Improvement of amorphous-carbon active-layer thin-film light-emitting diodes using room-temperature ultrasound treatment

Woo Yeong Cho\textsuperscript{a)} and Koeng Su Lim
Department of Electrical Engineering, Korea Advanced Institute of Science and Technology, 373-1 Kasong-dong, Yusong-gu, Taejon 305-701, Korea
S. Ostapenko
Center for Microelectronics Research, University of South Florida, Tampa, Florida 33620

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Ultrasound treatment (UST) applied at room temperature enhances electroluminescent intensity (maximum at 600 nm) and optical output in thin-film light-emitting diodes with hydrogenated amorphous-carbon as an active layer. This positive UST effect is attributed to a reduction of the diode series resistance caused by a change of the interface and contact resistances. The UST effect is saturated with increase of the ultrasound amplitude. © 1999 American Institute of Physics. [S0003-6951(99)02232-9]

Atomic hydrogenation introduced into a bulk or thin-film semiconductor can improve recombination and transport properties of electronic materials. A primary reason for such improvement is a hydrogen passivation of defects in the bulk and at the interface. This process can be strongly suppressed by hydrogen trapping, retarding the diffusion.\textsuperscript{1} It was found recently that ultrasound treatment (UST) applied to hydrogenated polycrystalline Si thin films effectively facilitates passivation of grain-boundary defects.\textsuperscript{2} As a result, UST processing can improve leakage current and threshold voltage of polycrystalline Si thin-film transistors.\textsuperscript{3} A physical reason of ultrasonically enhanced hydrogenation was attributed to a coupling of ultrasound with trapped hydrogen, release of the hydrogen from traps, which follows by a fast H diffusion and passivation of dangling Si bonds.\textsuperscript{4} It was anticipated that a similar UST process could benefit operational parameters in other hydrogen-based materials and devices.

In a previous study, it was reported that a hydrogenated amorphous-carbon (a-C:H) thin film on glass with the band gap of 3.2 eV is a promising luminescent material for the active layer in the injection-type thin-film light-emitting diode (TFLED).\textsuperscript{5} Specifically, the a-C:H active-layer TFLED showed a luminescence spectrum shifted toward higher energy (peak wavelength at 600 nm) compared to hydrogenated amorphous-silicon carbide (a-SiC:H, 2.4 eV) TFLED with a luminescence maximum at 720 nm. Additionally, the a-C:H active-layer TFLED had a threshold voltage of 3.5 V and on-current conductance of about 1.1 A/V/cm\textsuperscript{2}, which is noticeably lower than similar parameters of the a-SiC:H material, correspondingly, 7 V and 1.7 A/V/cm\textsuperscript{2}. However, the electroluminescence (EL) intensity of the a-C:H TFLED was weaker than that of the a-SiC:H TFLED, which was attributed to insufficient hydrogen passivation of nonradiative defects. In this study, we applied the UST technique to improve electrical and optical characteristics of the a-C:H light-emitting diodes.

The TFLEDs were fabricated using the photochemical vapor deposition (photo-CVD) cluster system, which consists of a load-lock chamber and three reaction chambers for a p layer of boron-doped \mu c-Si:H (2.3 eV, \sigma = 0.8 S/cm), an i layer of intrinsic a-SiC:H or a-C:H, and an n layer of phosphorus-doped \mu c-Si:H (2.4 eV, \sigma = 20 S/cm). The base pressure in each chamber was below 5 \times 10^{-7} Torr. Each layer was successively and independently processed in vacuum so that the interface layer conditions were free from experimental variations. Fabricated TFLEDs have an area of 0.0331 cm\textsuperscript{2} with a circular shape, showing reproducible electro-optical characteristics. Electroluminescence spectra were measured using a monochromator (Instrument S.A., HR. 640), a photomultiplier (Atago Bussan Co., No. 295), and a lock-in amplifier (Stanford Research System, SR510) under 200 Hz square wave with a peak-to-peak voltage (\textit{V}_{pp}) and a duty ratio of 20\% at an injection current density of 600 mA/cm\textsuperscript{2}. Optical output power was measured using an optical power meter Newport 1830-C with an 818-SL/CM-detector module. The film thickness and optical band gap (\textit{E}_{opt}) were calculated with a spectroscopic ellipsometer (JOBIN-YVON, UVISEL) using Tauc’s plot.

