IEEE Photonics Journal
An IEEE Photonics Society Publication

Invited Paper

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Volume 4, Number 2, April 2012

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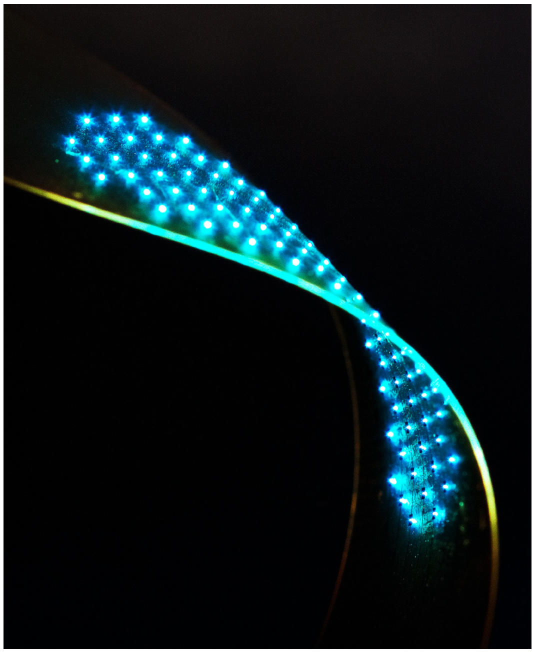

DOI: 10.1109/JPHOT.2012.2188998
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Microscale Inorganic Light-Emitting Diodes on Flexible and Stretchable Substrates

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(Invited Paper)

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DOI: 10.1109/JPHOT.2012.2188998
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Manuscript received February 15, 2012; accepted February 19, 2012. Date of current version April 20, 2012. Corresponding author: J. A. Rogers (e-mail: jrogers@illinois.edu).

Abstract: Recent work involving microscale inorganic light-emitting diodes on flexible and stretchable substrates is reviewed. Techniques for materials growth, device fabrication, and assembly are summarized, along with highlights of optical, electrical, and mechanical properties that can be achieved. Some examples of emerging applications in biomedical devices illustrate the value of these methods in areas where conventional approaches are unsuitable.

Index Terms: Microscale light-emitting diodes (LEDs), organic LEDs (OLEDs), flexible electronics, stretchable electronics, biomedical devices, transfer printing.

1. Introduction

The emergence of organic light-emitting diodes (OLEDs) as a commercial technology for lighting and display has stimulated interest in devices with ultrathin, flexible, lightweight construction [1], [2]. Recent research has explored strategies for using inorganic light-emitting diodes (referred to here as ILEDs to emphasize the difference) to similar ends, and in some cases with mechanical properties that extend beyond flexibility (i.e., bendability) to full, elastic responses under large strain deformation, i.e., stretchability [3]–[5]. The appeal of this technical approach is that it exploits properties, such as life span, brightness, and efficiency in ILEDs that exceed those of currently available OLEDs [6]. The most successful schemes combine inorganic material stacks grown on nonepitaxial substrates (e.g., glass [7]), in unconventional forms (e.g., three dimensionally structured templates [8]), or processed in unusual ways to yield releasable devices [3], [9] for integration onto surfaces of interest. The last process, when implemented with advanced techniques for deterministic device assembly, has many attractive features. Here, printing methods [10], [11] deliver organized arrays of microscale inorganic optoelectronic devices to sheets of plastic or slabs of rubber where they are integrated into functional systems. This type of technology has the potential to create wide-ranging classes of application, with characteristics and modes of use that cannot be addressed in any other way. Examples in biomedicine include components for optogenetics, blood oximetry, drug delivery, advanced surgical tools, and chemical sensors.

2. Transfer Printing

Deterministic assembly techniques that use elastomeric stamps to manipulate microscale ILEDs (μ-ILEDs) represent versatile, high-throughput manufacturing strategies for the systems reviewed

