A prototype active-matrix OLED using graphene anode for flexible display application

O Eun Kwon\textsuperscript{a}, Jin-Wook Shin\textsuperscript{a}, Himchan Oh\textsuperscript{a}, Chan-mo Kang\textsuperscript{a}, Hyunsu Cho\textsuperscript{a}, Byoung-Hwa Kwon\textsuperscript{a}, Chun-Won Byun\textsuperscript{a}, Jong-Heon Yang\textsuperscript{a}, Kang Me Lee\textsuperscript{a}, Jun-Han Han\textsuperscript{a}, Nam Sung Cho\textsuperscript{a}, Jong Hyuk Yoon\textsuperscript{a}, Seung Jin Chae\textsuperscript{a}, Jin Sung Park\textsuperscript{a}, Hyunkoo Lee\textsuperscript{a}, Chi-Sun Hwang\textsuperscript{a}, Jaehyun Moon\textsuperscript{a} and Jeong-Ik Lee\textsuperscript{a}

\textsuperscript{a}Reality Devices Research Division, Electronics and Telecommunications Research Institute (ETRI), Daejeon, Republic of Korea; \textsuperscript{b}Aerospace R&D Center, Hanwha Aerospace, Seongnam, Republic of Korea

ABSTRACT
From the very first time that graphene was used as a transparent electrode for OLED applications, the emergence of active-matrix (AM)-graphene OLED displays has been envisioned. Realizing this expectation, however, turned out to be difficult. Two obstacles are the growth and transfer of a large-area graphene film and the patterning of a graphene film into pixels. To solve these problems, a process of patterning a graphene film without surface contamination was developed. The fabrication of OLED panels by the patterned graphene anode on Gen 2(370 × 470 mm)-sized and flexible substrates was successfully demonstrated. In this work, oxide TFT arrays were combined as a switching backplane, and a pixelated graphene OLED was used as an emissive layer, to realize AM-graphene OLED displays. To explore the technical feasibility of flexible AM-graphene OLED displays, the aforementioned components were formed on a flexible substrate. For commercial-level production, all the processes that were used were chosen to be compatible with the conventional display processes.

1. Introduction
As a single entity, single-crystalline graphene has outstanding characteristics. The typical examples are high transmittance, outstanding chemical and thermal stability, high mobility, and mechanical bendability. Thus, graphene has been proposed as an active device component in transistors, sensors, and transparent electrodes. From the perspective of organic light-emitting diodes (OLEDs), graphene has drawn attention as a material for transparent electrode [1–3]. Due to the trade-off relation between sheet resistance and transmittance, it is not easy to find a material that is both highly conductive and transparent. In addition, graphene has good mechanical compliance, making it a prominent candidate in the field of flexible optoelectronics [4–7]. Graphene has been reported to have excellent bending characteristics compared to indium tin oxide (ITO), which is widely used as an OLED transparent electrode [8].

Since 2012, these researchers have focused on the use of graphene in OLEDs. In particular, a multi-layered graphene film was used as an anode in bottom-emission-type OLEDs. While graphene itself has outstanding properties, integrating it into a device is technically challenging. These researchers’ past research achievement may be summarized as overcoming the technical issues relevant to the graphene integration into OLEDs [9,10]. In the course of the research, graphene films obtained via chemical vapor deposition (CVD) were used. Thanks to the nature of the CVD growth method, uniform and large-area graphene can be obtained. In the beginning stage of the years 2012–2014, OLEDs with graphene anode suffered serious electrical instability. The instability was due to the surface roughness of graphene and the unfitting energy alignment to its adjacent hole transport layer. Later, the transfer of large-area graphene turned out to be difficult. Also, the fine patterning of graphene films into pixels emerged as an obstacle. Graphene has poor adhesion to glass or plastic substrates. In addition, due to the nature of the physical transfer process, it is practically impossible to achieve a defect-free status at the graphene/substrate interface. The patterning issue was overcome by the use of liquid bridging [11]. Liquid bridging brought forth three beneficial effects: increased adhesion of graphene to its substrate, elimination of the
Figure 1. (a) Gen 2 (370 × 470 mm) graphene anode OLED and (b) Flexible graphene anode OLED panels.

