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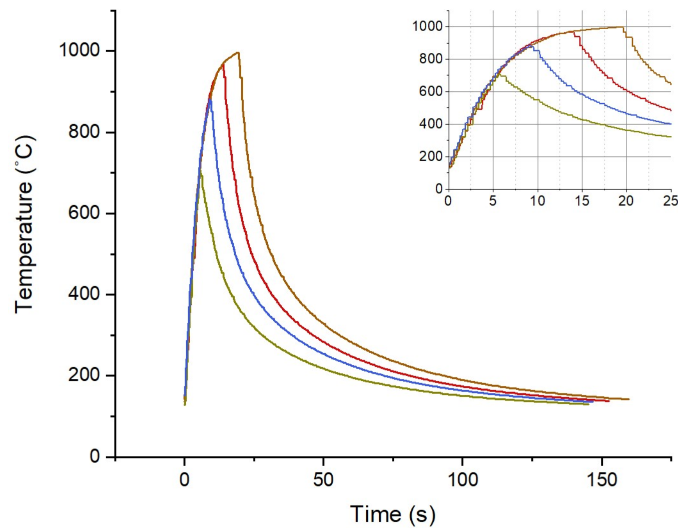
PULSED HEATING ATOMIC LAYER DEPOSITION (PH-ALD) FOR EPITAXIAL GROWTH OF ZINC OXIDE THIN FILMS ON C-PLANE SAPPHIRE

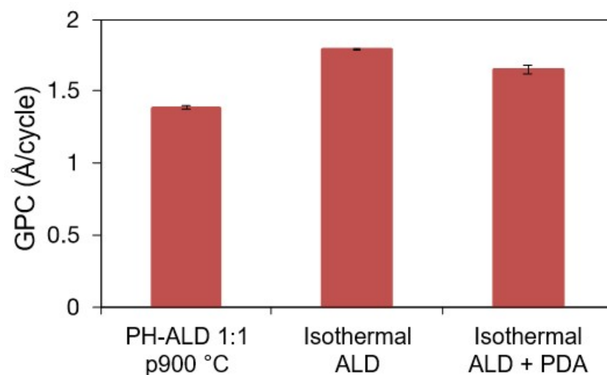
Brandon D. Piercy,[1] Jamie P. Wooding,[1] Shawn A. Gregory,[1] Mark D. Losego[1]

[1]School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, GA



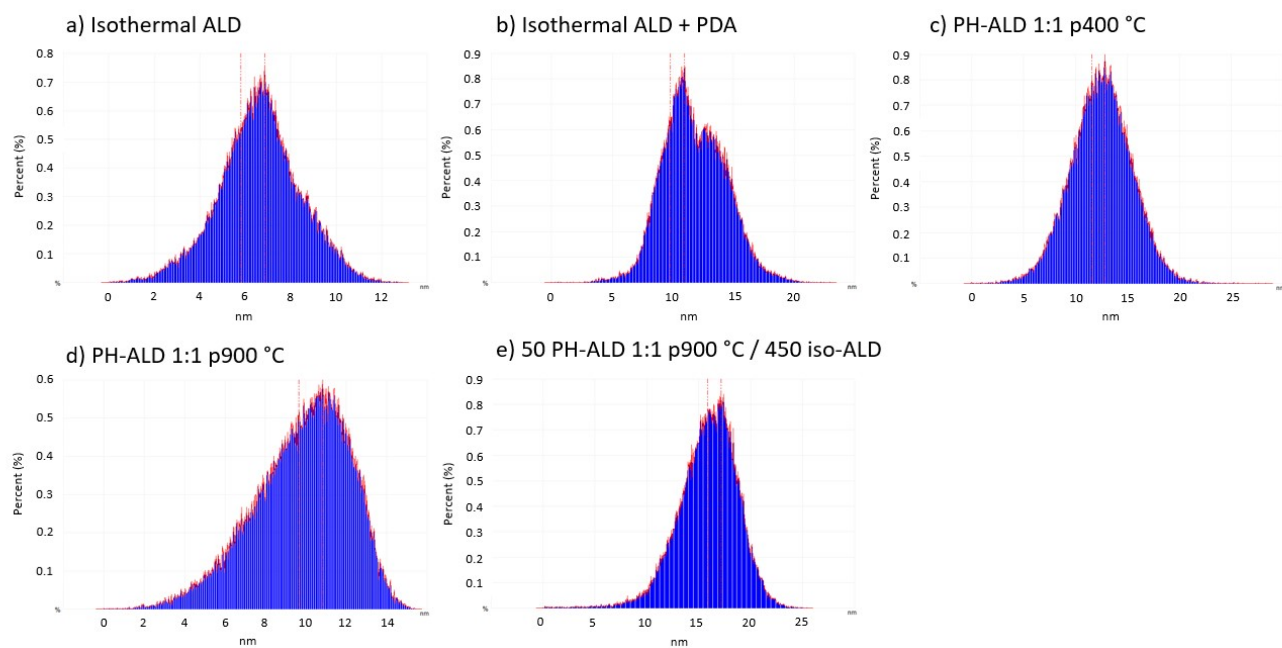
ESI Figure 1. *Thermal response of AlN/W heater during the heat pulse and cooling period for pulse durations of 5-20 seconds. (inset) Magnified view of the heat pulse period.*

Characteristic thermal responses for the AlN/W heater with heat pulses of 5-20 seconds are shown in ESI Figure 1. In this configuration, we can reach a temperature of 700˚C within 5 seconds and 1000 ˚C after 20 seconds. The time to return to the ambient chamber temperature of 110˚C remains about 3 minutes for all pulse temperatures. The thermal response remains constant over up to 500 cycles within a single run.



ESI Figure 2. *Growth rates between ALD, ALD with post-deposition anneal, and a 900 ˚C, 1:1 PH-ALD sample measured over 500 deposition cycles.*

ESI Figure *2* reports the growth per cycle (GPC) for PH-ALD 1:1 p900 ˚C, isothermal ALD, and isothermal ALD + PDA films. Isothermal ALD films consistently grew at a rate of 0.18 nm/cycle, consistent with literature reports for the ALD of ZnO from DEZ-H_2O chemistry. PH-ALD 1:1 samples under the most aggressive growth condition of a 900 ˚C heat pulse every ALD cycle did show a slight reduction in growth rate to 0.14 nm/cycle. This is likely caused by increased densification from the heat pulses, similar to behavior observed in PH-ALD reports in other material systems.[1-2] Regardless, the still relatively high growth rate also indicates that the system is not sensitive to any evaporation of the ZnO surface layers as sometimes reported by other techniques.[3]



ESI Figure 3. *Image histogram depth analysis for AFM results for a) isothermal ALD, b) isothermal ALD + PDA, c) PH-ALD 1:1 p 400 °C, e) PH-ALD 1:1 p900 °C, and e) 50 PH-ALD 1:1 p900 °C / 450 isothermal ALD.*

ESI Figure 3 presents the image histogram for a representative AFM image for the isothermal ALD films and selected PH-ALD films for reference.

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3. Ogata, K.; Maejima, K.; Fujita, S.; Fujita, S., Growth Mode Control of Zno toward Nanorod Structures or High-Quality Layered Structures by Metal-Organic Vapor Phase Epitaxy. *Journal of Crystal Growth* **2003**, *248*, 25-30.