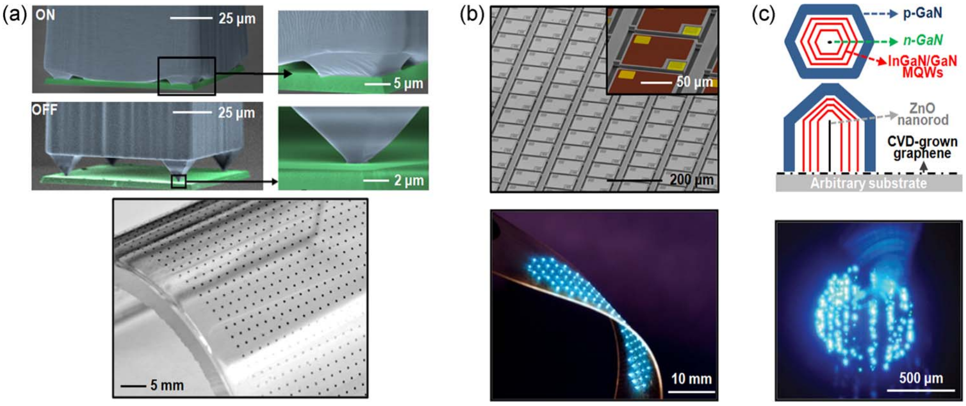

Fig. 1. (a) Deterministic assembly of microscale devices (green) using a structured elastomeric stamp (blue) with collapsible features of relief, shown here as colorized scanning electron micrographs in the adhesion ON (top) and OFF (middle) states. Reproduced with permission from [10]. Copyright National Academy of Sciences. The bottom frame shows an array of AlInGaP μ-ILEDs delivered by printing to the surface of a thin sheet of plastic that is subsequently bent around a glass cylindrical support. Reproduced with permission from [3]. Copyright American Association for the Advancement of Science. (b), (c) Unusual growth and processing strategies for InGaN LEDs and devices built with them. The top left frame shows InGaN μ-ILEDs released from an underlying silicon wafer growth substrate by anisotropic wet etching of the silicon; the frame below shows an array of such devices printed and interconnected on a thin, transparent sheet of plastic. Reproduced with permission from [4]. Copyright American Association for the Advancement of Science. The top right frame provides a schematic illustration of GaN and InGaN grown on nanorods of ZnO that form on graphene; the frame below shows a pattern of emission from a device made from such material. Reproduced with permission from [5]. Copyright Copyright WileyVCH Verlag.

here [10]. The underlying method, known as transfer printing, can be considered as a massively parallel “pick-and-place” technology that is compatible with extremely thin, fragile device components, originally developed for manipulating individual silicon transistors [10], [12], [13]. In this process, thin active layers or fully integrated devices formed on a growth wafer are released, retrieved with a stamp and then delivered to a foreign substrate. The key to successful operation is an engineered mechanism to modulate the adhesion to the stamp, from a strong state, for retrieval, to a weak one, for printing. Several approaches are available, ranging from those that use peel-rate dependent viscoelastic behaviors in the stamps [11], to pressure-modulated contact areas [10], to interfacial shear loading [14], each of which can be used for efficient transfer, even without separate adhesive layers on the target substrate. Fig. 1(a) shows, as an example of the second scheme, a contacting surface of a stamp of poly(dimethylsiloxane) (PDMS; blue) that supports compressible, pyramid-shaped features at the four corners [10]. Here, an applied preload collapses this relief to enable nearly full areal contact [adhesion ON state; Fig. 1(a) top] with a device element (green). Rapid retraction retrieves the device; shortly thereafter, elastic restoring forces push the collapsed region back into its initial, undeformed state, leaving contact only at the sharp tips of the pyramids [adhesion OFF state; Fig. 1(a) (middle)]. The adhesion is more than 1000 times stronger in the collapsed state than in the retracted one, due simply to differences in contact area for these two configurations. Fig. 1(a) (bottom) presents an image of a large array of AlInGaP μ-ILEDs printed onto a bent sheet of plastic using a related method for transfer printing [3].

