

## ***In Situ* TEM Observation on the Agglomeration of Nanoparticles in the Interface of SnO<sub>2</sub>**

Jun Young Cheong<sup>1</sup>, Joon Ha Chang<sup>1,2</sup>, Sung Joo Kim<sup>1,2</sup>, Chanhoon Kim,<sup>1</sup> Hyeon Kook Seo<sup>1,2</sup>, Jae Won Shin<sup>2</sup>, Jong Min Yuk<sup>1</sup>, Jeong Yong Lee<sup>1,2</sup> and Il-Doo Kim<sup>1</sup>

<sup>1</sup>. Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology, Daejeon, Korea (305-701)

<sup>2</sup>. Center for Nanomaterials and Chemical Reactions, Institute for Basic Science (IBS), Daejeon, Korea (305-701)

Direct observation of nanoparticles in high resolution has attracted considerable attention, as it can provide the fundamental understanding that is crucial to manufacturing the nanoscale materials that have different morphologies (e.g. sizes and shapes). Recently, very feasible *in situ* TEM platform called ‘graphene liquid cell’ (GLC) has been developed, which is easy to fabricate while maintaining high resolution imaging to better understand the dynamics of nanoparticles [1].

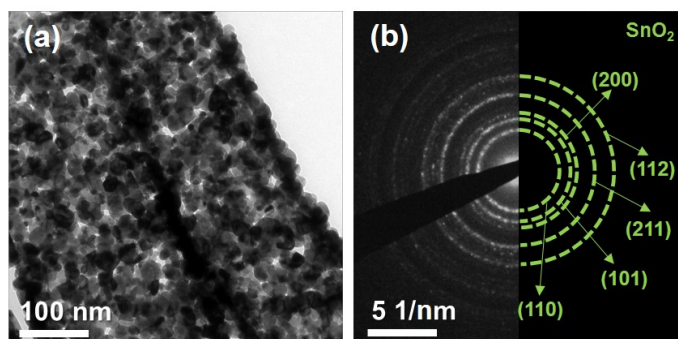
In the last two decades, coupled with the increasing energy demands and larger consumption of energy, various kinds of rechargeable energy storage systems have been highly researched [2]. Especially, much research has been devoted to utilize nanoscale materials as the feasible electrodes for rechargeable energy storage system [3]. In particular, nanoparticles have been researched as the potential electrode materials due to various merits that they have, such as larger reaction sites for energy storage [3]. Nevertheless, agglomeration of nanoparticles has been one of highly critical dilemmas, it leads to capacity fading stemming from the less number of reaction sites due to the enlargement of nanoparticles. To resolve this issue, incorporating nanoparticles into carbon matrix has been attempted previously [4], which may solve some immediate issues. Nevertheless, more comprehensive understanding on how the agglomeration of nanoparticles takes place still remains unknown as it is hard to conduct *in situ* observation on such phenomenon.

In this work, we have conducted *in situ* TEM observation on how the nanoparticles undergo agglomeration during lithiation (when lithium ions react with the electrode materials) to better understand the overall dynamics. Using GLC, we have encapsulated the SnO<sub>2</sub> nanotubes inside the graphene sheets, with an electrolyte composed of 1.3 M of lithium hexafluorophosphate (LiPF<sub>6</sub>) in ethylene carbonate(EC):diethylene carbonate (DEC) (v/v = 3:7) with 10 wt% of fluoroethylene carbonate (FEC). TEM images of SnO<sub>2</sub> nanotubes in low magnification and SAED patterns of SnO<sub>2</sub> nanotubes are shown in Figure 1, exhibiting polycrystalline nature of SnO<sub>2</sub>. The dynamics of nanoparticles in the interface of SnO<sub>2</sub> were visualized in a time-series from 0 s to 100 s. It is revealed based on the microscopic observation that the individual nanoparticles, once discretely separated from one another, later undergo agglomeration to be formed into larger nanoparticles. At 100 s, it can be suggested that the interface layer is formed on the surface of SnO<sub>2</sub>, which are composed of agglomerated nanoparticles, which are quite different from the TEM image at 0 s. As can be seen, agglomeration behaviour of nanoparticles upon lithiation is a common phenomenon, which requires additional analyses for further elucidation.