For the UST of hydrogenated TFLEDs, ultrasound vibrations were applied to the TFLEDs through a glass substrate using an external circular piezoceramic transducer of 70 mm diam. The transducer was driven by a function generator and a power amplifier adjusted to the main resonance frequency of the radial vibrations at 25 kHz. To maintain the system in the acoustic resonance during UST processing, the resonance frequency was \textit{in situ} controlled and automatically adjusted using a high-sensitive UST probe. In these experiments, the UST amplitude was controlled by ac voltage applied to the transducer. The UST time was fixed at 30 min at every processing step. The UST effect was monitored by measurement of the \textit{I–V} curves, EL spectrum, and optical output power. The on-current conductance (\sigma_{on}) was calculated as a line slope in the range of exponentially increasing current of the \textit{I–V} curves.

Figure 1 shows \textit{I–V} curves measured on a-C:H TFLED structures before and after room-temperature UST. Every \textit{I–V} curve corresponds to the individual device, which was...
processed for 30 min at different ac voltage applied to the ultrasound transducer. Importantly, before the UST, all the devices had identical $I-V$ curves similar to the initial curve in Fig. 1. The inset in Fig. 1 shows a dependence of the ultrasound amplitude on the UST transducer versus ac voltage. The UST amplitude is linearly increased between 0.5 and 1.0 V, and saturated above 2.0 V. This range of applied voltages and UST amplitudes was used in a current study. A major UST effect on $I-V$ curves is exhibited as a gradual decreasing of TFLED series resistance measured above 2.5 V of the voltage applied to the diode. Especially, significant changes were observed over a threshold voltage at 3.5 V. Furthermore, there was no variation of leakage current on UST-processed diodes. It appeared that the material properties of each layer in UST-processed diodes ($p,i,n$, SnO$_2$) did not change compared to the initial electrical characteristics of each layer.

We performed a statistical verification of this positive UST effect for TFLED series resistance. In Fig. 2 we present UST-induced changes of on-current conductance of the TFLEDs and corresponding values of their threshold voltages. For each UST amplitude, seven diodes fabricated on the same substrate were processed by ultrasound. Initially, the TFLEDs had a threshold voltage of 3.9 ± 0.03 V. The threshold voltage remained essentially the same after UST at 0.5, 1.0, and 1.5 V. Concurrently, the on-current conductance of all identical initial diodes was increased by a factor of 2 compared to the initial value and saturated at UST voltage on the transducer above 1.0 V.

EL spectra of $a$-C:H TFLEDs before and after UST were measured under the same injection current density of 600 mA/cm$^2$. At this injection level a conduction mechanism in the diode is attributed to Fowler–Nordheim tunneling, as was shown in the previous study. Figure 3 shows the dependence of the EL spectra on UST amplitude. EL intensity increased linearly with UST voltage up to 1.0 V, and then saturated at a level almost doubled compared to the initial intensity. This behavior is depicted in the inset graph. The EL spectra displayed the same peak wavelength of 600 nm before and after room-temperature USTs and a similar half width.

These results are consistent with the on-current conductance improvement shown in Fig. 2, and can be explained as follows. The voltage applied to the ultrasonically processed TFLED, which is required to flow the injection current of 600 mA/cm$^2$, is lower than the voltage for identical injection to the TFLED before UST. Therefore, due to the higher field applied to the untreated diode, the majority of the injected electrons and holes passed through the luminescent active $a$-C:H layer under the tunneling mechanism without recombination. Only a small portion of injected carriers participates in a radiative recombination. However, all injected carriers withstood series resistance at the $p/i$, $i/n$, SnO$_2/p$, and $n/Al$ interfaces. After UST, which reduces the series resistance, a lower electric field is applied to the luminescent active $a$-C:H layer to provide the same injection level. As a
result, tunneling carriers have a higher probability to recombine radiatively, which is reflected as ultrasonically increased EL intensity.

Figure 4 shows the optical output power of the a-C:H TFLED versus injection current density for three diodes processed at different UST voltages. The optical power is increased at all injection levels after room-temperature UST by a factor of 2 compared to the initial diode, which was kept as a reference. This experiment proves that the UST improves the luminescent efficiency at the same carrier injection. In other words, the probability of radiative recombination of the a-C:H TFLED is increased after the UST, because a lower electric field due to increased on-current conductance is applied to the active layer.

In conclusion, we demonstrated that room-temperature ultrasound treatment applied to a-C:H TFLEDs substantially reduces the series resistance of the diodes. Concurrently, no change was observed in the threshold voltage, peak position, and half width of the EL spectrum. An important result is the substantial factor of 2 increase of the EL intensity and optical output power of TFLEDs subjected to UST. This increase can be attributed to enhanced radiative recombination resulting from the lower field applied to the active a-C:H layer under the same current injection conditions. In the follow-up study, we further observed that UST applied at temperatures up to 130 °C also improves the recombination parameters of a-C:H layers. Our experiments allow extending an application of UST technology to electroluminescent materials for flat-panel displays.