

Improved Operational Stability of Polymer Light-Emitting Diodes Based on Silver Nanowire Electrode Through Pre-Bias Conditioning Treatment

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For the first time, highly efficient and flexible polymer light emitting diodes (PLEDs) based on silver nanowire (AgNW) electrode, with improved operational stability by simply applying pre-bias conditioning treatment, are demonstrated. Reverse bias conditioning performed before J - V - L measurement of the PLEDs enables the rough AgNW networks to function properly as a bottom electrode by stabilizing current characteristics, and the devices continue to show consistent operational performances. Conditions of applied bias and thicknesses of active layer are controlled for optimization and it is found that high reverse voltage is required to obtain current stabilization. Adequate thickness of polymer is also necessary to avoid breakdown induced by reverse bias. The essential effect of pre-bias conditioning on the improved performances of PLEDs is investigated, and it is found that morphological change of AgNW networks contribute to the improvement in device performance. Some of the AgNWs that appear to be pathway of leakage current are deformed, and surface roughness (RMS) of the AgNW film is decreased while the sheet resistance of the film is maintained when the reverse bias conditioning is applied. It is also revealed that pre-bias conditioning is independent from directionality of the applied bias when utilizing insulating polymer sandwiched between two electrodes.

used transparent electrodes are indium tin oxides (ITO) which have remarkably low sheet resistance ($\approx 10 \Omega \text{ sq}^{-1}$) and high transparency ($> 85\%$). However, many efforts of finding new alternatives for ITOs have been actively tried due to their problems containing mechanical brittleness and the scarcity of rare-earth (indium) resources.^[4–6]

To replace ITO electrode, various transparent electrodes have been emerged such as carbon nanotubes (CNTs), graphene, and networks of metallic nanowires.^[7–15] Among them, silver nanowire (AgNW) electrodes take center stage for its low sheet resistance and high transmittance, which are very close to the ITOs.^[12–15] However, AgNW networks formed on flexible substrates directly from a dispersive solution are restricted in their application, especially for organic thin-film devices, due to their inherent surface roughness which leads to short-circuit across the thin-film devices. The rough morphology is attributed to the intrinsic three-dimensional shape of

silver nanowire itself which leads to height variation of the films more than 300 nm in stacked structures.^[16–19] In a PLED having AgNW film as a bottom electrode, a high level leakage current that flows across the device takes place due to the features of AgNWs protruding into a thin polymer layer places decreasing operational stability. Here, the leakage current is denoted as current that flows without emission of light before the device is turned on. Thus, most of the researches that utilize a pristine AgNWs film which refers to AgNWs randomly

1. Introduction

With increased demand for next generation display, organic materials-based display, especially, polymer light emitting diodes (PLEDs) have attracted the spotlight due to their functionality and practicality; ultra-thin and light-weight device architecture and solution-processability in large area that brings cost reduction.^[1–3] Transparent electrodes have been essential part of the PLEDs as pathways of the emitted light. The most widely

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dispersed on the substrates without any additional treatment to flatten the morphological features have been limited to organic photovoltaic cells where thick active layer (>200 nm) is necessary for the enlarged light absorption.^[20–23] Zeng et al. demonstrated a method of reducing the surface roughness of the AgNW networks by burying them in the transparent polymer matrix, and Yu et al. reported AgNW electrode having a smooth surface obtained by utilizing transparent and cross-linkable polymer.^[16–19] However, the previous works required cumbersome steps for fabrications of organic thin-film devices based on the AgNW film.

Here, we demonstrate highly efficient AgNW-based PLEDs with improved operational stability realized by conducting pre-bias conditioning treatment on the devices. Fluctuating features of leakage current become stabilized and the device performance is improved in the resulted PLEDs after running the devices once in reverse bias (0 to -20 V). Such stabilized current characteristics are similar to those of ITO-based devices, even if the roughness (RMS) of the AgNW film used here is 18 nm in average which is too large to be compatible with the PLED where the optimized thickness of active layer is usually less than 100 nm. Unlike PLEDs having ITO electrode where electrical charges are uniformly injected through the interface between polymer and electrode, electrical charges are irregularly injected from the rough surfaces of randomly dispersed AgNWs in the PLEDs having AgNW electrode, and they are irregularly recombined at arbitrary points inside active layer, leading to low efficiency. By implementing pre-bias conditioning on the AgNW-based PLEDs, the device efficiency increased as leakage current was stabilized, and the device continue to show consistently stable operational characteristics. We investigated the reason behind current stabilization induced by pre-bias conditioning, revealing that some protruding features of AgNWs that cause leakage current were melted out by high reverse bias. We further investigated to check whether the same effect should be on with a high forward bias or not. However, it was not easy to observe that effect in PLEDs, since forward bias over 10 V leaded to failure of the devices due to the property of PLEDs in which a high level of current flowed in a forward direction. By utilizing insulating sandwiched between top and bottom electrodes, instead of using organic semiconducting materials, we were able to check whether directionality of applied bias showed up in stabilization of current of the device or not.

