

## **Form factor-free, printed power sources**

\*Sang-Young Lee<sup>1)</sup>

<sup>1)</sup>*Department of Energy Engineering, School of Energy and Chemical Engineering,  
UNIST, Ulsan 44919, Korea*

<sup>1)</sup>[syleek@unist.ac.kr](mailto:syleek@unist.ac.kr)

### **ABSTRACT**

With the advent of ubiquitous electronics era, high-performance power sources with aesthetic diversity are indispensably needed as a key-enabling technology. . From a power source point of view, conventional rechargeable batteries (represented by lithium-ion batteries) with fixed shapes and dimensions have suffered from lack of variety in form factors, thus imposing formidable challenges on their integration into versatile-shaped electronic devices. Here, as an innovative strategy to address the aforementioned longstanding challenges, we demonstrate a new class of printed rechargeable power sources (including lithium-ion batteries and supercapacitors) with exceptional form factors and aesthetic versatility, which lie far beyond those achievable with conventional battery technologies. A key-enabling technology for this new battery system is to design battery inks (specifically, electrode and electrolyte inks), with a focus on their chemistry and rheology. Particular attention of this talk is devoted to discussing effects of the battery inks on printing processability, microstructure (focusing on bicontinuous ion/electron transport channels) and electrochemical performance of the printed batteries.

### **1. INTRODUCTION**

Forthcoming flexible/wearable electronics, the Internet of Things (IoT) and electric vehicles, which are expected to bring us unforeseen ubiquitous connections via electronic transmission, have spurred the relentless pursuit of advanced power sources with flexibility and reliable electrochemical performance. Enormous emphasis should be paid to form factors and flexibility of the batteries to achieve this challenging goal, along with continuing efforts for development of new electrochemically active materials. Meanwhile, from the battery architecture point of view, conventional batteries with fixed shapes and sizes are fabricated by winding or stacking cell components (such as anodes, cathodes, and separator membranes) and then packaging them with (cylindrical-/rectangular-shaped) metallic canisters or pouch films, finally followed by injection of liquid electrolytes. Under the conventional battery materials and assembly processes, the resulting batteries have limitations in form factors and mechanical

flexibility, thus imposing formidable difficulties on their integration into versatile-shaped electronic devices.

“Printed power sources” have recently emerged as a new battery system to address the aforementioned issues on the design diversity and flexibility. The printed batteries are fabricated through simple, low-cost and scalable printing processes. Their salient features include various form factors, shape conformability and monolithic integration with devices, which are difficult to achieve with conventional battery technologies. Printing technology [ref] is widely used in a variety of application fields (in particular, printed electronics) due to simple processability, reproducibility and versatility, in which rationally designed inks (including pastes) are printed in pre-designed forms. Research direction of the printed batteries is focused on design of battery shape and configuration, synthesis of battery component (*i.e.*, electrodes, electrolytes and separator membranes) inks with tunable rheological properties and electrochemical performance and adoption of suitable printing techniques, eventually enabling us to reach an ultimate goal of so-called “all-printed-batteries”. This talk presents current status and challenges of the printed batteries, with a focus on form factors, battery component inks, printing techniques, electrochemical performance and integration with other systems.

## **2. Results and Discussion**

### **I. Printed Solid-State Lithium-Ion Batteries**

To fabricate printed solid-state batteries (referred to as “PRISS” batteries), their major components (solid composite electrolyte (SCE) layer and SCE matrix-embedded electrodes) are consecutively printed on arbitrary objects of complex design/geometries through simple stencil printing process (followed by ultraviolet (UV) crosslinking). Notably, this printing process for PRISS batteries does not require traditional processing solvents such as acetone, water and N-methyl pyrrolidone (that are essentially used for preparation of conventional electrodes/self-standing polymer electrolyte films and also demand time-/energy-consuming drying processes), liquid-electrolyte injection and conventional microporous separator membranes, eventually leading to the fabrication of fully-integrated, multilayer-structured PRISS batteries with shape conformability and design universality far beyond those accessible with conventional cell components and manufacturing processes. Furthermore, such process simplicity and scalability of PRISS batteries allow seamless integration into complex-shaped electronic devices, thus enabling the realization of power source-unitized electronics.

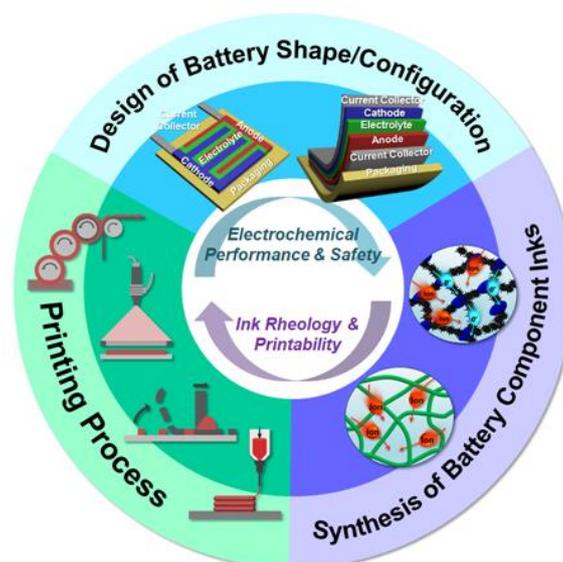
### **II. Monolithically integrated Lithium-Ion Batteries and Photovoltaic Cells**

