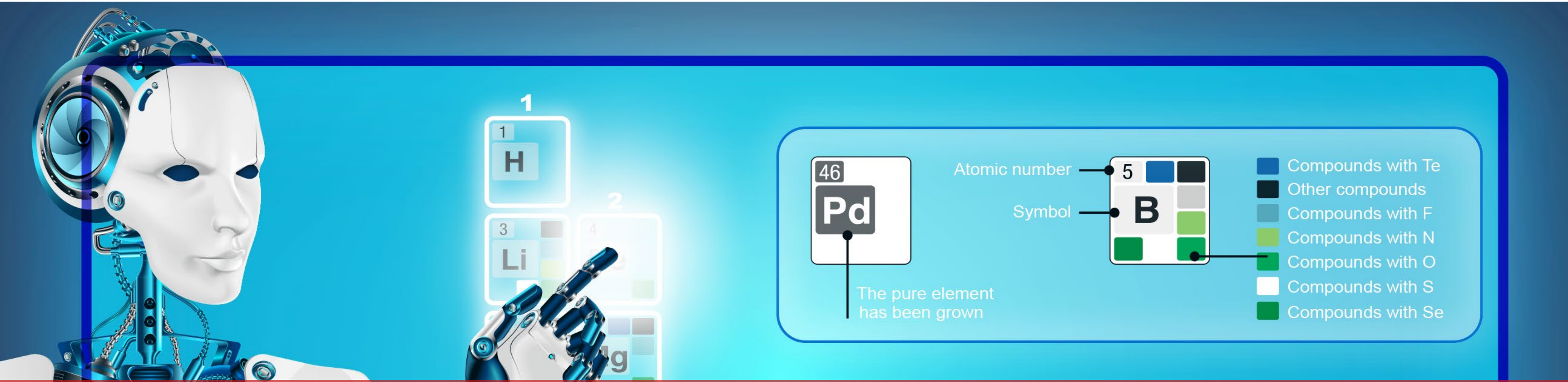


# Current and Future Perspectives on Atomic Layer Deposition

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Department of Applied Physics

AtomicLimits

TU/e EINDHOVEN UNIVERSITY OF TECHNOLOGY

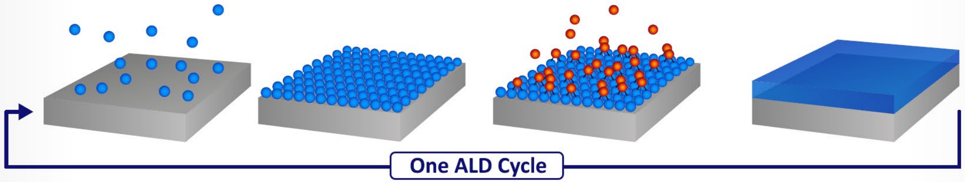
AVS ALD/ALE 2026

June 28-July 1, 2026 | Tampa, Florida

# Atomic scale processing toolbox

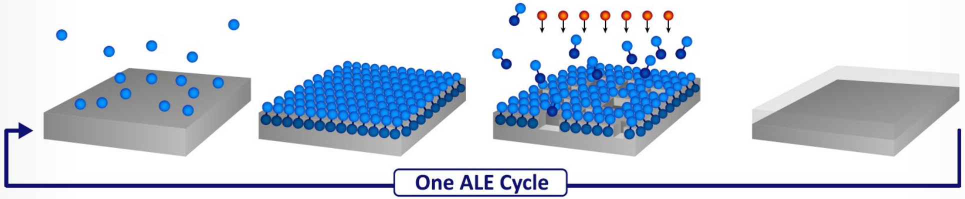
## Providing complementary routes to build, remove, and pattern materials atom by atom

**ALD**
**Atomic Layer Deposition**



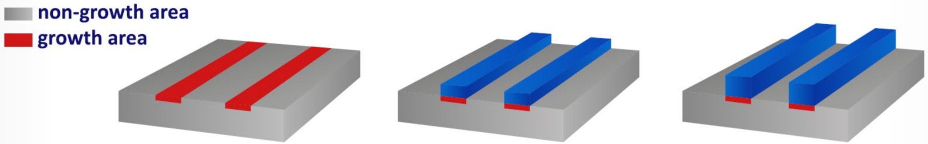

- Thermal ALD
- Plasma-enhanced ALD

**ALE**
**Atomic Layer Etching**




| | PLASMA | THERMAL |
|---|---|---|
| Isotropic | ✓ | ✓ |
| Anisotropic | ✓ | ✗ |

**ASD**
**Area-Selective Deposition**




- Intrinsic
- Activation
- Deactivation
  - Self-assembled monolayers
  - Small molecule inhibitors



Image: Atomic Limits – TU/e (2025)

# Outline

1. **Quick introduction to ALD**
2. ALD process development
3. Towards AI-enabled ALD
4. Challenges in frontier materials