...air pore defects at the graphene/substrate interface, and surface planarization of the graphene film. These features allowed these researchers to implement photolithographic patterning methods with a clean graphene film surface. Figure 1 shows the two major breakthroughs these researchers have achieved. Figure 1(a) shows a fully functional Gen 2 (370 × 470 mm)-sized graphene anode OLED. This result signifies the technical feasibility of graphene as a commercially serviceable component in OLED displays. Figure 1(b) shows a two-colour flexible OLED panel. In addition to the accurate patterning of graphene on a polyimide (PI) film, flexible thin-film encapsulation and a laser lift-off process were realized without damaging the organic components of the OLED pixels. The example shown in Figure 1 is a passive-matrix graphene OLED.

The aim of this work was to develop a prototype that is directly applicable to the fabrication of flexible active-matrix (AM) graphene anode OLEDs. In addition to the various technical issues dealt with in the past, a thin-film transistor (TFT) array must be fabricated, and its driving scheme must be established. Briefly, an oxide TFT array was formed on a PI film/glass support. On the planarization layer of the TFT array, pixelated graphene was formed. Electrical connection between each TFT and graphene pixel was established through the standard via holes and contact pads.

2. Experiments

Figure 2 shows the detailed structure and process flow for the fabrication of a flexible AM-graphene OLED panel on a PI substrate. PI varnish was spin-coated on a cleaned glass substrate, and was thermally cured. The PI film was about 2 μm thick. A SiNₓ (20 nm)-SiO₂ (100 nm) buffer layer was deposited on the PI film via plasma-enhanced chemical vapor deposition (PECVD). Bottom-gate/top-contact (BGTC) oxide TFT arrays were prepared for driving the OLED device.

The structure of the BGTC TFT is shown in Figure 3, and the fabrication procedure is as follows. The OLED pixel driving circuit was composed of two TFTs and one capacitor (2T-1C). An Mo gate electrode was deposited on the buffer layer through a sputtering process, and was patterned. A SiOₓ gate insulator was deposited on top of the gate electrode to a thickness of 200 nm, using PECVD. Then an aluminum-doped indium tin oxide (Al-IZTO) active layer was deposited using the sputtering process. The composition of Al-IZTO was Al:In:Zn:Sn = 1.7:24.3:34:40, respectively (total thickness: 30 nm). After the patterning of the active layer, thermal annealing was done at 200°C in a vacuum atmosphere. The gate insulator layer was patterned for source/drain (S/D) electrode-gate electrode metal contact. The S/D electrode was deposited using Mo metal. The passivation layer for the planarization and protection of the TFT array was designed with an organic/inorganic multilayer structure. On the top of the passivation layer, SiOₓ was deposited for facilitating the graphene transfer.

To contact the graphene anode with TFT, a via hole and a transparent contact pad were formed at the top of the passivation layer. The contact via holes were opened from the top of the passivation layer to the driving TFT. The transparent contact pad was deposited via sputtering. The graphene film was transferred to the top of the prepared passivation layer. The four-layered graphene film was used as an anode; it had 70 Ω/sq sheet resistance and 80% transmittance. The adhesion to the substrate was improved by the combined liquid bridge-vacuum thermal treatment of the graphene film [11–14]. This treatment prevented the graphene film from peeling off.
Figure 2. Process flow of the flexible AMOLED panel.

Figure 3. Schematic cross section diagram of oxide TFT.

during the photolithography process. To fabricate the OLED anode, a graphene film was patterned via photolithography. The graphene surface was coated with photoresist (PR) using spin coating, and baking was performed on a hot plate. To pixelate the graphene film, a photomask was placed on top of the PR, after which it was exposed to ultraviolet (UV) light, followed by the development process. The opened area of the graphene film was removed by O2 plasma at 50 Watts, and the PR residual was cleaned with PR remover.

Red bottom emission OLEDs were fabricated on the PI substrate to evaluate the graphene patterning process at the OLED device level. The thermal evaporation method was used for fabricating the organic and metal cathode layer of the OLED. The structure of the OLEDs consisted of a pixelated graphene anode/hole transport layer (HTL) of 1,3-Bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPB) (70 nm)/cathode of lithium fluoride (LiF) and aluminum (Al) (total thickness: 150 nm). The electrical and optical properties of the OLEDs were measured using a source meter (Keithley 280) and a goniometer-equipped spectroradiometer (Minolta CS-2000). The OLED was encapsulated using an Al2O3 layer and was bonded with a commercial barrier film. A 30-nm-thick Al2O3 layer was formed via atomic layer deposition (ALD). Finally, the AM-graphene OLED panel was peeled off from the glass substrate using the laser lift-off process.

