode of st-OSCs. Our results show that the transmission of the electrode is improved complementarily in long (due to Si NPs) as well as short wavelength regions (due to Alq3 layer) with additional synergetic improvement (due to hybrid M/NP/D nanostructure). Subsequently, the enhanced average visible transmittance (AVT) up to 32% is achieved for the proposed st-OSCs. Simultaneously, compared to the optimized control st-OSC with the bare ultra-thin Ag electrode, the power conversion efficiency (PCE) for top-illumination case is improved by about 34%, with a recovery of efficiency up to 68%. Moreover, angular dependence of short circuit current (Jsc) of st-OSCs with the hybrid electrode can be also alleviated. Consequently, the proposed transparent hybrid electrode can contribute to the emerging semi-transparent optoelectronics.

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Semi-transparent organic solar cells (st-OSCs) not only inherit the unique properties of organic solar cells (OSCs), but also bring innovation to both anode and cathode to preserve high transparency [1-9]. The commercialization of the st-OSCs can extend the potential application of OSCs such as the solar power-generating window, foldable solar curtains [4,6,10]. Nevertheless, the performance of st-OSCs is still low compared to opaque OSCs (o-OSCs) due to single pass of the light through devices and thus reduction of light absorption [8,9,11].

The recovery of efficiency here is defined as the ratio between the efficiency of the semi-transparent solar cell from top/bottom-illumination to that of its opaque one. An ideal st-OSC is expected to achieve high recovery of efficiency with sunlight illumination from both electrodes. However, the severe imbalance of device efficiency can exist between top- and bottom-illumination cases [4,12-14]. Studies in addressing these issues are limited especially the relative low recovery of efficiency from top-illumination [9]. Thus it’s desirable to improve the device efficiency when sunlight illuminates from top transparent electrode and thus simultaneously achieves high recovered efficiency for both sides illumination for emerging applications.

Transparent ultra-thin metal film has been reported as a good candidate being top transparent electrode due to its easy processing, high conductivity and long skin depth [4,10,15-17]. However, the trade-off between conductivity and transparency is a critical issue for highly efficient st-OSCs [17,18]. For instance, the transmission of 10 nm Ag film is less than 70% in near ultraviolet region, especially, the transmission will decrease to below 50% after the wavelength of 660 nm [18,19]. Meanwhile, studies of the issue in reducing the reflectivity of transparent metal electrode of st-OSCs are limited [20,21]. Developing a low surface reflectivity of the transparent electrode is essential to achieve high efficiency st-OSCs.

The issue of the low transmittance of ultra-thin metal electrode can degrade the light incoupling into the active layer of st-OSCs, the antireflective coating (ARC) layer is widely used to reduce the interfacial reflection thus increase the transmittance of metal electrode by light wave interference through finely tuning the thickness of the ARC layer [22-25]. Nevertheless, the use of ARC layer on ultra-thin metal film that functions as top electrode is not well studied in st-OSCs. In addition, the improvement from mere adoption of planar ARC layer would be weakened under sunlight with an oblique incident angle, due to the deviation of wave destructive interference condition.

Another potential solution is the plasmonic effect of metal nanomaterials and nanostructures, which is emerging as an excellent tool to enhance light harvesting by its highly confined near field distribution [26-34]. For st-OSCs, the incorporation of metal nanomaterials into device would increase some degree of surface roughness, it would bring difficulties to form high quality and uniform ultra-thin metal film, leading to poor electrical properties and thus hindering to achieve high efficiency of st-OSCs [15,20,35]. Therefore, the incorporation of metal nanomaterials into st-OSCs can only be expected to be located out of the device, where the light incoupling improvement mainly come from the scattering effect of metal nanomaterials other than the highly localized near field [36]. However, the use of metal nanomaterials at top electrode side will introduce the intrinsic loss as light impinging and be detrimental to achieve high transparency of ultra-thin metal electrode [36,37].