# Recent review paper on ALD – Nature Reviews Methods Primers

nature reviews methods primers

https://doi.org/10.1038/s43586-025-00435-6

Primer

Check for updates

# Atomic layer deposition

Erwin Kessels[1], Anjana Devi[2,3,4], Jin-Seong Park[5], Mikko Ritala[6], Angel Yanguas-Gil[7] & Claudia Wiemer[8]

**Abstract**

Atomic layer deposition (ALD) is a surface-controlled chemical vapour deposition method, in which materials are prepared one atomic layer at the time. With ALD, film thickness can be controlled very precisely, and it allows the user to cover large areas and surfaces with a complex three-dimensional structure uniformly and conformally. ALD is used for the deposition of high-quality thin films and nanostructures, as well as for surface functionalization and interface engineering in a wide range of applications, both from a research and development perspective, as well as for high-volume manufacturing. This Primer

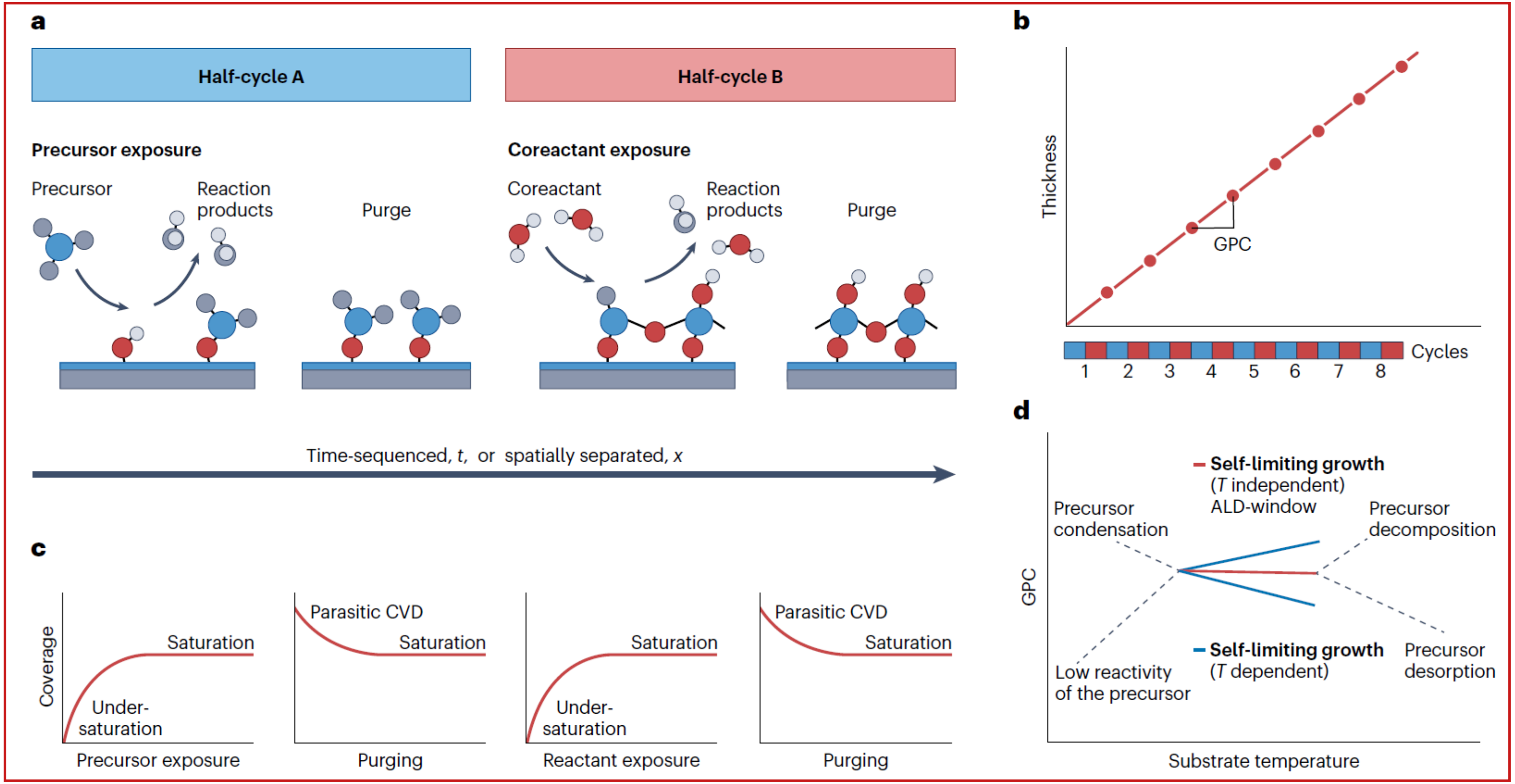
**Sections**





Kessels *et al.*, Nat. Rev. Methods Primers 5, 66 (2025)