3. Results and discussion

3.1. Pixelated graphene anode

The purpose of this work was to pave the way for the use of graphene as the anode for the active-matrix OLED (AMOLED). Therefore, it is necessary to accurately pattern graphene on the AMOLED backplane. In a commercial display process, the transparent electrode is generally patterned via photolithography. Transferred graphene, however, has weak adhesion, and as such, it can be easily peeled off and damaged during the photolithography process.

The “liquid bridging process” was developed to overcome the aforementioned technical issue. This process improves the adhesion of the transferred graphene, enabling the subsequent photolithography patterning process to be carried out [11]. Figure 4 shows the optical microscope (OM) and scanning electron microscopy (SEM) images of the pixelated graphene electrodes formed on the planarization layer. The graphene pixels had a geometrically accurate size, as designed. As shown
in Figure 4, these images demonstrate the technical feasibility of forming graphene pixels on top of the AMOLED backplane in a stable state, without failure.

3.2. Device characteristics

As a preliminary test, phosphorescent red OLEDs with an array of patterned graphene films were fabricated as pixel anodes (Figure 5). Figure 5(a) shows the current density ($J$)-voltage ($V$)-luminance ($L$) plot. The $J$-$V$ and $L$-$V$ characteristics showed the typical behavior of a normally functional OLED. The $J$ and $L$ levels saturate around $10^{-1}$ A/cm² and $3 \times 10^4$ cd/cm². The inset of Figure 5(a) shows the actual emission images of the OLED with pixelated graphene anodes. As can be seen, all the pixels are working without deteriorated marks. The luminance efficiency (LE, lm/W) exhibits the typical roll-off behavior (Figure 5(b)). A 25 lm/W LE was achieved at the $7.5 \times 10^{-3}$ A/cm² $J$ level. The Figure 5(b) inset shows the normalized EL spectra of the OLEDs. The EL spectra of the graphene OLED show an aberrational feature, with the central peak at 625 nm. If the graphene surface has contaminants, the OLED device characteristics are expected to be either aberrant or non-working. The study results in Figure 5 show that the proposed graphene film patterning process can be used to fabricate OLEDs.

The fabricated oxide TFT exhibited 25 cm²/Vs mobility ($\mu$), as shown in Figure 6. Such $\mu$ may suffice for driving an AMOLED pixel. The leakage level does not exceed $10^{-12}$ A up to an applied voltage of 10 V, indicating the outstanding property of the proposed gate dielectrics.

3.3. Panel design of the AM-Graphene OLED prototype

Figure 7(a) shows the layout of the AM-graphene OLED panel, and (b) is the actual image of the fabricated panel on the PI film/glass support. The 2T-1C structure is employed for driving an AMOLED panel. Figure 7(c,d) are the unit pixel layout and schematics of the 2T-1C circuit, respectively. In the panel layout, test pixels and align keys were placed for process defect confirmation.

Figure 8 shows the operating image of the fabricated AMOLED panel prototype with the graphene anode and
red bottom emission OLED. The unit pixel size of the AMOLED panel was 222 × 222 μm. The panel resolution was 320 × 240 QVGA, and the screen size was 3.5 inches. From this panel, a flexible display could be obtained by separating PI from the glass using the LLO process.

4. Conclusion

In the past, these researchers successfully demonstrated passive-matrix-type organic light-emitting diode (OLED) panels with pixelated graphene films as transparent electrodes. To be specific, fully functional Gen
2(370 × 470 mm)-sized and flexible graphene anode OLEDs were demonstrated. With the aim of developing fully functional flexible AM-graphene OLED panels, the relevant technical issues were presented, and associated solutions were suggested, both on the integration and component levels. The importance of graphene film fine patterning and the use of the laser lift-off technology for realizing flexible panels on a realistic-sized substrate was emphasized. For demonstration purposes, an active-matrix OLED (AMOLED) panel with a QVGA (320 × 240) resolution and a 2T-1C per-pixel scheme was prepared. On a PI/glass support, an oxide thin-film transistor (TFT) array, a planarization layer, and a pixelated graphene array were sequentially formed. The fabrication steps are compatible with those of the existing display processes. It is believed that the proposed approach suggests a meaningful advancement in realizing the ultra-thin flexible display, in which the unique properties of graphene are crucial.

