ALD & ALE
Room Tamna Hall A - Session ALDALE-MoA

Student Award Session

Moderators: Jihwan An, Pohang University of Science and Technology (POSTECH), **Parag Banerjee**, University of Central Fllorida

1:30pm **ALDALE-MoA-1 ALD Student Award Finalist Talk: Integrating Machine Learning into Atomic Layer Deposition: A Case Study on Hafnium Oxide Process Optimization***, **Minjong Lee**, Doo San Kim, Thi Thu Huong Chu, Dushyant Narayan, Dan Le, Soubhik De,* University of Texas at Dallas*; Si Joon Kim,* Kangwon National University, Republic of Korea*; Jiyoung Kim,* University of Texas at Dallas

Industry 4.0 integrates intelligent information technologies into manufacturing, with machine learning (ML) offering significant potential to enhance flexibility, improve efficiency, and minimize process errors.[1] ML integration is particularly valuable in semiconductor fabrication, where deposition processes such as ALD require a comprehensive understanding of numerous parameters—especially with the rise of three-dimensional (3D) structures. Leveraging ML is expected to accelerate process optimization and expand ALD applications in cutting-edge semiconductor manufacturing. While real-time monitoring with self-feedback capabilities is the most effective strategy for ML-driven ALD, the increased processing time and maintenance demands of *in-situ* monitoring systems present challenges. This study introduces a “pipeline research” approach, outlining strategies for integrating ML into ALD processes as a foundation for digital twin technologies.

Case studies focus on ALD HfO_x film deposition, extending beyond conventional ML applications that have primarily been restricted to film thickness prediction.[2] While thickness remains a key metric within ALD windows, it provides only a partial understanding of film quality. To address this, film density was incorporated as an additional critical factor to ensure optimal film characteristics. A deep neural network (DNN) model was employed instead of traditional regression-based ML algorithms to improve predictive accuracy by capturing the complex, non-linear features of ALD processes. Such a model not only enhances film property predictions but also generates prediction maps of film properties, which were further analyzed to explore broader parameter spaces across an extended range. Additionally, investigations were conducted to determine the optimal dataset size required for effectively training a DNN system, providing key guidelines for integrating ML into ALD processes. The advantages of using DNNs for ALD process optimization will be comprehensively highlighted, particularly in their ability to efficiently handle high-dimensional process parameters.

This analysis demonstrates how ML-driven innovations simplify experimental design and reshape ALD process optimization strategies. The presentation will cover technical methodologies, integration strategies, and the workflow of prototype implementations for ML-driven ALD studies, offering insights into the future of autonomous ALD process control systems.

This work was supported by KEIT granted by MOTIE (Nos. 20010806 and 1415187770) and NRF (RS-2024-00450836).

[1] K. J. Kanarik et al., Nature 616, 707 (2023).[2] A. Arunachalam et al., JVST A 40, 012405 (2022).

1:45pm **ALDALE-MoA-2 ALD Student Award Finalist Talk: The AtomicLimits ALD/E Database: Unlocking the Future of ALD/E with Large Language Models***, **Eleni Poupaki**,* Eindhoven University of Technology, Netherlands*; Sameer Sadruddin, Jennifer D’Souza,* TIB Leibniz Information Centre for Science and Technology, Germany*; Alex Watkins, Bora Karasulu,* University of Warwick, UK*; Sören Auer,* TIB Leibniz Information Centre for Science and Technology, Germany*; Adrie Mackus, Erwin Kessels,* Eindhoven University of Technology, Netherlands

Launched by TU/e in 2019, the *AtomicLimits* ALD/E database [1,2] has become a pivotal crowd-sourcing, open-domain platform for the atomic scale processing community by providing a comprehensive overview of processes reported in the scientific literature. In its current form it compiles basic information, such as deposited materials, reactants used, and relevant references. For ALE processes, it also reports whether the process is isotropic or anisotropic. Despite its extensive repository, vast amounts of (unstructured) data remain unexploited limiting the database’s potential for advancing material science discovery in ALD and ALE processes. Artificial Intelligence (AI), and particularly Large Language Models (LLMs), present transformative opportunities to bridge this gap. LLMs can solve tasks beyond their initial training scope, excelling at understanding, generating and processing text. These capabilities make them highly suitable for recognizing patterns, synthesizing knowledge and producing human-like outputs. Thus, they provide a powerful and scalable alternative for structured data extraction from the vast literature on ALD/E. [3]

