## Secondary Layer Evolution of Bilayer Graphene on Copper

Ning Yang and Hyung Gyu Park\*

Nanoscience for Energy Technology and Sustainability Department of Mechanical and Process Engineering ETH Zürich Zürich CH-8092, Switzerland \* parkh@ethz.ch

## Abstract

The synthesis of bilayer graphene by chemical vapor deposition (CVD) on copper has been challenged by its self-limiting nature. The formation of continuous graphene layer prevents carbon supply from arriving at the underneath second layer, thus limiting the second-to-first-layer yield of the bilayer graphene. Here, we report a simple regrowth method capable of synthesizing the second layer graphene selectively with minimal change in size of the first layer. This regrowth method consists of two sequencing CVD processes with different hydrogen partial pressures and/or temperatures. We allocate the edge termination state of top layer graphene as the key parameter for this layer selective growth. A reaction mechanism based on hydrogen and carbon adatoms diffusion as well as carbon incorporation is applied to describe the saturation behaviour of the second layer graphene growth. The growth limiting step is found to be time dependent, transitioning from diffusion-limited to reaction-limited regimes.



## Figures