2. Results and Discussion

J - V - L characteristics of AgNW-based PLEDs were obtained after running the devices once in either forward bias or reverse bias where an electric field is applied across the vertically stacked layers. In forward bias scan, pre-bias ranged from 0 to 6 V was applied, this type of pre-bias conditioning was conducted for reliable operation of the device. Maximum applying voltage (6 V) in pre-bias conditioning was determined by observing that high scanning voltages more than 8 V in this case resulted in severe degradation of the device. In reverse bias conditioning, the device was scanned from 0 to

-20 V. J - V - L and efficiency curves obtained after an application of either forward or reverse bias are presented in Figure 1a,b. In the case of forward bias conditioning, a high level of leakage current over 1 mA cm^{-2} already existed below turn-on voltage at which the device starts to emit light (2.4 V), which were not desirable for PLEDs since they brought the device unstable and consequently resulted in lowered efficiencies. The high level of leakage current is resulted from the rough surface characteristics of the AgNWs being protruding into polymer layer. Surface morphology of AgNW film is much coarser than that of ITO, and the roughness (RMS) of AgNW film ($60 \text{ } \Omega \text{ sq}^{-1}$) is 18 nm, which is considerably larger compared to surface roughness of ITO (1 nm), (Supporting Information, Figure S1). With reverse bias conditioning treatment, interestingly, the leakage currents were reduced more than 100 times in the low voltage region (below 2 V), and the decrease of leakage current was accompanied by improved power efficiency, 6.2 lm W^{-1} at 3.5 V while comparing 3.9 lm W^{-1} at 7.5 V for forward bias conditioning (Figure 1b). Such an improvement in power efficiency was made by stabilizing the leakage currents which flow across the device but result in no emission. The obtained stabilized current characteristics and improvement in efficiencies after reverse bias conditioning were not observed in ITO based PLEDs with the same structural configuration (Supporting Information, Figure S2). It is notable that overall thickness of the polymer layers containing PEDOT:PSS (50 nm) and SY-PPV (80 nm) used is just 130 nm which is the typical thickness in the polymer based light-emitting diodes (We do not increase polymer layer thickness intentionally in order to avoid leakage currents). We confirmed that a 250 nm of polymer layers containing 50 nm of PEDOT:PSS and 200 nm of SY-PPV was required in AgNW based devices to obtain stabilized current characteristics without reverse bias conditioning. However, thicker polymer layer is undesirable due to severely lower brightness and efficiency which are resulted from the lowered electric field across the devices (Supporting Information, Figure S3). Figure 1c shows the result of repetitive J - V - L measurement of the PLEDs with the reverse bias treatment. Stable J - V - L curves were obtained consistently (upto 5 times), which implied there was a permanent change in the morphological characteristics of the AgNW film when the reversed bias conditioning was applied.

Figure 2a shows J - V - L curves of AgNW-based PLEDs with different pre-bias conditionings. To compare the amount of leakage currents which flow across the devices before turn-on voltage (2.4 V), we arbitrary plot current density at 1 V of each case on Figure 2c. As a range of applied voltages became larger, current density at 1 V became lowered. Until the range became from 0 to -5 V upto 0 to -15 V, the corresponding devices still showed unstable characteristics of the current. Voltages over -15 V were required to reach the level that leakage currents were sufficiently suppressed, below the level of $10^{-3} \text{ mA cm}^{-2}$ at 1 V. A result of the repetitive pre-bias conditioning with 0 to -5 V upto 10 times is shown in Figure 2b indicating still the high level of leakage current exists. See that there was still 100 times higher current density at 1 V compared to the case of which high reverse bias conditioning (0 to -20 V) is applied, and no sooner had we apply voltage to the PLEDs than the