In this work, we present the current state of the *AtomicLimits* ALD/E database and demonstrate how LLMs can be employed to enhance it with additional structured data. We highlight a case study where LLMs extract valuable ALD-related information from unstructured literature text using a structured framework. This framework is iteratively generated by LLMs and refined through ALD-expert feedback. The final structure, combined with ALD literature, enables knowledge extraction using LLMs. The extracted data is then represented as a knowledge graph, specifically using the Open Research Knowledge Graph (ORKG). [4] This approach not only enables the creation of extensive datasets for ALD and ALE research but also lays the foundation for a new era of data-driven material science discovery in the atomic-scale processing domain.

[1] DOI: 10.6100/alddatabase

[2] DOI: 10.6100/aledatabase

[3] Schilling-Wilhelmi, M. *et al.* From text to insight: large language models for chemical data extraction. *Chem. Soc. Rev.* 10.1039.D4CS00913D (2025) doi:10.1039/D4CS00913D

[4] Stocker, M. et al. FAIR scientific information with the Open Research Knowledge Graph. FAIR Connect 1, 19–21 (2023).

2:00pm **ALDALE-MoA-3 ALD Student Award Finalist Talk: Influence of Hydrocarbon Chain Length in Phenyl(Alkyl)trimethoxysilane Inhibitors on AS-ALD Selectivity: Comparison of Adsorption Mechanisms in Gas-phase and Liquid-phase***, **Hae Lin Yang**, Minchan Kim,* Hanyang University, Korea*; Eun Chong Cho,* Sungkyunkwan University, Korea*; Seunghwan Lee, Beomseok Kim, Changhwa Jung, Hanjin Lim,* Samsung Electronics Co., Inc., Republic of Korea*; Jung-Hoon Lee, Youngkwon Kim,* Korea Research Institute of Chemical Technology (KRICT), Republic of Korea*; Jin-Seong Park,* Hanyang University, Korea

Area-selective atomic layer deposition (AS-ALD) has become a critical technique for precise material fabrication, particularly in complex nanoarchitectures. Achieving high selectivity in AS-ALD requires the strategic use of effective small molecular inhibitors (SMIs) to prevent undesired growth on non-target surfaces[1,2]. In this study, we investigated how variations in the hydrocarbon chain length (n = 1–6) of Phenyl(Alkyl)trimethoxysilane inhibitors influence their adsorption and precursor-blocking performance during the AS-ALD of vanadium dioxide (VO_2). To analyze the adsorption characteristics, we employed two different adsorption methods: (1) liquid-phase adsorption via spin-coating and (2) gas-phase adsorption by supplying vaporized SMIs into the chamber. The SMI-coated SiO_2 surfaces formed through liquid-phase adsorption exhibited highly similar characteristics, with water contact angle (WCA) variations within 5° among different SMIs. Furthermore, after 100 ALD cycles of VO_2 deposition, all samples demonstrated high selectivity above 90%, regardless of the SMI used. However, in the gas-phase adsorption process, significant differences were observed depending on the chain length. The WCA varied by more than 20°, indicating substantial surface property differences. Additionally, after 100 cycles, selectivity ranged from a maximum of 89.9% (n = 2) to a minimum of 4.5% (n = 6), revealing a significant disparity in selectivity. To elucidate these differences, we employed density functional theory calculations and random sequential adsorption simulations[3]. Experimental validation was conducted using WCA, X-ray photoelectron spectroscopy, X-ray fluorescence, and scanning electron microscopy. These findings highlight the importance of appropriate molecular characteristics and structures depending on the adsorption method and underscore the necessity of tailored molecular design to achieve optimal AS-ALD performance.