3. ILEDs on Flexible Substrates

3.1. Devices in Planar Layouts

Various techniques exist for releasing planar layers of active materials grown on wafer substrates [3], [4], [15]–[18]. A recent approach that avoids cumbersome processes based on laser liftoff exploits InGaN stacks formed on silicon wafers with (111) orientation [4]. Etching through the InGaN defines lateral dimensions of devices that form by defining contacts to the p and n regions. Immersing the processed substrate in a bath of potassium hydroxide undercuts the silicon, which leaves freely suspended, but fully formed, ultrathin μ-ILEDs, each tethered at two of their corners to unetched regions of silicon (i.e., anchor bars). Typical lateral dimensions are 100×100 μm^2 and thicknesses are $\sim$4.8 μm. Fig. 1(b) (top) shows a scanning electron micrograph of a collection of InGaN μ-ILEDs and a colorized image in the inset (yellow—ohmic contacts; brown—current spreading layer), after undercut release from the silicon substrate (gray). Transfer printing allows the assembly of selected sets of these devices into desired layouts on plastic sheets or other foreign substrates, where they can be electrically interconnected. An example appears in Fig. 1(b) (bottom). Here, the small sizes of the devices facilitate efficient passive thermal spreading, to allow operation even on plastics or other materials with low thermal conductivity. The interconnects in this case rely on a backside optical exposure technique, in which a spin-cast, photocurable layer of benzocyclobutene serves as a planarizing film that is removed in regions shadowed by the opaque p and n contact pads. This process is important because it avoids the need for device-level packaging and wire-bonding, or for demanding alignment in photolithography; the result is then scalable to large collections of devices in arbitrary layouts over large areas. These procedures are immediately applicable with μ-ILEDs that have dimensions from 150×150 μm^2 to 25×25 μm^2, with even smaller dimensions readily achievable. Color modulation from blue to white is possible using patterned films of yellow phosphor dyes.

3.2. Devices in Nonplanar Layouts

As an alternative to GaN formed using planar strategies, GaN/ZnO coaxial nanorod heterostructures, including multiple quantum wells (MQWs) can be created by growth on films of graphene, as illustrated in the top schematic illustration of Fig. 1(c) [5]. Here, GaN forms epitaxially on ZnO structures first grown on the graphene [9]. Etching away an underlying layer of SiO_2 after growth yields released nanorod ILEDs on graphene, suitable for transfer onto plastic coated with copper for backside contacts. Devices with this configuration are mechanically flexible and capable of operation without noticeable degradation after 100 cycles of bending. The bottom frame of Fig. 1(c) shows representative patterns of emission. Although less technically mature than the strategies of Section 3.1, these ideas have merit and warrant further attention, particularly in aspects of the growth. For example, initial studies suggest that the grain structure in the graphene correlates to the densities of the resultant of GaN/ZnO nanorods. As evidence, graphene grown on copper foil leads to low densities of ZnO nanorods and high degrees of crystallinity, due at least in part to the large grains observed in graphene on copper. By comparison, graphene grown on nickel has small grains, which leads to densities in ZnO nanorods that are too high for effective growth of GaN.

4. ILEDs on Stretchable Substrates and Applications in Biomedicine

The concepts of Sections 2 and 3 can yield advanced systems that offer not only the mechanics of a flexible plastic sheet but also a stretchable rubber band. This latter capability is important because it enables integration of ILED technologies directly and intimately with the soft, curvilinear surfaces of the human body, in a noninvasive fashion [19]. Potential applications range from health monitors, to oximeters and highly functional surgical tools. The following summarizes the approaches and provides several device examples.

4.1. Interconnect Strategies for Stretchable ILEDs

Flexible lighting and display systems of the type described previously follow simply from the use of thin μ-ILEDs on thin, plastic substrates, sometimes in neutral mechanical plane layouts to enhance further the degree of bendability. Stretchable characteristics demand additional attention to the mechanics in order to avoid fracture of brittle, inorganic materials during large-scale deformations, where overall strains can, in certain cases, exceed 100%. The most powerful schemes incorporate layouts in which the interconnects absorb the applied strain in a way that mechanically isolates the μ-ILEDs. Interconnects with noncoplanar geometries in straight or serpentine shapes [3], [15], [20],