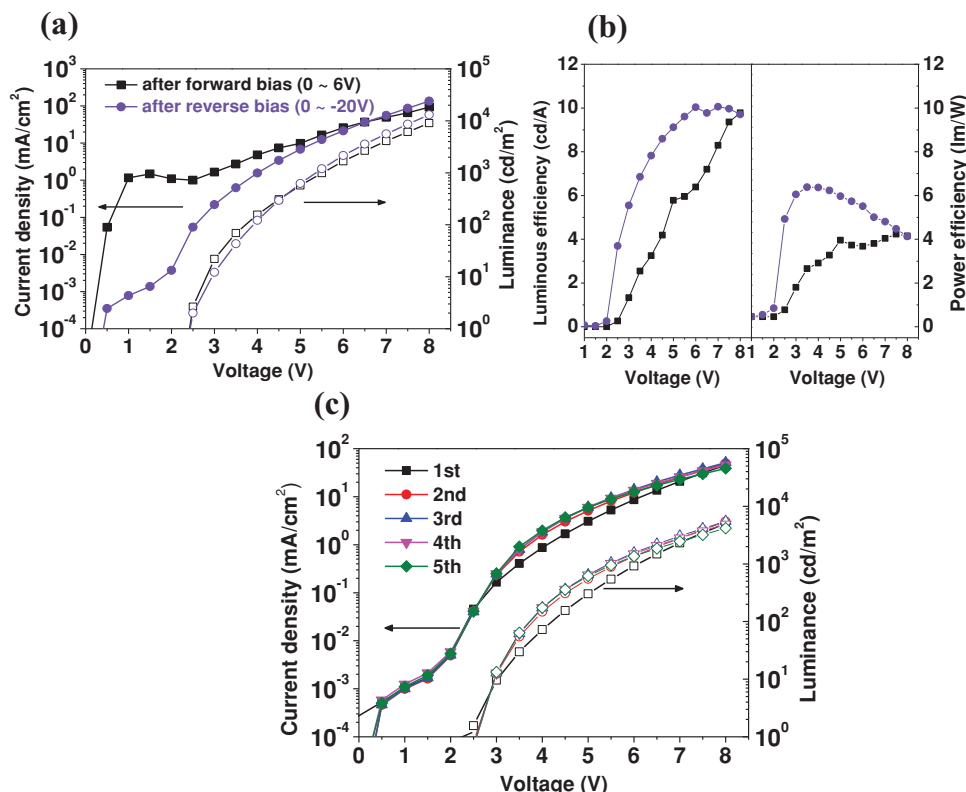


Figure 1. a) J - V - L characteristics and b) luminous efficiency (cd A^{-1}) and power efficiency (lm W^{-1}) for PLEDs as a function of an applied voltage after the following; pre-bias conditioning in forward bias (square) and reverse bias (circle). In forward bias conditioning treatment, the voltage applied to AgNW anode is higher than that to aluminum cathode (0 to 6 V). In reverse bias conditioning treatment, the voltage applied to aluminum cathode is higher than that to AgNW anode ranged from 0 to -20 V. c) Repetitive measurements of J - V - L characteristics after reverse bias voltages (0 to -20 V) were applied to the devices.

device started to show linear increase in current density without any suppression before turn-on voltage. We observed that there was limitation to obtain stabilized current characteristics, if low voltage pre-bias conditioning was used, no matter how repetitively we implemented pre-bias conditioning on PLEDs with low voltages.

We varied thickness of active layer from 30 nm to 120 nm while keeping pre-bias conditioning as 0 to -20 V. J - V - L and efficiency curves of the devices are shown in **Figure 3a,b**, respectively. Both current density and luminance increased as active layer thickness decreased from 120 nm to 60 nm due to the increased electric field. As active layer thickness became 30 nm thick, however, no luminance was observed even though current still flowed through the device. The device with 30 nm of active layer was totally degraded by reverse bias conditioning. As polymer layers sandwiched between top and bottom electrodes become thinner, higher electric field is developed through the device at a constant voltage. This higher electric field eventually results in the breakdown which refers to rapid increase of current and leads to failure of the devices.^[24] To check whether breakdown is taking place in the PLEDs, current flowing through the devices with different active layer thickness was measured during reverse bias conditioning (Supporting Information, Figure S4a). A low-level of reverse currents was detected in the PLED having 80 nm of active layer and that was desirable characteristic in the diodes in which little