Compared to metal nanomaterials, the dielectric nanomaterials conversely have no such metal loss, and are an excellent candidate to improve light incoupling by the scattering effect [38-40]. Previous literatures report the utilization of whispering-gallery mode from the highly close-packed silica (SiO2) sphere for improving light incoupling in amorphous silicon (Si) solar cells [41,42], while the size of the sphere should be on the magnitude of several hundred nanometers because of the relatively low refractive index. Besides, through employing the two-dimensional (2D) photonic pattern that was made by interference lithography and soft-imprint lithography, an ultra-low reflectance of Si wafer from the top electrode can also be achieved for Si solar cells [43,44]. However, the direct integration of photonic patterns on thin top electrode for st-OSCs through the lithography process may damage the ultra-thin metal electrode and the organic active layer. Consequently, it is of great importance to offer a simple and efficient structure by using dielectric materials to realize low reflectivity for ultra-thin metal electrode and enhance the light incoupling of the active layer for highly efficient st-OSCs.

Moreover, performances of the planar st-OSCs will degrade as the incident light tilt away from the normal following the Lambert’s cosine law, which indicates the incident light intensity would vary as the cosine of incident angle of light [40,45,46]. Therefore, an alleviated angular dependence of device performance on polarization and incident angle of light is highly desirable for the commercialization of st-OSCs.

Here, we proposed the metal/nanoparticle/dielectric (M/NP/D) nanostructure as transparent electrode, which consists of high index scatter of Si nanoparticles (Si NPs) together with index matching material (here we use Tris(8-hydroxyquinolinate) aluminum, Alq3) as a hybrid optical structure on top of ultra-thin Ag film for enhancing the light incoupling. The hybrid electrode (Ag/Si NPs/Alq3) can improve average visible transmittance (AVT, defined as an average of transmission in wavelength region 380-740 nm) to about 72% with an enhancement ratio of 24% compared to that of the bare Ag thin film electrode (AVT=58%). After utilizing the proposed M/NP/D nanostructure as top electrode, the AVT of 32% is achieved for the proposed st-OSCs. Simultaneously, the power conversion efficiency (PCE) of st-OSCs can be improved by about 34% compared to the optimized control device without any optical incoupling structures. Moreover, by simultaneously utilizing Si NPs hybridized with Alq3 layer, the transmission is improved complementarily in long (due to Si NPs) and short wavelength regions (due to Alq3 layer) with additional synergetic improvement (due to hybrid M/NP/D nanostructure), resulting in a broadband absorption enhancement that almost cover the whole visible spectrum. Compared to the opaque counterpart, a recovery of efficiency up to 68% and 87% can be attained in our proposed st-OSCs in top and bottom
illuminations cases, respectively. Meanwhile, the theoretical and experimental results show that the utilization of Si NPs cannot only promote active layer absorption at long wavelength region but also favor an alleviated angular dependence of device performance in top-illumination.

Experimental section

Device fabrication

The OSCs with the device structure of ITO/ZnO(22 nm)/poly[2,6,7,8,11,12,14,15,18,19-decabinyl[2,7-bis(2-bromothien-5-yl)pyrrolo[3,4-c]pyrrole-1,4,5,8-tetrayl]dibenzo[1,2-b:3,4-b']dithiophene-alt-5-diindolylmethylene]-6,11-dithieno[3,2-b:2',3'-d']thieno[3,4-c]pyrrole-1,4-dione](PBDTT-DPP):(6,6)-phenyl C71-butyric acid methyl ester (PC71BM) (80 nm)/MoO3(9 nm)/Ag electrode are fabricated. PBDTT-DPP is a commercially available low bandgap material. The ITO/glass substrate is pre-cleaned with detergent water, acetone, ethanol and UV-ozone treatment for each of 15 min. The sheet resistance of the ITO/glass is 15 $\Omega \square^{-1}$. The ZnO is synthesized with a diameter of 5 nm by using the method reported by Beek et al. [47]. The solution of 15 mg ml$^{-1}$ ZnO in n-butanol is spin-coated at 3000 rpm on the prepared ITO substrate with a thickness of 22 nm. The active layer is prepared by spin-coating the blended PBDTT-DPP:PC71BM (6 mg ml$^{-1}$: 12 mg ml$^{-1}$) in DCB solution of 120 °C at 2000 rpm. The thickness of active layer is about 80 nm. The hole transport layer of MoO3 with a thickness of 9 nm is fabricated by thermal evaporation of the powder at the rate of 0.1 nm s$^{-1}$. By controlling the thermal evaporation, Ag films with thickness of 100 nm and 10 nm are formed as electrodes of o-OSCs and st-OSCs, respectively. For the opaque counterpart solar cells, the Ag electrode of 100 nm is evaporated with a shadow mask of 0.06 cm$^2$. For the st-OSCs, a 10 nm layer of Ag electrode is firstly evaporated at the rate of 0.5 nm s$^{-1}$, followed by spin-coating the Si NPs dispersion at 3000 rpm for 40 s with the optimized concentration of 0.1 mg ml$^{-1}$ and evaporating 50 nm of Alq3 layer at the rate of 0.5 nm s$^{-1}$ on the top electrode. The Si NPs dispersions are prepared by dispersing 150 nm Si NPs (INTERNATIONAL LAB) into ethanol at different concentrations with two hours ultrasonic bath. The st-OSCs with electrode of Ag/Si NPs only and Ag/Alq3 only are also fabricated in the same process and the characteristics of device performance were summarized in Supporting information.