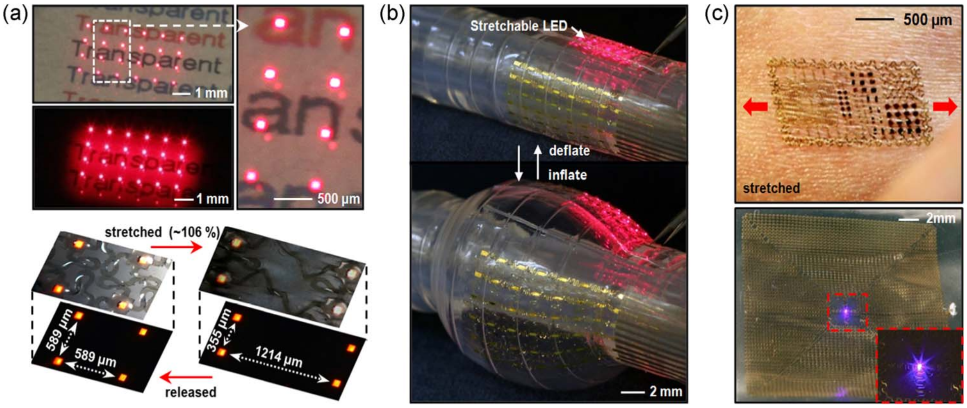

Fig. 2. (a) Collections of AlInGaP μ-ILEDs with transparent graphene interconnects on elastomeric substrates. Reproduced with permission from [20]. Copyright American Chemical Society. Applications of stretchable arrays of μ-ILEDs in advanced surgical tools (b) "instrumented" balloon catheters [23] and skin-mounted monitors as (c) "epidermal" electronic systems [24]. Reproduced with permission from [23] and [24], respectively. Copyright Nature Publishing Group for [23] and American Association for the Advancement of Science for [24].

[21], on either flat or structured elastomer supports, are effective. In these cases, controlled buckling and associated out-of-plane motions accommodate in-plane deformations, such that the strains in all of the constituent materials, except the elastomeric substrate, are small (e.g., $<$ 0.25%). Optimized designs, guided by quantitative mechanics modeling, enable stretching to 150% or more, without inducing fracture in any of the functional layers. The use of structured elastomers [22] or multilayer stacked configurations [15] enables this type of mechanics, even in systems that involve high areal coverage of active devices. In most cases, the interconnects consist of trilayer stacks of polymer/metal/polymer. Recent work shows that graphene can be used as an alternative, of interest here due to its optical transparency and its excellent mechanical properties [20]. The same concepts of buckling apply to graphene defined in narrow strips. The extremely low flexural rigidity of the graphene allows it to conform to the topography defined by the μ-ILEDs; the result facilitates efficient coverage on the contacts, over the edges of the devices. Examples of an array of AlInGaP μ-ILEDs with graphene interconnects appear in Fig. 2(a), demonstrating stretchability that exceeds 100%.

4.2. Applications in Biomedicine

Stretchable arrays of μ-ILEDs can be readily integrated into platforms that are suitable for natural, "soft" interfaces to the human body. For example, devices can be bonded to the surfaces of catheter balloons, to add advanced functionality to this otherwise conventional surgical tool. Applications for photoactivation of drugs, *in situ* spectroscopy or even optical ablation are possible, in minimally invasive modes. Fig. 2(b) shows an example that includes not only μ-ILEDs but also electrodes and various sensors, configured as a tool for monitoring and therapy in the context of cardiac surgery [23]. In another example, μ-ILEDs can be integrated with electronics in formats that have physical properties, ranging from thickness to bending rigidity, modulus and mass loading, all matched to the epidermis [24]. Mounting on the skin leads to a highly functional interface to the body, for health/wellness monitors, oximetry diagnostics, human–machine interfaces and others. Fig. 2(c) provides images of representative devices. In this case, inductive coupling provides a source of power.

5. Summary

Advanced methods in materials growth, processing, mechanics, thermal design, and system manufacturing combine to enable unusual modes of use for ILEDs. The outcomes have the potential to lead to applications that can complement those already well addressed by conventional forms of ILEDs or OLEDs. The range of engineering opportunities and avenues for basic scientific investigations create an appealing set of circumstances for future research and development.