currents were flowing in reverse region. In the case of a PLED with 30 nm of active layer, however, current density dramatically increased as applied voltage became higher, and the device went as far as to emit light during reverse bias conditioning because considerable amounts of holes and electrons were injected even in reverse bias regime due to the increased electric field (Supporting Information, Figure S4b). Fortunately, the maximum reverse voltage (-20 V) to be applied to AgNW-based PLEDs that have active layer thickness of 80 nm seems to be within the range that do not cause breakdown of the devices, because little currents are flowing even at -30 V, and a high level of reverse current comparable to the counterparts in a forward bias region begins to flow when reverse voltage become higher than -35 V (Supporting Information, Figure S4c). It is notable that proper thickness is required to prevent deterioration of the PLEDs when applying reverse bias conditioning. Active layer thickness of 60–80 nm would be suitable for PLEDs with this configurational device structure, considering that maximum current efficiencies and power efficiencies of the devices were 10.1 cd A^{-1} at 4.5 V, 8.3 lm W^{-1} at 3.5 V for 60 nm and 10.3 cd A^{-1} at 6 V, 7.0 lm W^{-1} at 3.5 V for 80 nm of active layer, respectively, and those values were comparable to those of ITO-based devices reaching 10.4 cd A^{-1} at 4.4 V, 8.9 lm W^{-1} at 3.2 V. However, the range of thickness of polymer layers that pre-bias conditioning is markedly working can be varied, depending upon the device architectures, materials, and the sheet resistance of AgNW film.

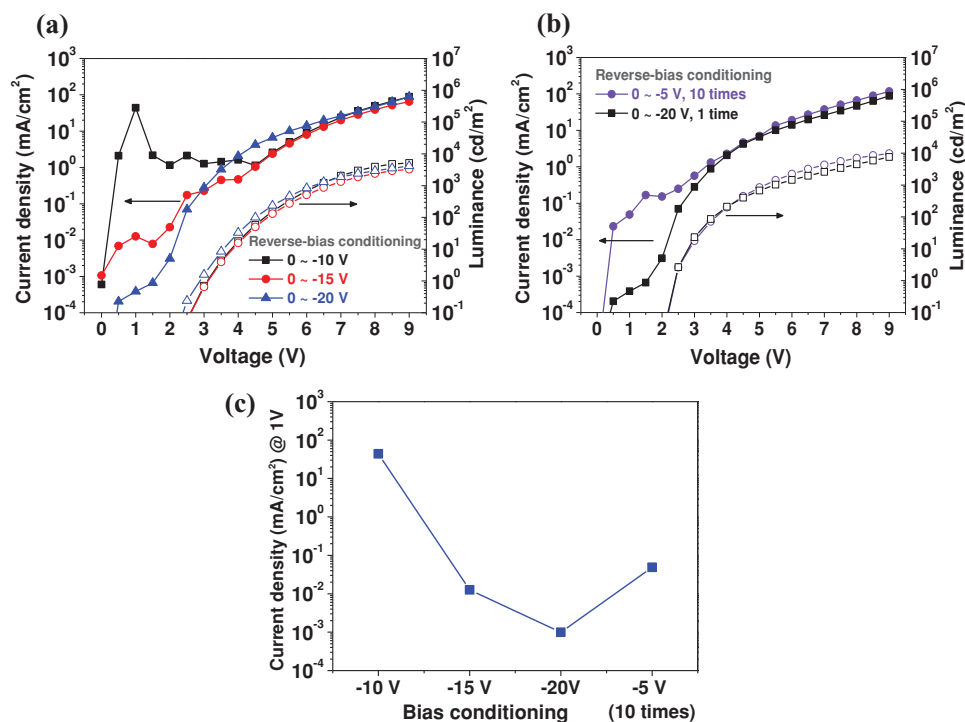


Figure 2. *J*-*V*-*L* characteristics of PLEDs a) with different reverse bias conditioning (square: 0 to -10 V, circle: 0 to -15 V, and up-triangle: 0 to -20 V), b) after repetitive bias conditioning with low voltages (0 to -5 V) up to 10 times. Results for the case where voltage range from 0 to -20 V is applied were also shown for reference. Active layer thickness for each case was 80 nm. c) Current density for PLEDs with various bias conditionings measured at 1 V.