Device characterization and modeling

The current density-voltage ($J-V$) characteristics of OSCs are measured by using a Keithley 2635 source meter and an ABET AM 1.5G solar simulator with a light intensity of 100 mW cm$^{-2}$. The incident photon-to-current conversion efficiency (IPCE) of different OSCs are measured by a home-built system with a Newport Xenon lamp, an Acton monochromator, a pre-amplifier and a Stanford lock-in amplifier. The absorption coefficient of active layer was measured by Woollam spectroscopic ellipsometry. The transmission and angular dependence of OSCs were measured by a home-built

Figure 1  The transmission of the transparent Ag film with and without light incoupling structure. (a) Experimental measurement (on quartz) and (b) theoretical calculation, (c) the transmission enhancement over the bare transparent Ag film by adopting different light incoupling architectures. The legend of “SI NPs + Alq3 (Summation)” represents linear summation of the enhancement from Si NPs only and Alq3 only. “SI NPs/Alq3 (Hybrid)” represents the measured enhancement from the hybrid M/NP/D nanostructure and (d) the schematically depicted total internal reflection of the light path in hybrid transparent film with M/NP/D nanostructure.
system with a Newport Xenon lamp, a stepper motor turntable, an Oriel spectrometer integrated with CCD (charge-coupled device), and a Keithley 2635 source meter. The SEM images were captured by using LEO 1530 FEG Scanning Electron Microscope. The AFM images were captured by using NTegra Solaris from NT-MDT.

For the theoretical results, Finite difference time domain method was adopted to carry out the theoretical calculation of transparent metal film with and without high index Si NPs and Alq3 [48,49]. For model involved Si NPs, the results were averaged over period of 400, 500, 600 and 700 nm to smooth the influence originated from periodicity. The result of angular dependent scattering for single Si NP was calculated by Mie theory [36].

Result and discussion

Transmission properties of the hybrid electrode and st-OSCs

To obtain high transparent metal electrode, an investigation of the ultra-thin Ag film with and without light incoupling architecture has been carried out experimentally along with theoretical analysis. A 10 nm transparent Ag film was fabricated on quartz as a reference through thermal vacuum evaporation. The transmission of the ultra-thin Ag film was measured as shown in Figure 1a (black square line). A roll-off transmission can be observed from near ultraviolet to near-infrared region due to the high reflection of metal properties of Ag film.

Alq3 effects and Si NP effects

A thickness of 50 nm Alq3 film as anti-reflector was evaporated on the transparent Ag film to reduce the reflection. The transmission of Ag/Alq3 film is enhanced particularly in the blue region (around 450 nm-600 nm) as shown in Figure 1a. The AVT is improved from 58% to 68%. The pronounced peak around 460 nm in transmission spectrum is due to the destructive interference of incident and reflected wave [11,50,51]. The occurrence of a dip around 400 nm in transmission spectrum is ascribed to the Alq3 absorption and the existence of surface plasmon along the interface of Ag and Alq3 film that results in reducing light transmission.