2:15pm **ALDALE-MoA-4 ALD Student Award Finalist Talk: Diffusion Behavior Study for Vapor Phase Infiltration Using Quartz Crystal Microgravimetry and its Application in Energy Storage Materials***, **Rongliang Shang**, Jin Xie,* ShanghaiTech University, China

Vapor phase infiltration (VPI), derived from atomic layer deposition (ALD), facilitates the growth of inorganic components into organic polymers, emerging as an effective technique for fabricating organic-inorganic hybrid materials. VPI involves prolonged exposure of gas-phase precursors (typically several minutes to hours), allowing their diffusion into porous substrates. However, the complexity of diffusion behavior during the VPI process presents challenges in studying diffusion kinetics, particularly for highly reactive precursor-polymer systems such as trimethylaluminum (TMA) and poly(ethylene oxide) (PEO). This limitation hinders the study of VPI kinetics in PEO systems.

In this work, we investigate the VPI process of TMA in PEO using in-situ quartz crystal microgravimetry (QCM), which enables measurement of diffusion behavior and kinetics with high temporal resolution (0.1 s). PEO film was spin-coated on the QCM crystal, and then sent to VPI. In a typical mono-pulse VPI process, TMA was pulsed into a reaction chamber, followed by prolonged exposure and then nitrogen purging. The mass of the film was recorded during the whole process.

QCM measurements indicated that the VPI process consists of two main regions: a rapid diffusion process, corresponding to the initial penetration of the precursor into the film, followed by a slower relaxation process, attributed to the ongoing chemical reaction. The equivalent diffusion coefficient (D_e) was estimated to be on the order of 10^{-9} cm^2/s at 100 °C. In multi-pulse VPI, the mass gain per cycle decreased with successive pulses, attributed to the reduction in available reactive sites and the reduction of diffusion coefficient. When water was introduced as a co-reactant in multi-cycle VPI processes, the formation of Al–O clusters further inhibited diffusion, shifting the process from reaction-limited to diffusion-limited behavior. Finally, VPI-modified PEO was used as solid polymer electrolytes (SPEs) for lithium metal batteries as a proof-of-concept, demonstrating enhanced mechanical properties, suppressed dendrite growth, and higher Coulombic efficiency (89.5% after 50 cycles vs. 60% for unmodified PEO).

This study provides a novel understanding of the VPI process for TMA in PEO, highlights the critical role of diffusion and reaction kinetics in determining material properties, and ultimately contributes to the development of next-generation materials for energy storage and beyond.

2:30pm **ALDALE-MoA-5 ALE Student Award Finalist Talk: Lateral Etching of 2D MoS_2 Crystalline Layers Using Sequential Ozone and Thionyl Chloride Exposures***, **Janine Sempel,*** University of Colorado at Boulder*; Taewook Nam,* Sejong University, Republic of Korea*; Tianyi Zhang, Jing Kong,* Massachusetts Institute of Technology*; Steven George,* University of Colorado at Boulder

Atomic layer controlled etching of 2D MoS_2 crystalline layers is important for the fabrication of MoS_2 channel transistors. Individual 2D MoS_2 layers must be removed to reduce MoS_2 multilayers to MoS_2 bilayers and monolayers. The removal of individual MoS_2 layers requires lateral etching in the 2D plane of each MoS_2 layer. In this study, the lateral etching of 2D MoS_2 crystalline layers was demonstrated using two sequential reactions for surface modification and volatile release of the modified layer. O_3 (ozone) was used for MoS_2 oxidation to MoO_3 and $SOCl_2$ (thionyl chloride) was used for the volatilization of MoO_3 as MoO_2Cl_2.