Acknowledgment

T. Kim and R.-H. Kim contributed equally in this paper.

References

[1] F. So, J. Kido, and P. Burrows, "Organic light-emitting devices for solid-state lighting," *MRS Bull.*, vol. 33, no. 7, pp. 663–669, Jul. 2008.
[2] S. R. Forrest, "The path to ubiquitous and low-cost organic electronic appliances on plastic," *Nature*, vol. 428, no. 6986, pp. 911–918, Apr. 2004.
[3] S.-I. Park, Y. Xiong, R.-H. Kim, P. Elvikis, M. Meitl, D.-H. Kim, J. Wu, J. Yoon, C.-J. Yu, Z. Lui, Y. Huang, K.-C. Hwang, P. Fereira, X. Li, K. Choquette, and J. A. Rogers, "Printed assemblies of inorganic light-emitting diodes for deformable and semitransparent displays," *Science*, vol. 325, no. 5943, pp. 977–981, Aug. 2009.
[4] H.-S. Kim, E. Brueckner, J. Song, Y. Li, S. Kim, C. Lu, J. Sulkin, K. Choquette, Y. Huang, R. G. Nuzzo, and J. A. Rogers, "Unusual strategies for using indium gallium nitrides grown on silicon (111) for solid-state lighting," *Proc. Nat. Acad. Sci. USA*, vol. 108, no. 25, pp. 10 072–10 077, Jun. 2011.
[5] C.-H. Lee, Y.-J. Kim, Y.J. Hong, S.-R. Jeon, S. Bae, B. H. Hong, and G.-C. Yi, "Flexible inorganic nanostructure light-emitting diodes fabricated on graphene films," *Adv. Mater.*, vol. 23, no. 40, pp. 4614–4619, Oct. 2011.
[6] P. Patel, "Solid-state lighting: The future looks bright," *MRS Bull.*, vol. 36, pp. 678–680, Sep. 2011.
[7] J. H. Choi, A. Zoulkarneev, S. I. Kim, C. W. Baik, M. H. Yang, S. S. Park, H. Suh, U. J. Kim, H. B. Son, J. S. Lee, M. Kim, J. M. Kim, and K. Kim, "Nearly single-crystalline GaN light-emitting diodes on amorphous glass substrates," *Nat. Photonics*, vol. 5, no. 12, pp. 763–769, 2011.
[8] E. C. Nelson, N. L. Dias, K. P. Bassett, S. N. Dunham, V. Verma, M. Miyake, P. Wiltzius, J. A. Rogers, J. J. Coleman, X. Li, and P. V. Braun, "Epitaxial growth of three-dimensionally architecture optoelectronic devices," *Nat. Mater.*, vol. 10, no. 9, pp. 676–681, Jul. 2011.
[9] K. Chung, C.-H. Lee, and G.-C. Yi, "Transferable GaN layers grown on ZnO-coated graphene layers for optoelectronic devices," *Science*, vol. 330, no. 6004, pp. 655–657, Oct. 2010.
[10] S. Kim, J. Wu, A. Carlso, S. H. Jin, A. Kovalsky, P. Glass, Z. Liu, N. Ahmed, S. L. Elgan, W. Chen, P. M. Ferreira, M. Sitti, Y. Huang, and J. A. Rogers, "Microstructured elastomeric surfaces with reversible adhesion and examples of their use in deterministic assembly by transfer printing," *Proc. Nat. Acad. Sci. USA*, vol. 107, no. 40, pp. 17 095–17 100, Oct. 2010.
[11] M. A. Meitl, Z.-T. Zhu, V. Kumar, K. J. Lee, X. Feng, Y. Y. Huang, I. Adesida, R. G. Nuzzo, and J. A. Rogers, "Transfer printing by kinetic control of adhesion to an elastomeric stamp," *Nat. Mater.*, vol. 5, no. 1, pp. 33–38, Jan. 2006.
[12] E. Menard, K. J. Lee, D.-Y. Khang, R. G. Nuzzo, and J. A. Rogers, "A printable form of silicon for high performance thin film transistors on plastic substrates," *Appl. Phys. Lett.*, vol. 84, no. 26, pp. 5389–5400, Jun. 2004.
[13] K. J. Lee, M. J. Motala, M. A. Meitl, W. R. Childs, E. Menard, A. K. Shim, J. A. Rogers, and R. G. Nuzzo, "Large-area, selective transfer of microstructured silicon: A printing-based approach to high-performance thin-film transistors supported on flexible substrates," *Adv. Mater.*, vol. 17, no. 19, pp. 2332–2336, Oct. 2005.