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Notes on contributors

O Eun Kwon received his B.S. and M.S. Electrical Engineering and Electric Physics degrees from Kwangwoon University, Seoul, South Korea in 2011 and 2015, respectively. He joined Electronics and Telecommunications Research Institute (ETRI), Daejeon, South Korea in 2017. His current research interests include printed electronics, flexible substrate for organic light-emitting diodes (OLEDs), graphene electrode application for OLEDs, and implantable biomedical devices.

Jin-Wook Shin received his B.S. and M.S. degrees from Myongji and Kwangwoon University, respectively. In 2018, he received his Ph.D. degree from Tohoku University, Japan. He joined ETRI in 2009. His current research interests include flexible OLED displays with graphene films, microdisplays for augmented reality/virtual reality (AR/VR), and implantable biomedical devices.

Himchan Oh received his B.S. and M.S. Materials Science & Engineering degrees from Seoul National University, Seoul, South Korea in 2008 and 2010, respectively. He is currently a senior researcher at ETRI in Daejeon, South Korea. His main research interests are the device and material aspects of the transition metal oxide semiconductor.

Chan-mo Kang received his B.S. and Ph.D. Electrical and Computer Engineering degrees from Seoul National University in 2008 and 2014, respectively. He is currently a senior researcher at ETRI in Daejeon, South Korea. His research interests include device engineering and physics in organic electronics, thin-film transistors (TFTs), flexible and printed electronics, and organic/inorganic hybrid devices.

Hyunsu Cho received his B.S. and Ph.D. Electrical Engineering degrees from Korea Advanced Institute of Science and Technology (KAIST), Daejeon, South Korea in 2008 and 2014, respectively. He joined ETRI Daejeon, South Korea in 2014, where he is currently a senior researcher. His research interests include the device physics and optical design of optoelectronics devices like OLEDs.

Byoung-Hwa Kwon received his B.S., M.S., and Ph.D. degrees from the Department of Materials Science & Engineering of Hanyang University, Seoul, South Korea, Pohang University of Science and Technology (POSTECH), and KAIST, respectively, in 2006, 2008, and 2012. After graduating, he worked at the Department of Materials Science & Engineering of the University of Florida in Gainesville, U.S.A. as a postdoctoral associate, and at LG Chem Ltd. as a senior researcher. He has been a senior researcher at ETRI since 2014. His current research interests are optoelectronic materials and devices like OLEDs and QD-LEDs, and thin-film encapsulation for flexible devices.

Chun-Won Byun received his B.S. and M.S. Electrical and Computer Engineering degrees from Hanyang University, Seoul, South Korea in 2002 and 2007, respectively. From 2007 to 2010, he worked for ETRI in Daejeon, South Korea, and from 2011 to 2013, he worked for Samsung Display, South Korea. Since 2013, he has been working at the Reality Devices Research Division of ETRI as a senior researcher. His research interests include the microdisplay for AR/VR, driving methods for new displays, and implantable biomedical devices.


**Jong-Heon Yang** received his B.S. and Ph.D. Electrical Engineering degrees from KAIST, South Korea in 2000 and 2018, respectively, and his M.S. Electronic Engineering degree from POSTECH, South Korea in 2002. He joined ETRI, South Korea in 2002. His research has focused on silicon nano-scale transistors, silicon nanowire biosensors, oxide memory TFTs, and active-matrix flat-panel displays using oxide TFTs.

**Kangme Lee** received her B.S. Polymer Engineering degree from Chungnam National University in 2015. Since 2015, she has been working at ETRI. Her current work involves the fabrication and optimization of OLED devices.

**Jun-Han Han** received his B.S. and M.S. Electronic Engineering degrees from Hanyang University, Seoul, South Korea in 2007 and 2009, respectively. He joined ETRI in Daejeon, South Korea in 2009 and continues to research on OLED lighting applications and next-generation displays.