The studies were performed using high quality MoS_2 bilayers on silicon coupons. The etching of the 2D MoS_2 was examined by optical microscopy and atomic force microscopy (AFM) at 175°C. A decrease in the optical contrast of the MoS_2 bilayer during MoS_2 etching was visualized by optical microscopy. Etching was observed by AFM as triangular etch pits inside the 2D MoS_2 crystalline domains and removal of MoS_2 from the edge of 2D MoS_2 crystalline domains. The triangular etch pits and the gap between crystalline domains grew progressively versus number of etching cycles (Figure 1). The depth of the triangular etch pits was equal to one MoS_2 bilayer.

The AFM images were consistent with lateral etching at step edges of the 2D MoS_2 crystalline layer. The lateral etching could be quantified by going back to the same location after various numbers of etching cycles. The enlargement of the triangular etch pits versus number of etching cycles was “inside-out” lateral etching (Figure 2A). The loss of MoS_2 from the edge of 2D MoS_2 crystalline domains versus number of etching cycles was “outside-in” etching (Figure 2B). These AFM collocation experiments determined that “inside-out” and “outside-in” 2D lateral etching rates were equivalent at ~5 Å per etching cycle at 175°C (Figure 3). This lateral etch rate represents ~2 MoS_2 units removed at the step edge of the 2D MoS_2 layer.

2:45pm **ALDALE-MoA-6 ALE Student Award Finalist Talk: A Sustainable and Precise Solution to IGZO Etch Residual Challenges Using Transient-Assisted Processing (TAP)***, **Atefeh Fathzadeh,*** KU Leuven and Imec, Belgium*; Philippe Bezard, Thierry Conard, Frank Holsteyns,* IMEC Belgium*; Stefan De Gendt,* KU Leuven and Imec, Belgium

The increasing demand for memory-intensive applications, such as AI, necessitates significant advancements in DRAM performance and power efficiency. A promising approach to address these challenges is the 2T0C (2 Transistors, 0 Capacitor) cell architecture, where the write transistor relies on a semiconductive oxide channel to minimize leakage, ensuring long retention times and low power consumption. Among candidate materials, $InGaZnO_4$ (IGZO) stands out due to its superior electrical properties. However, the etching process used for IGZO patterning can generate residues contributing to leakage. Superior device performance has been demonstrated using conventional plasma etching over ion-beam etching due to its ability to reduce these unwanted residues[1]. Among possible etchants, CH_4–based plasma allows the formation of volatile by-products such as $(CH_3)_3In$, $(CH_3)_3Ga$, and $(CH_3)_2Zn$. However, Ga_xO_y is slower to be etched than In_xO_y and ZnO. As a result the plasma-exposed surfaces become Ga-rich, leading to residue formations. Even Atomic Layer Etching (ALE), despite its excellent control capabilities, still exhibits Ga residues for CH_4/Ar mixture. Introducing Cl_2 to the process enables the formation of volatile $GaCl_3$, which enhances Ga removal. However, non-volatile $InCl_3$ and $ZnCl_2$ are also produced, requiring precise chlorine dosing to minimize residues. This presents a challenge even for ALE unless the different gases are separated into dedicated cycles- an approach that dramatically increases processing time, cost, and environmental impact.

Transient-Assisted Plasma Processing (TAP) provides a breakthrough solution to these challenges. TAP operates in controlled cycles, where reactive species are briefly injected into a carrier gas, creating a transient decrease in their concentration[2]. The plasma is ignited at the most convenient timing to ensure optimal ion over neutral ratio (similar to ALE) but also to control the nature of the formed species. This control over the dosage and timing of reactive gases enables balanced etching of different elements, ensuring uniform material removal, preserving surface composition and minimizing residues. Notably, TAP’s benefits extend beyond IGZO, making it applicable to a wide range of materials and semiconductor applications[3–6].

This paper presents a scalable and sustainable IGZO patterning process that minimizes residues and gas consumption while achieving near-vertical profiles at 28 nm pitch—the tightest pitch enabled by 0.33 NA EUV lithography to ensure scalability, as illustrated in Figure 1. The demonstrated approach enables production-compatible processing times, with a linear control of the etched depth through the number of cycles,supporting the advancement of energy-efficient memory technologies.

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