Besides the considerable enhancement of light incoupling in short wavelength region achieved by the adoption of Alq3 film, it is highly desirable to further improve the transmission of ultra-thin Ag film in near-infrared region. From our theoretical studies, the Si NPs scatter can be used to improve the transmission in near-infrared region. As shown in Figure S1, through changing the size of Si NPs, the forward scattering can be well tuned. By using Si NPs with a size of 150 nm, a strong forward scattering peak at around 660 nm is theoretically obtained which can be explained by the excited magnetic dipole resonance of Mie resonant scatter [38,39] and is evidenced by the electric field distribution as shown in Figure S2a.

We therefore experimentally adopt Si NPs with a size of 150 nm to further enhance transmission of ultra-thin Ag film in near-infrared region. The scanning electron microscopy (SEM) of Si NPs dispersed on the ultra-thin Ag film is shown in Figure S3. From Figure 1a and c, a notable transmission improvement is experimentally demonstrated at near-infrared region for Ag/Si NPs film with the optimized concentration of 150 nm Si NPs (0.1 mg ml−1, shown in Figure S5). Consequently, due to total forward scattering effect of Si NPs at the wavelength of 660 nm (as shown in Figure S2b), the light incoupling of ultra-thin Ag film is improved in the near-infrared region.

Hybrid M/NP/D nanostructure effect

Interestingly, by using hybrid structure of Si NPs(150 nm)/Alq3(50 nm), we can obtain an additional synergetic improvement in transmission enhancement, which is more than the linear summation of the enhancement from Si NPs only and Alq3 only as described in previously. The transmission enhancement of Si NPs only (grey-circle line) and Alq3 only (olive-square line) are shown in Figure 1c, respectively, and the linear summation of the enhancement from Si NPs only and Alq3 only is denoted by dark-yellow star line. It can be observed in Figure 1c that the hybrid structure of Si NPs/Alq3 can offer an additional improvement (pink down-triangular line). This additional transmission enhancement introduced by Si NPs in hybrid electrode with M/NP/D nanostructure can be explained by light harvesting as shown in Figure 1d. Theoretically, the adoption of Alq3 as ARC layer ideally can lead to zero reflection as light is incident on Alq3 layer at wavelength 460 nm, therefore the reflection existing in the hybrid electrode mainly comes from the reflective metal surface as green line shown in Figure 1d (denoted by I). After introducing Si NPs in hybrid electrode Ag/Si NPs/Alq3, some amount of the reflected wave by Ag interface will enter the Si NPs (line denoted by II). The reflected wave will circulate in Si NPs and not leak out of Si NPs because of the existence of total internal reflection (TIR) caused by a relative large refractive index difference between silicon (n4=4.5) and Alq3 (n1=1.8). Without the hybrid M/NP/D nanostructure, the light circulation in Si NPs will not exist. It also cannot be possible for the light inside Si NPs to re-enter the OSCs through the Ag film for further improving the transmission and enhancing device performances. The theoretical transmission of the proposed hybrid M/NP/D electrode (Figure 1b) shows the same trend as experimental measurement (Figure 1a) which again confirms the efficiency of the hybrid transparent electrode with M/NP/D nanostructure. The effect of TIR by Si NPs will introduce an additional average transmission enhancement ratio of 18% for Ag/Si NPs/Alq3 hybrid electrode compared to linear summation of Si NPs only and Alq3 only electrode. Consequently, the improved AVT of 72% is achieved for the hybrid M/NP/D nanostructure due to the introduction of multiple effects including destructive wave interference, magnetic dipole resonance and TIR. These effects will enhance the light incoupling through the Ag transparent electrode into OSCs for improving the device performance.

The schematic device structure of st-OSCs with the hybrid transparent electrode is depicted as shown in Figure 2a. The transmission spectra of the st-OSCs with and without hybrid electrode are measured as shown in Figure 2b, the considerably enhanced device transmission is observed in the region 450-650 nm. The AVT of st-OSCs with hybrid electrode is 32% revealing an enhancement ratio of 61% compared to that of st-OSCs with bare ultra-thin Ag electrode. The device transmission of top illumination is enhanced around 400 nm which is mainly due to the increased surface roughness of the film [52,53]. Thus, the hybrid M/NP/D nanostructure can efficiently
enhance light incoupling and improve device transparency, and will contribute to enhance the transparency of emerging transparent optoelectronic devices.

