

**Atomistic observation of the lithiation and delithiation behaviors of silicon nanowires using reactive molecular dynamics simulations**Sang Soo Han<sup>1,\*</sup>, Hyun Jung<sup>1,2</sup>, Minho Lee<sup>1</sup>, Byung Chul Yeo<sup>1</sup>, Kwang-Ryeol Lee<sup>1</sup><sup>1</sup>KIST; <sup>2</sup>Hanyang University

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In this talk, we will discuss the mechanisms for the lithiation and delithiation of Si NWs determined using a large-scale molecular dynamics (MD) simulation with a reactive force field (ReaxFF). The ReaxFF is developed using results from first-principles calculations for various crystals and molecules. During the lithiation process, Li atoms penetrate into the lattices of the crystalline Si (*c*-Si) NWs preferentially along the  $\langle 110 \rangle$  or  $\langle 112 \rangle$  direction, and then the *c*-Si changes into amorphous  $\text{Li}_x\text{Si}$  (*a*- $\text{Li}_x\text{Si}$ ) phases due to the simultaneous breaking of Si-Si bonds as a result of the tensile stresses between Si atoms. Before the complete amorphization of the Si NWs, we observe the formation of silicene-like structures in the NWs that are eventually broken into low-coordinated components, such as dumbbells and isolated atoms. Additionally, during delithiation of  $\text{Li}_x\text{Si}$  NWs, we observe the formation of a small amount of *c*-Si nuclei in the *a*- $\text{Li}_x\text{Si}$  matrix below a composition of  $\text{Li}_{1.4}\text{Si} \sim \text{Li}_{1.5}\text{Si}$ , and we demonstrate that the two-phase structure can be thermodynamically more favorable than the single-phase *a*- $\text{Li}_x\text{Si}$ .