[14] A. Carlson, H.-J. Kim-Lee, J. Wu, P. Elvikis, H. Cheng, A. Kovalsky, S. Elgan, Q. Yu, P. M. Ferreira, Y. Huang, K. T. Turner, and J. A. Rogers, "Shear-enhanced adhesiveless transfer printing for use in deterministic materials assembly," *Appl. Phys, Lett.*, vol. 98, no. 26, pp. 264104-1–264104-3, Jun. 2011.
[15] R.-H. Kim, D.-H. Kim, J. Xiao, B. H. Kim, S.-I. Park, B. Panilaitis, R. Ghaffari, J. Yao, M. Li, Z. Lui, V. Malyarchuk, D. G. Kim, A.-P. Le, R. G. Nuzzo, D. L. Kaplan, F. G. Omenetto, Y. Huang, Z. Kang, and J. A. Rogers, "Waterproof AlInP optoelectronics on stretchable substrates with applications in biomedicine and robotics," *Nat. Mater.*, vol. 9, pp. 929–937, Nov. 2010.
[16] W. S. Wong, T. Sands, N. W. Cheung, M. Kneissl, D. P. Bour, P. Mei, L. T. Romano, and N. W. Johnson, "$In_xGa_{1-x}N$ light emitting diodes on Si substrates fabricated by Pd–In metal bonding and laser lift off," *Appl. Phys. Lett.*, vol. 77, no. 18, pp. 2822–2824, Oct. 2000.
[17] E. Yablonovitch, T. Gmitter, J. P. Harbison, and R. Bhat, "Extreme selectivity in the lift-off of epitaxial GaAs films," *Appl. Phys. Lett.*, vol. 51, no. 26, pp. 2222–2224, Dec. 1987.
[18] J. Yoon, S. Jo, I. S. Chun, I. Jung, H.-S. Kim, M. Meitle, E. Menard, X. Li, J. J. Coleman, U. Paik, and J. A. Rogers, "GaAs photovoltaics and optoelectronics using releasable multilayer epitaxial assemblies," *Nature*, vol. 465, no. 7296, pp. 329–334, May 2010.
[19] J. A. Rogers, T. Someya, and Y. Huang, "Materials and mechanics for stretchable electronics," *Science*, vol. 327, no. 5973, pp. 1603–1607, Mar. 2010.
[20] R.-H. Kim, M.-H. Bae, D. G. Kim, H. Cheng, B. H. Kim, D.-H. Kim, M. Li, F. Du, H.-S. Kim, D. Estrada, S. W. Hong, Y. Huang, E. Pop, and J. A. Rogers, "Stretchable, transparent graphene interconnects for arrays of microscale inorganic light emitting diodes on rubber substrates," *Nano Lett.*, vol. 11, no. 9, pp. 3881–3886, Sep. 2011.
[21] D.-H. Kim, J. Song, W. M. Choi, H.-S. Kim, R.-H. Kim, Z. Lui, Y. Huang, K.-C. Hwang, Y.-W. Zhang, and J. A. Rogers, "Materials and noncoplanar mesh designs for integrated circuits with linear elastic responses to extreme mechanical deformations," *Proc. Nat. Acad. Sci. USA*, vol. 105, no. 48, pp. 18 675–18 680, Dec. 2008.
[22] J. Lee, J. Wu, M. Shi, J. Yoon, S.-I. Park, M. Li, Z. Liu, Y. Huang, and J. A. Rogers, "Stretchable GaAs photovoltaics with designs that enable high areal coverage," *Adv. Mater.*, vol. 23, no. 8, pp. 986–991, Feb. 2011.
[23] D.-H. Kim, N. Lu, R. Ghaffari, Y.-S. Kim, S. P. Lee, L. Xu, J. Wu, R.-H. Kim, J. Song, Z. Liu, J. Viventi, B. D. Graff, B. Elolampi, M. Mansour, M. J. Slepian, S. Hwang, J. D. Moss, S.-M. Won, Y. Huang, B. Litt, and J. A. Rogers,

Materials for multifunctional balloon catheters with capabilities in cardiac electrophysiological mapping and ablation therapy," *Nat. Mater.*, vol. 10, no. 4, pp. 316–323, Apr. 2011.

[24] D.-H. Kim, N. Lu, R. Ma, Y.-S. Kim, R.-H. Kim, S. Wang, J. Wu, S. M. Won, H. Tao, A. Islam, K. J. Yu, T.-I. Kim, R. Chowdhury, M. Ying, L. Xu, M. Li, H.-J. Chung, H. Keum, M. McCormick, P. Liu, Y.-W. Zhang, F. G. Omenetto, Y. Huang, T. Coleman, and J. A. Rogers, "Epidermal electronics," *Science*, vol. 333, no. 6044, pp. 838–843, Aug. 2011.