Device performances of the hybrid electrode OSCs.

The o-OSCs and control st-OSCs are also fabricated and characterized. PCE of the optimized o-OSCs and st-OSCs are 7.19% and 3.61%, respectively (see Supporting information, Figures S6, S7 and Tables S1, S2 for the optimization of active layer thickness of OSCs). The hybrid electrode with structure of Ag/Si NPs/Alq3 is then adopted to improve light incoupling in st-OSCs. The corresponding current density–voltage ($J$–$V$) characteristics of the o-OSCs and st-OSCs with and without the proposed light incoupling nanostructure are shown in Figure 3a. By introducing the hybrid transparent electrode of Ag/Si NPs/Alq3, the short circuit current density ($J_{SC}$) of st-OSCs can be improved from 8.12 to 10.36 mA cm$^{-2}$. PCE of st-OSCs with the light incoupling structure is enhanced by about 34% (from PCE of 3.61%–4.83%). The summarized photovoltaic characteristics are also listed in Table 1. Importantly, the st-OSCs with light incoupling structure brought the recovery of device efficiency from 50% to 68% compared to that of its opaque counterpart. Moreover, it should be noted that, for bottom-illumination case (from ITO side), the PCE of st-OSCs with the hybrid electrode can reach to 6.22% corresponding to the recovery of efficiency up to 87% (Table 1), which is one of the highest reported value [4,5,9,10].

To further clarify the improvement brought by the hybrid electrode of M/NP/D nanostructure, the incident photon-to-current conversion efficiency (IPCE) is measured as shown in Figure 3b. A conspicuous broadband enhancement of IPCE can be observed from 430 nm to 800 nm in the main absorption spectrum region of the active layer. The IPCE enhancement in short wavelength is attributed to the enhanced light incoupling by Alq3 layer while the forward scattering of Si NPs mainly contributes to enhancement at long wavelength region as described by the theoretical results above. Consequently, the cooperative light incoupling effect of Si NPs/Alq3 hybrid structure ultimately induces a broadband enhancement of the IPCE and a 34% increase of the PCE.

In addition, the spatially distributed absorptive power of the active layer that average over $xoy$ plane is also obtained through theoretical simulation, which confirms the synergetic enhancement effect by the hybrid electrode as shown in Figure 4. The absorptive power of the active layer reveals the different enhancement regions in visible spectrum through employing light incoupling architecture of Si NPs only, Alq3 only and a hybrid of Si NPs/Alq3 as shown in Figure 4b-d, respectively. The theoretical results further confirm the broadband light incoupling effects induced by the proposed M/NP/D nanostructure, which are coincident with IPCE results.
Angular response of the hybrid electrode and st-OSCs

The angular dependence of incident light is another issue which should be considered in realistic cases. The transparent electrode with high transparency over a wide range of angle of incidence (AOI) for both p- and s-polarized light is greatly desirable for future photovoltaic applications. Remarkably, the hybrid electrode with the deposited Si NPs/Alq3 layer on transparent ultra-thin Ag film reveals properties of an ultra-low reflectivity over a wide angle of incidence. A theoretical investigation of the reflection of the hybrid electrode with M/NP/D nanostructure for both p- and s-polarization has been carried out. As shown in Figure 5a and b, the reflection is below 20% over a wide range of angle of incidence for both s- and p-polarization light at 660 nm (magnetic dipole resonance of Si electrode with high transparency over a wide range of angle of incidence (AOI) for both p- and s-polarized light is greatly desirable for future photovoltaic applications. Remarkably, the hybrid electrode with the deposited Si NPs/Alq3 layer on transparent ultra-thin Ag film reveals properties of an ultra-low reflectivity over a wide angle of incidence. A theoretical investigation of the reflection of the hybrid electrode with M/NP/D nanostructure for both p- and s-polarization has been carried out. As shown in Figure 5a and b, the reflection is below 20% over a wide range of angle of incidence for both s- and p-polarization light at 660 nm (magnetic dipole resonance of Si

![Figure 4](image_url)

Figure 4 Spatially distributed absorptive power of the device with semi-transparent hybrid electrode of (a) Ag, (b)Ag/Si NPs, (c) Ag/Alq3 and (d)Ag/Si NPs/Alq3. The white dot line regions denote the active layer of st-OSCs. The absorptive power is averaged over the xoy plane that represents the position of each layer.