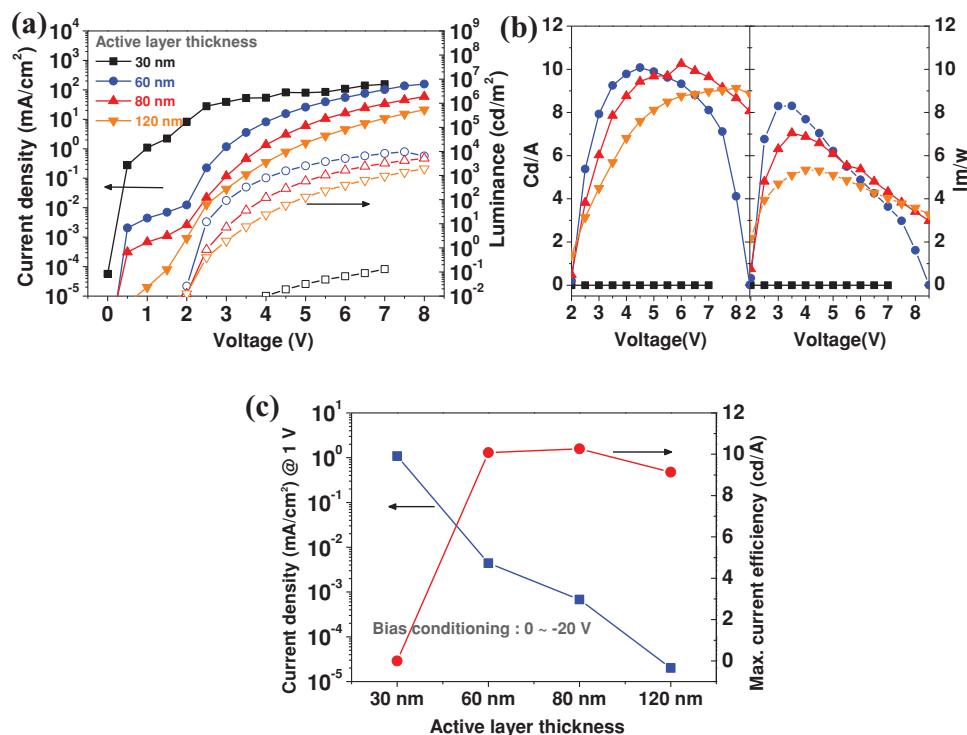


Figure 3. a) *J*-*V*-*L* and b) efficiency curves with different thickness of active layer, AgNW/PEDOT:PSS(50 nm)/SY-PPV(given above)/LiF(1 nm)/Al(120 nm), equivalent reverse bias conditioning (0 to -20 V) is applied to each sample. c) Current density measured at 1 V and maximum current efficiency of the PLEDs with various active layer thickness at fixed bias conditioning (0 to -20 V).

To investigate essential effect of reverse bias conditioning on AgNW film morphology, upper layers were peeled off from the fabricated PLEDs. The detailed process is described in the experimental section. Distilled water was used to dissolve hydrophilic PEDOT:PSS polymer, and consequentially its upper layers containing SY-PPV, LiF and Al cathode were come away into the water. To confirm the peel-off process does not damage AgNW networks, we measured electrical conductivity of the AgNW film and found out that there was no increase in sheet resistance of the film after the peel-off process (Supporting Information, Figure S5a). Measurement of sheet resistance of bare AgNW film in distilled water under ultra-sonication indicates that AgNW networks remain intact under the harsh condition until 5 min (Supporting Information, Figure S5b). Note that the peel-off process was completed within 3 min under relatively mild condition (without ultra-sonication).

Figure 4a displays SEM image of bare AgNW/PET without any bias application. AgNWs randomly dispersed in the PET substrate meet one another and pile up at some points. These points are regarded as mountains that induce current leakage by protruding into the polymer layers in the PLEDs. SEM images of the AgNW film on PET substrate which was used as an electrode for fabrication of PLED and on which reverse bias treatment is applied (0 to −20 V) and the peel-off process is conducted sequentially are shown in Figure 4b,c. Some deformed features of nanowires that looked like darker shades were observed in many places, and the AFM topographic image

and line profile shown in Figure 4d reveal that the shades are melted parts of the nanowires. The features melted by applied bias might include nanowires that might be directly in contact with the Al electrode or even impurity particles that might be unintentionally added during the fabrication of the devices and could be a pathway to leakage current, even though it is difficult to be observed. Such observed morphological change of AgNWs is correlated with welding of metal nanowires by Joule heating or thermal treatment.^[26,27] Sheet resistance of AgNW electrode is decreased as the junctions of nanowires are welded together at a high annealing temperature. However, continued thermal annealing process eventually leads to disconnection of Ag nanowires with increased sheet resistance of the electrode. In the light of disconnection of AgNW by heat, the melted parts of the Ag nanowires is considered to be formed by Joule heating under the high electric field.^[14] The morphological deformation of the AgNWs was observed especially at the junctions of nanowires. Typically, junctions of nanowires refer to the points that are piled up together and closest to the Al cathode, so that current is easily flowing through the junctions by small amount of applied voltages, causing leakage currents through the PLED structures. Meanwhile, Joule heating takes place through AgNW networks as voltages are applied, especially at the junctions of nanowires that have the highest electrical resistance in the AgNW networks,^[13] since