**Nam Sung Cho** received his B.S. Chemistry degree from Chung-Ang University in Seoul, South Korea in 2000, and his M.S. and Ph.D. Chemistry degrees from KAIST in Daejeon, South Korea in 2002 and 2006, respectively. From 2006 to 2008, he was a postdoctoral associate at University of California Santa Barbara in Santa Barbara, CA, U.S.A. From 2008 to 2012, he worked on materials development for OLEDs at LG Display R&D Center in Paju, South Korea. He joined ETRI in Daejeon, South Korea in 2012. His current research interests include OLED structures, OLED materials, and white OLEDs.

**Jong Hyuk Yoon** received his B.S. Materials Science and Engineering degree from Kumoh National Institute and Technology in 2004, and his M.S. degree from University of Florida, Gainesville, FL, U.S.A. in 2007. He has been with Hanwha Aerospace (previously Samsung Techwin), South Korea since 2008, and has worked on graphene and its application research for commercialization, such as the high-quality and large-area graphene fabrication process for transparent conducting electrodes, sensors, barrier films, etc.

**Seung Jin Chae** received his B.S Semiconductor degree from Chonbuk National University in 2007, and his M.S. and Ph.D. degrees from Sungkyunkwan University in 2013. He was with Dongwoo FineChem, South Korea from 2013 to 2017, and worked on touch sensor fabrication with ITO and AgNW. He moved to Hanwha Aerospace in 2017, and his current research interests include high-quality and large-area graphene growth for various applications, such as transparent conducting electrodes, sensors, and barriers.

**Jin Sung Park** received his B.S. and M.S. degrees from Sungkyunkwan University, South Korea in 2001 and 2003, respectively, and his Ph.D. Physics degree from Tohoku University, Japan in 2008. He was also a postdoctoral associate at Tohoku and Kyushu University in Japan from 2008 to 2012. He joined Hanwha Aerospace in Seongnam, South Korea in 2012. His current research interests include large-area graphene film fabrication and its commercial applications.

**Hyunkoo Lee** received his B.S. and M.S. Electrical Engineering degrees from Seoul National University in 2004 and 2006, respectively. He was a junior researcher in the Monitor Circuit Design Team of LG Display, South Korea from 2006 to 2009. He received his Ph.D. Electrical Engineering degree in 2013, also from Seoul National University. He has been with ETRI as a senior researcher since 2013. His research interests include the device physics of organic semiconductors and organic/inorganic hybrid materials, and their applications to optoelectronics devices like OLEDs, QD (quantum dot)-LEDs, and printed electronics. His current research interests are optoelectronic devices with graphene and conducting polymer electrodes, thin-film encapsulation for flexible devices, and microdisplays for AR/VR.

**Chi-Sun Hwang** received his M.S. degree from Seoul National University in 1991, and his Ph.D. degree from KAIST in 1996, both in physics. From 1996 to 2000, he made DRAM devices at Hyundai Semiconductor Inc., South Korea. Since 2000, he has been with ETRI. His research has focused on display technologies based on the AM flat-panel display using TFTs, especially oxide TFTs. His current research interests are digital holography, large-area flexible electronics, and novel switching devices.

**Jaehyun Moon** received his B.S. degree from Korea University, Seoul, South Korea in 1995, and his Ph.D. Materials Science and Engineering degree from Carnegie Mellon University, Pittsburgh, Pennsylvania, U.S.A. in 2003. From 2003 to 2004, he was a postdoctoral associate at Max Planck Institute, Stuttgart, Germany. He joined ETRI in 2004. His current research interests include flexible electronics, functional nanomaterials, and OLEDs.

**Jeong-Ik Lee** received his B.S., M.S., and Ph.D. Chemistry degrees from KAIST in Daejeon, South Korea in 1992, 1994, and 1997, respectively. After graduating, he joined IBM Almaden Research Center in San Jose, CA, U.S.A. as a postdoctoral associate and worked on OLED materials. He joined ETRI in 1999 and continued his
research on OLED materials and devices. He has led the Reality Devices Research Division of ETRI since 2017 and has worked on the convergence of display and sensor technologies.

**ORCID**

*Hyunsu Cho* http://orcid.org/0000-0003-0182-6376

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