Tunable and Scalable Synthesis of ZnO Nanostructures using ALD Seed Layers

Alondra M. Ortiz-Ortiz¹, Andrew J. Gayle¹, Jing Wang¹, Daniel O. Delgado², Daniel Penley¹, Hannah R. Faustyn¹, Kalyn M. Fuelling², Ashley R. Bielinski^{1,3}, Caleb Sherwood¹, Neil P. Dasgupta^{1,2}

¹Department of Mechanical Engineering, University of Michigan ²Department of Materials Science and Engineering, University of Michigan ³Materials Science Division, Argonne National Laboratory

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Abstract: There has been significant advancement in the synthesis of nanostructured materials for use in biological, environmental, and energy fields in recent decades, with many novel processes demonstrated at the laboratory scale. Nevertheless, challenges remain at the nanomanufacturing frontier, where it is necessary to maintain tunable material structure and properties



using high-volume, high-throughput, and low-cost processes. One of the most facile techniques for the tunable and scalable synthesis of nanostructured materials is atmospheric-pressure hydrothermal synthesis of ZnO, which occurs under mild conditions. We have previously demonstrated that by using Atomic Layer Deposition (ALD) to deposit tunable "seed layers" with sub-monolayer precision in composition and structure, we can programmably tune geometric parameters (spacing, length) of the resulting ZnO nanostructures [1]. We refer to this process as "surface directed assembly", which has been used to enable applications ranging from photocatalysis to anti-biofouling surfaces [2,3].

In this study, we explore a third dimension of programmable control: using ALD seed layers to tune the ZnO nanostructure shape. Specifically, we demonstrate synthesis of Al-Zn-O nanosheets (NSs) using the surface directed assembly process by depositing a seed layer of Al_2O_3 . The formation of a zinc-aluminum layered double hydroxide phase occurs from the interactions between the Al and Zn^{2+} ions during the hydrothermal synthesis, as confirmed by X-ray Diffraction (XRD). We demonstrate that the density of the ZnO NSs can be controlled by the number of cycles of Al_2O_3 . Furthermore, Scanning Electron Microscopy (SEM) analysis shows that a transition from NS to nanowires (NWs) occurs after extended growth time. To rationalize this behavior, we propose a "phase diagram" for surface directed assembly of nanostructures using ALD. Additionally, we demonstrate the ability to scale-up this surface-directed assembly process

onto non-planar, large (cm-scale) surfaces facilitated by the design of a customized flow reactor. This work provides a new pathway to scalable nanomanufacturing enabled by the precise interfacial tunability of ALD for macroscopic applications, including medical devices and marine sensors.

References:

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