$$P = IV = I^2 R$$

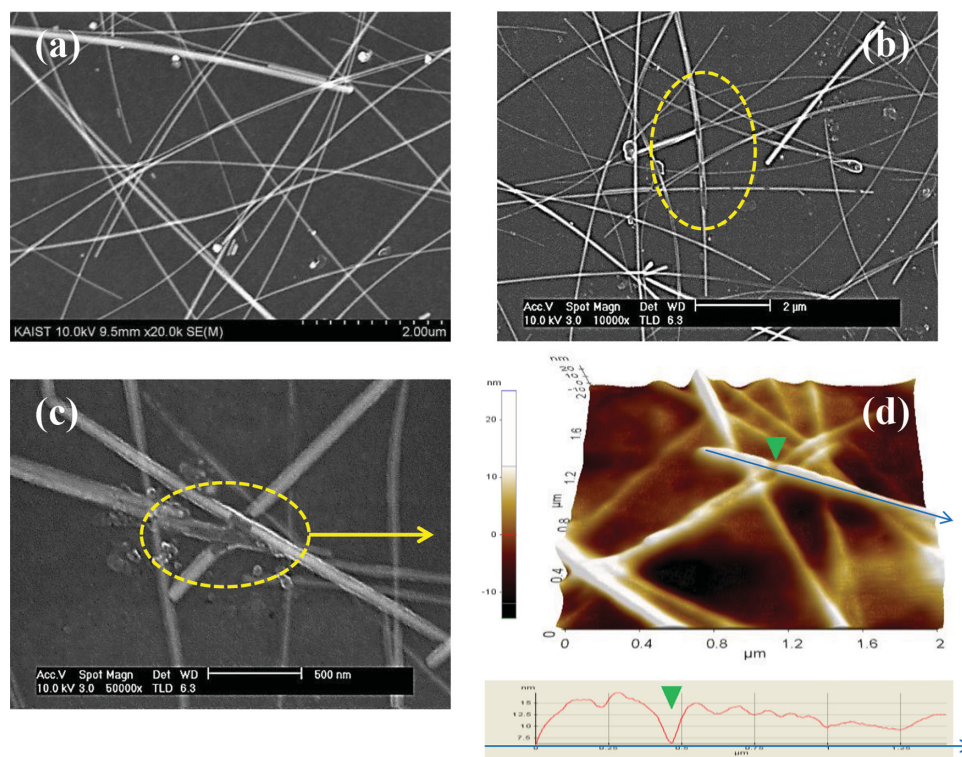


Figure 4. SEM images of a) as prepared AgNW/PET film, AgNW/PET film after reverse bias treatment is applied on the fabricated PLED, and upper layers peeled off in b) low magnification and c) high magnification. Note that there are some deformed features especially in the intersections where nanowires are crossing one another, it is different from nanowires shown in image (a). d) AFM topographic image and line profile of the deformed features of a nanowire.

where R is the resistance.^[15,25] Judging from that point, we can speculate the application of high reverse bias made some parts of the Ag nanowires melted by generating intensive heat especially on the junctions, eliminating the features of AgNWs that induced leakage current across the devices. Surface roughness (RMS) measured for the pristine AgNW/PET film and AgNW film where reverse bias treatment (0 to -20 V) was applied is shown in Figure S6, and it was found out that RMS value was decreased from 18 nm to 7 nm after reverse bias treatment, and we confirmed that reverse bias conditioning made some features of AgNWs related to the leakage currents be melted away and sequentially led to surface flattening of the AgNW electrode that was regarded as a crucial factor for the stable operation of the AgNW-based PLEDs.^[17] Sheet resistance of the AgNW electrode in the PLEDs after reverse bias conditioning treatment (0 to -20 V) was measured after peeling off the upper layers with DI-water, since some melted parts in Ag nanowires especially at the junctions and even disconnected AgNWs were observed and expected to increase the sheet resistance of the electrode. Interestingly, sheet resistance did not increase but was maintained regardless of the morphological changes of the AgNWs networks (Supporting Information, Figure S7). It is considered that some of the partially melted Ag nanowires do not act as setbacks in electrical connections of AgNW networks, and strongly welded junctions of Ag nanowires by electric field-induced Joule heating rather make a positive contribution to the electrical property of the film.