Table 1 The summary of photovoltaic characteristics of OSCs with different electrodes.

<table>
<thead>
<tr>
<th>Top electrode</th>
<th>Voc (V)</th>
<th>Jsc (mA cm^-2)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
<th>Efficiency recovery</th>
</tr>
</thead>
<tbody>
<tr>
<td>o-OSC w Ag-100 nm</td>
<td>0.73</td>
<td>15.70</td>
<td>63.15</td>
<td>7.19</td>
<td>-</td>
</tr>
<tr>
<td>st-OSC w Ag-10 nm^a</td>
<td>0.72</td>
<td>8.12</td>
<td>61.63</td>
<td>3.61</td>
<td>50%</td>
</tr>
<tr>
<td>st-OSC w hybrid electrode^a</td>
<td>0.73</td>
<td>10.36</td>
<td>63.86</td>
<td>4.83</td>
<td>68%</td>
</tr>
<tr>
<td>st-OSC w Ag-10 nm^b</td>
<td>0.72</td>
<td>14.27</td>
<td>58.34</td>
<td>6.04</td>
<td>84%</td>
</tr>
<tr>
<td>st-OSC w hybrid electrode^b</td>
<td>0.72</td>
<td>14.64</td>
<td>58.62</td>
<td>6.22</td>
<td>87%</td>
</tr>
</tbody>
</table>

^a st-OSCs were illuminated from the transparent electrode side.

^b st-OSCs were illuminated from ITO side.
NPs), which offers an improved light incoupling compared to bare transparent ultra-thin Ag electrode.

The characteristics of device performance varied by the incident angle of illumination light are also investigated for the st-OSCs with and without the hybrid electrode structure. Notably, the incorporation of Si NPs/Alq3 hybrid structure on ultra-thin Ag layer can reduce light reflection over a wide incident angle as shown in theoretical results of Figure 5a and b as well as experimental results of Figure 5c and d. The normalized $J_{SC}$ and PCE in the case of the control semi-transparent electrode without any light incoupling structures reveal a clear obedience to Lambert’s cosine law. The reason for the angular alleviated dependence in the case of hybrid electrode is mainly attributed to light incoupling effect from Si NPs in the hybrid electrode as shown in Figure S9, which indicates the increase of the angular absorptive power of the active layer. It should be noted that the normalized photovoltaic characteristics of $V_{OC}$ and FF showed almost no dependence on angle of incidence (as expected) in the case of semi-transparent electrode with or without light incoupling M/NP/D nanostructure. Consequently, the angular dependence of $J_{SC}$ and PCE is improved by introducing the light incoupling structure.

**Conclusion**

In conclusion, we have proposed a novel hybrid electrode based on ultra-thin Ag film atop by integrating with Si NPs and Alq3. By using the hybrid electrode with M/NP/D nanostructure, the transmission of the electrode is improved complementary in long (due to Si NPs) as well as short wavelength regions (due to Alq3 layer) with additional synergetic improvement (due to the hybrid M/NP/D nanostructure). The overall transparency of the st-OSCs is improved by 61%. Meantime, st-OSCs with hybrid electrode achieved PCE enhancement of about 34% compared to the optimized st-OSCs without light incoupling structure. In addition, the recovery of efficiency of 68% and 87% are demonstrated in top and bottom illuminated devices, respectively. Notably, the large enhancement of light incoupling as well as the alleviated angular dependence of device performance are also achieved. Consequently, the hybrid transparent electrode with M/NP/D nanostructure can be beneficial to the future transparent photovoltaic applications.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.nanoen.2015.08.014.

References


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