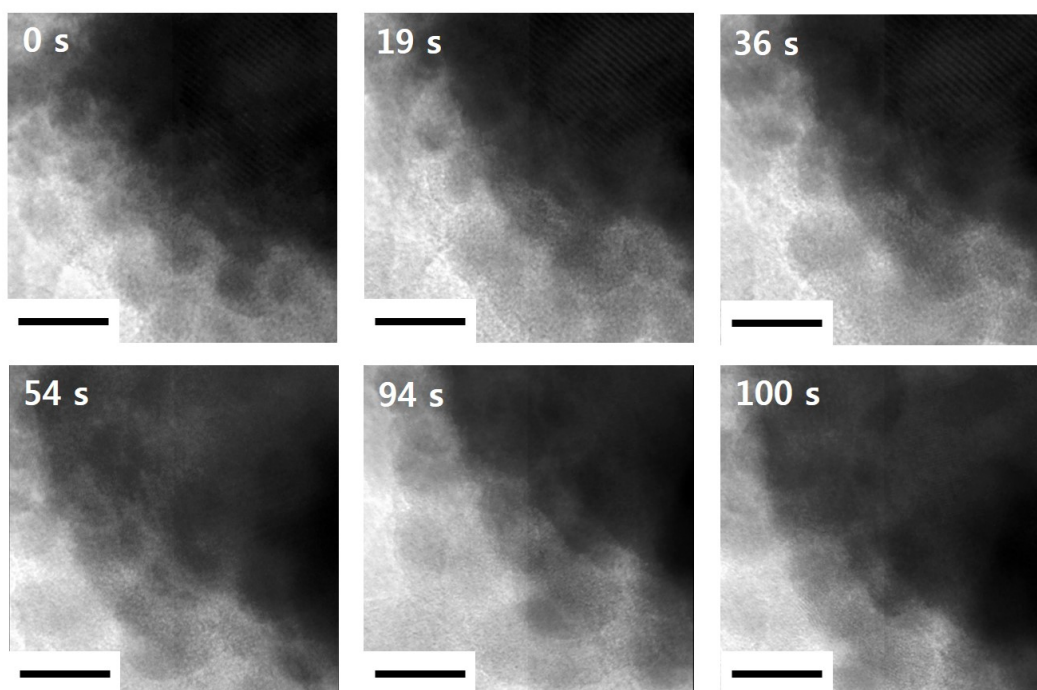
To conclude, we have firstly attempted to understand how the nanoparticles behave in the interfaces of active materials upon lithiation by conducting *in situ* TEM observation. In the interfaces of SnO<sub>2</sub>, the case material in this study, the individual nanoparticles start to merge with each other as time proceeds and the larger nanoparticles are formed as time proceeds. Based on this study, it is expected that more *in situ* TEM observation could be conducted by using GLC to probe the dynamics of nanoparticles in the liquid condition [5].

## References:

- [1] J. M. Yuk *et al*, *Science* **336** (2012), p. 61.  
[2] P. G. Bruce *et al*, *Nat. Mater.* **11** (2012), p. 19.  
[3] A. S. Arico *et al*, *Nat. Mater.* **4** (2005), p. 366.  
[4] A. Jahel, *et al*, *Adv. Energy Mater.* **4** (2014), p. 1400025.  
[5] The authors acknowledge the funding from Korea CCS R&D Center (KCRC) grant by the Korea government (Ministry of Science, ICT & Future Planning) (No. NRF-2014M1A8A1049303), Wearable Platform Materials Technology Center (WMC) (NR-2016R1A5A1009926), and Institute for Basic Science (IBS) (IBS-R004-G3).



**Figure 1.** (a) TEM images and (b) SAED patterns of SnO<sub>2</sub> nanotubes.



**Figure 2.** Time-series TEM images showing the agglomeration of nanoparticles in the interface of SnO<sub>2</sub> during lithiation.