We applied a voltage of -15 V across two ends of the flexible AgNW/PET film itself which do not have any stacking layers for device operation to check an equivalent morphological change of the AgNW film. The results obtained were different from those of the AgNW film prepared by pre-bias conditioning treatment mentioned above. Temperature of the film gradually increased due to Joule heating induced by current flowing, after a few seconds, however, current flowing through the AgNW film is stopped and do not flow anymore. SEM images of the resulted AgNW/PET film indicate that AgNWs were disconnected in the middle of the film accompanied by deformation of PET film that made the disconnection of the current flow (Supporting Information, Figure S8). A lot of bundles of Ag nanowires near the deformed PET were melted and even burnt away. Extremely high level of current flowing through the highly conductive metal nanowires across the two ends of the AgNW film which is originated from an absence of electrical resistance that regulates the amount of electrical current generated the large amount of heat throughout the film and induced the unexpected cut-off of the electrical flows at uncontrollable points that could even deform the PET film. Even when we decreased the applied voltage to -5 V, electrical current flowing through the AgNW film is also eventually stopped in the same manner due to the high level of electrical flows. These undesirable consequences were avoided and improved surface characteristics of the AgNW networks for application to PLEDs was controllably realized by application of reverse bias conditioning to the PLED structures in which organic materials that function as electrical resistances were vertically stacked that the restricted electrical currents were flowing even at high reverse bias.

Provided that a high reverse bias made some features of the AgNW networks melted by generating heat, the same effects

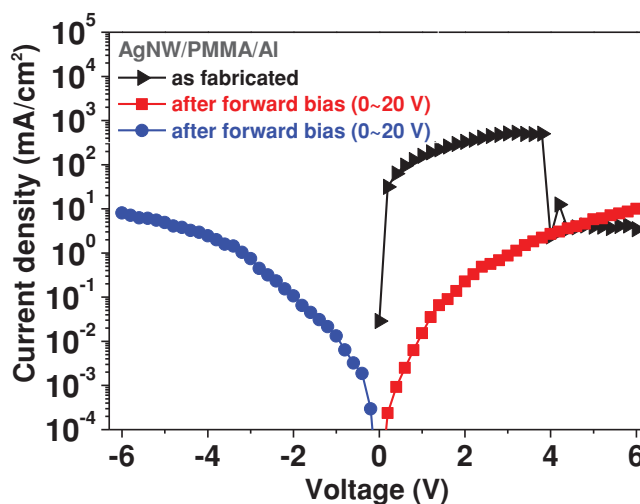


Figure 5. J - V measurement of AgNW/PMMA (100 nm)/Al (120 nm) after forward bias conditioning (0 to 20 V), a filled triangle denotes J - V curve of as-fabricated device without any bias conditioning treatment.

should be on by a high forward bias and not be dependent on directionality of an applied bias. However, it was not easy to observe that effect in PLEDs, since a forward bias over 10 V led to failure of the devices due to the high level of current flowing in a forward direction. To check whether current stabilization is not dependent on directionality of applied bias and not limited in the diode structures, we fabricated a device that has insulating polymer rather than organic semiconducting materials which bring about biased currents that are mostly flowing in one direction. A well known insulating polymer, poly methyl methacrylate (PMMA) was used in, to obtain AgNW/PMMA/Al structure. We ascertained that the high level of leakage current flows in the as-fabricated AgNW/PMMA/Al structure due to rough morphology of AgNW networks, unless we implemented bias treatment. However, the high level of leakage current was stabilized by implementing forward bias conditioning (0 to 20 V) to the devices (Figure 5). Current density at 1 V decreased by more than 1000 times after forward bias conditioning was applied on the device. Considering that change in the PLEDs after pre-bias conditioning was that some features of metal nanowires were melted, current stabilization in forward bias is a matter of course. In the current-voltage curves shown in Figure 5, it is worthy of notice that current densities are symmetric in both forward and reverse region in AgNW/PMMA/Al structure, and this is dissimilar to those observed in the PLEDs structures of which current asymmetrically flows biased in forward direction. A high level of leakage current is also stabilized by reverse bias conditioning in the AgNW/PMMA/Al structure (Supporting Information, Figure S9). We confirmed that stabilization of currents was not limited to the diode structures but was also observed in the AgNW/PMMA/Al structure by both forward and reverse bias conditionings, since how much currents were flowing from one metal (AgNW) to another metal (Al) was not much influenced by the directions of applied bias. From the obtained results, we can conclude that the high level of leakage current is not related to organic interlayers but caused by metallic nanowire electrode itself, and stabilization of

current by pre-bias conditioning is made by eliminating some features of AgNWs that induce leakage currents and is not dependent on a direction of applied bias.