Combining energy generation and energy storage systems is an ultimate solution to meet the ever-increasing demand for high-energy-density power sources. Here, we demonstrate a new class of monolithically integrated, photo-rechargeable portable power sources based on miniaturized crystalline Si photovoltaics (c-Si PVs) and printed solid-state lithium-ion batteries (LIBs). To enable the seamless architectural/electrical connection of the two different energy systems, an aluminium (Al) metal electrode of the c-Si PV module is simultaneously used as a current collector of the LIBs. The c-Si

PV module is fabricated by interconnecting 25 units of Si mini-cells, and it shows a high output voltage of 14.1 V and a PCE of 15.8%. The solid-state LIB with a bipolar cell configuration is directly fabricated on the Al metal electrode of the c-Si PV module via an ultraviolet (UV) curing-assisted printing process, which eventually enables the seamless architectural/electrical connection of the two different energy systems. The single-unit PV-LIB device shows exceptional electrochemical performance that lies far beyond those achievable by conventional PVs or LIBs alone, including fast, low-light-intensity, and high-temperature photo-charging, a photo-electric conversion/storage efficiency of 7.61%, sustainable cycling performance, and continuous discharging at an extremely high current density of 28 C under sunlight illumination.

### III. Monolithically integrated Lithium-Ion Batteries and Photovoltaic Cells

We present a new class of solid-state flexible power sources that are fabricated directly on conventional A4 paper using a commercial desktop inkjet printer. A salient feature of the inkjet-printed power sources is their monolithic integration with paper, i.e., they look like inkjet-printed letters or figures that are commonly found in office documents. A supercapacitor (SC), which is composed of activated carbon/carbon nanotubes (CNTs) and ionic liquid/ultraviolet-cured triacrylate polymer-based solid-state electrolyte, is chosen as a model power source to explore feasibility of the proposed concept. Cellulose nanofibril-mediated nanoporous mats are inkjet-printed on top of paper as a primer layer to enable high-resolution images. In addition, CNT-assisted photonic interwelding of Ag nanowires is introduced onto the electrodes to further improve electrical conductivity of the electrodes. The inkjet-printed SCs can be easily connected in series or parallel, leading to user-customized control of cell voltage and capacitance. Notably, a variety of all-inkjet-printed SCs featuring computer-designed artistic patterns/letters are aesthetically unitized with other inkjet-printed images and smart glass cups, underscoring their potential applicability as an unprecedented object-tailored power source.



**Figure 1** Research strategies for development of printed power sources (Lee, 2018).

#### 4. Conclusion

The printed batteries presented herein have garnered considerable attention as an innovative technology that breaks our common beliefs on power sources from the viewpoint of cell design. This talk describes an overview of the printed batteries, with a focus on battery component inks, printing technology, electrochemical performance, shape conformability/form factors and integration with devices. Future research of the printed batteries will be directed to synthesis of new battery component inks (based on chemical/rheological consideration) and adoption of newly emerging printing technologies such as high-fidelity inkjet and 3D printing. We envision that the printed batteries will open up a new avenue towards form factor-free/monolithic integrated power sources with object-tailored design versatility, which play a vital role in the upcoming flexible/wearable electronics, IOTs and ubiquitous energy applications.

#### Acknowledgments

This work was supported by the Basic Science Research Program (2017M1A2A2044501, 2018R1A2A1A05019733) and Wearable Platform Materials Technology Center (2016R1A5A1009926) through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and future Planning.

#### REFERENCES.

- 1) Keun-Ho Choi, David B. Ahn, and Sang-Young Lee\*, "Current Status and Challenges in Printed Batteries Toward Form Factor-Free, Monolithic Integrated Power Sources" **ACS Energy Lett.** **2018**, 3, 220.
- 2) Eun-Hye Kil, Keun-Ho Choi, Hyo-Jeong Ha, Sheng Xu, John A. Rogers, Mi Ri Kim, Young-Gi Lee, Kwang Man Kim, Kuk Young Cho\*, Sang-Young Lee\* "Imprintable, bendable, and shape-conformable polymer electrolytes for versatile-shaped lithium-ion batteries" **Advanced Materials** **2013**, 25, 1395.
- 3) Se-Hee Kim, Keun-Ho Choi, Sung-Ju Cho, Sinho Choi, Soojin Park, and Sang-Young Lee\* "Printed Solid-State Lithium-Ion Batteries: A New Route Toward Shape-Conformable Power Sources with Aesthetic Versatility for Flexible Electronics" **Nano Letters** **2015**, 15, 5168.
- 4) Keun-Ho Choi, JongTae Yoo, Chang Kee Lee and Sang-Young Lee\* "All-inkjet-printed, solid-state flexible supercapacitors on paper" **Energy & Environ. Sci.** **2016**, 9, 2812.
- 5) Han-Don Um, Keun-Ho Choi, Inchan Hwang, Se-Hee Kim, Kwanyong Seo, and Sang-Young Lee\*, "Monolithically integrated, photo-rechargeable portable power sources based on miniaturized Si solar cells and printed solid-state lithium-ion batteries", **Energy & Environ. Sci.** **2017**, 10, 931.