

# Elastic Spongy Graphene-Functionalized Silicon Anode with Excellent Cycle Stability in Li battery

Byong-June Lee, Chunfei Zhang, Tong Hyeon Kang, and Jong-Sung Yu\*

Department of Energy Science and Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

E-mail: [jsyu@dgist.ac.kr](mailto:jsyu@dgist.ac.kr)

Graphite plays a prominent role as a typical anode material in the lithium ion batteries (LIBs) because of its high lithiation-dilithiation reversibility and low voltage window. Unfortunately, the capacity is limited to 372 mAh g<sup>-1</sup>[1,2]. To search for materials with higher lithium storage capacity, a great number of investigations on metal oxides (or sulfides), Sn, P, and Si have been carried out in recent decades. Among these materials, silicon can make alloy with lithium in the form of Li<sub>12</sub>Si<sub>5</sub> to deliver a highest theoretical gravimetric capacity of ~4200 mAh g<sup>-1</sup>, and thus is considered to be one of the most promising anode materials for next generation LIB. It is worth mentioning that its quite low delithiation potential and high lithium storage capacity can provide a wide working voltage window and energy density, which enable promising potential application in electric vehicles. However, those advantages are seriously offset by a great challenge of large volume expansion during lithiation process and the resultant breakage of bulk silicon particles and solid electrolyte interface (SEI), which causes a serious damage to the electrode structure and thus gives rise to a fast decay of the specific capacity [3].

In this work, novel 3D spongy grapheme (SG)-functionalized silicon is for the first time demonstrated by chemical vapor deposition for a LIB anode, which can overcome the common silicon anode issues such as poor conductivity and volume expansion of Si as well as transfer of Li ion towards the Si. The elastic feature of graphene has excellent function to self-adaptively buffer the volume variation during charge-discharge process. In particular, different from traditional graphene or carbon shells (core-shell and yolk-shell), the spongy 3D graphene networks provide much improved unique functions with excellent long-cycle stability and rate capability. The Si@SG electrode exhibits excellent cycling performance with high reversible specific capacity [4]. A superior 95% capacity retention is achieved after 510 cycles. All the electrochemical performances get benefits from the well-designed functional SG shells, where interconnected nano-graphene structure not only guarantees a high conductive network but also provides more free paths for excellent mass transfer in addition to self-adaptive buffering capability.

## References:

- [1] B. Fang, J. H. Kim, M.-S. Kim, J.-S. Yu, *Acc. Chem. Res.* 46 (2013) 1397-1406.
- [2] C. Zhang, J.-S. Yu, *Chem. Eur. J.* 22 (2016) 4422-4430.
- [3] M. Zhou, X. Li, B. Wang, Y. Zhang, et al. *Nano Lett.* 15 (2015) 6222-6228.
- [4] C.Zhang, T.-H. Kang, J.-S. Yu, *Nano Research*, 11(2018) 233-245.