3. Conclusions

In summary, we demonstrate efficient PLEDs based on flexible AgNW electrode by simply applying pre-bias conditioning. This is the first report to apply rough AgNW networks as a bottom electrode into the PLEDs without intentionally increasing thickness of polymer layer in order to avoid leakage currents. This simple method do not require any additional surface treatment on the silver nanowire networks. The high level of leakage current which comes from the rough surface characteristics of nanowire networks was stabilized and the device efficiencies of the PLEDs were improved after running the device once in 0 to -20 V, and the device continued to show stable operation consistently. We found out that considerably high reverse bias (larger than -15 V) was required to observe clear-cut effect of stabilization of current, and there was limitation to obtain stabilized current characteristics, no matter how repetitively we implemented pre-bias conditioning with low voltages (0 to -5 V). By varying thickness of active layer of PLEDs, it was revealed that sufficiently thick active layer was necessary to avoid deterioration of the device during reverse bias conditioning, and 60–80 nm of active layer was found to be appropriate for stable operation in this device architecture, showing the highest device efficiencies. As work to examine the change of AgNW film morphology after reverse bias conditioning, we peeled off upper layers by immersing the device in distilled water and discovered that some parts of nanowires, which were suspected to cause leakage current, were melted by applying reverse bias. We also found out that the surface roughness (RMS) of AgNW networks was significantly decreased after reverse bias treatment and we speculate the flattened morphology of AgNW networks contributed to the improved operational stability of the PLEDs. An equivalent effect of forward bias treatment on the operational stability of the PLEDs was not able to be verified in the PLED structures in which organic semiconducting materials are stacked between two electrodes, due to the severe degradation of the devices being operated at high voltages by the extremely high current density in a forward direction. By utilizing insulating polymer, we confirmed that stabilization of current was made by both forward and reverse bias conditionings and was not dependent on a direction of applied bias.

4. Experimental Section

Fabrication of AgNW-based PLEDs: AgNW electrodes formed on flexible PET substrates were prepared in accordance with the previous procedures and was used without any further cleaning.^[15] PEDOT: PSS (Clevios P VP Al4083) dispersed in water was spin-cast at 1500 rpm for 60 s to form thin layer of 50 nm on top of the AgNW anode of which sheet resistance was $60 \Omega \text{ sq}^{-1}$, and heated at 130°C for 30 min to remove residual water. A light-emitting material, SY-PPV (PDY132, Merck) dissolved in toluene (anhydrous, 99.8%, Sigma Aldrich) with different concentration (3 mg mL^{-1} to 8 mg mL^{-1}) was spun onto the PEDOT layer followed by thermal annealing at 100°C for 1 h in a

nitrogen-filled glove box. Thickness of the SY-PPV layer was varied by controlling concentration of solution at a fixed spin speed (3000 rpm). To measure thickness of polymer layers used in PLED with AgNW electrode, polymer solutions were spun on ITO/glass substrate under the same condition. Thicknesses of polymers on ITO/glass substrate were measured by surface profiler (Tencor α -step 500). Finally, LiF (1 nm) and Al cathode (120 nm) were thermally evaporated under high vacuum condition (1×10^{-7} Torr). For fabrication of AgNW/PMMA/Al device structure, PMMA (Sigma Aldrich) dissolved in toluene (30 mg mL^{-1}) was spun onto the AgNW film to be 100 nm and followed by Al deposition.

Pre-Bias Conditioning Treatment and Device Characterization: J - V - L characteristics were measured by a Keithley 2635A source meter unit and a Minolta CS2000 spectro-photometer in a dark room. Before J - V - L measurement, two types of pre-bias conditionings were accompanied in the devices; forward bias conditioning and reverse bias conditioning. In forward bias conditioning, an electrical potential of the AgNW anode was higher than that of the Al cathode and the range of scanning voltage was 0 to 6 V. On the other hands, higher voltages ranged from 0 to -20 V where an electrical potential of the AgNW anode was lower than that of the Al cathode were applied in the case of reverse bias conditioning. An interval of the applied voltage was 0.2 V for both cases. Non-contact mode AFM (NC-AFM) was conducted to evaluate the morphological property of the AgNW film. SEM images were obtained by using Hitachi S-4800 scanning electron microscope and Philips XL30 scanning electron microscope. For an observation of AgNW morphology on PET film after PLED fabrication, the PLED was immersed in a beaker containing distilled water. The layers containing PEDOT: PSS, SY-PPV, and LiF/Al cathode were peeled off easily, because the underlying PEDOT: PSS layer is easily dissolved in water. The peel-off process was completed within 3 min. Sheet resistance of AgNW film was measured by using four-point probe instrument (CMT SR1000N, AIT).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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