# ALD in one slide: self-limiting cycles enable precise growth

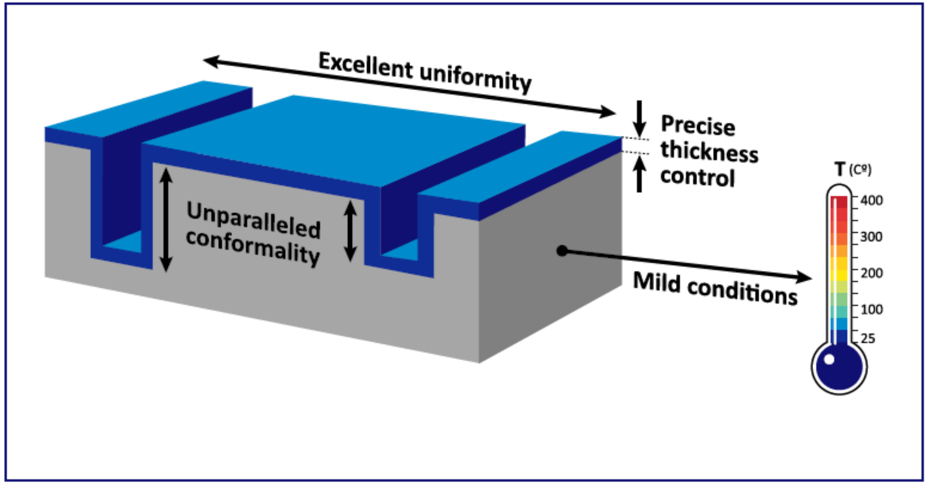


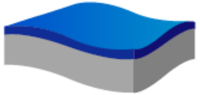



Kessels *et al.*, Nat. Rev. Methods Primers 5, 66 (2025)

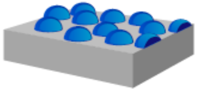
# ALD: enabling nanotechnology through precision and conformality

## ALD enables controlled materials synthesis on complex surfaces and nanostructures

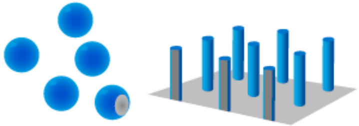


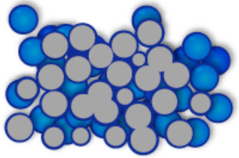
*Flexible foils*



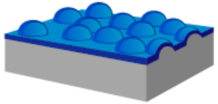
*Size-controlled nanoparticles*



*Conformal coating of nanoparticles and nanowires*

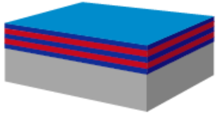


*Conformal penetration of porous substrates*

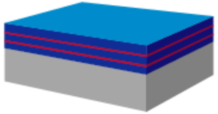


*Thin film encapsulation*

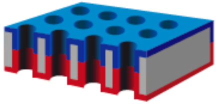
*Selective deposition on prepatterned surfaces*



*Nanolaminates*



*Delta-doping*

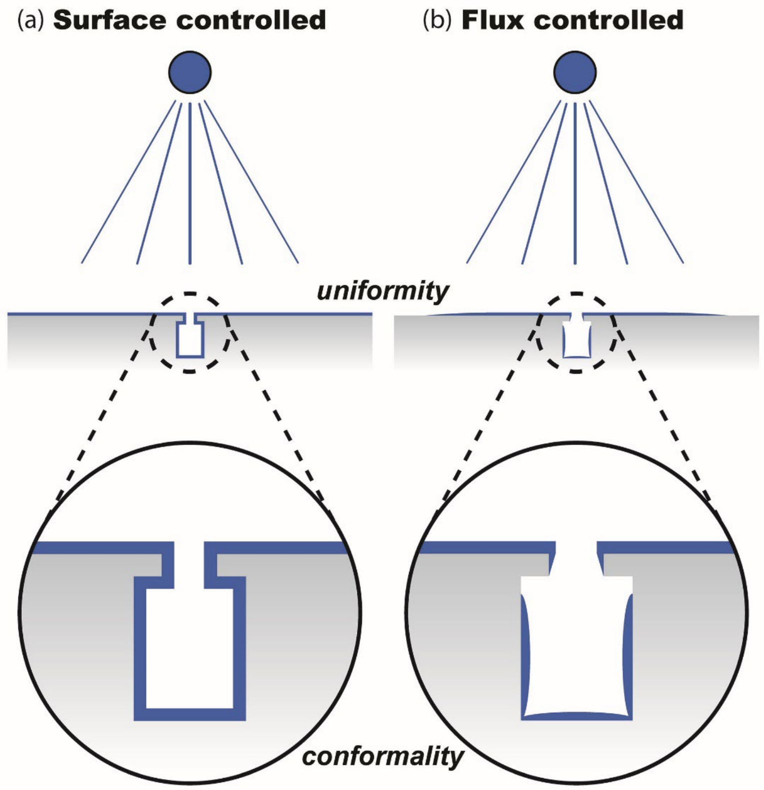


*Nanoscale engineering*

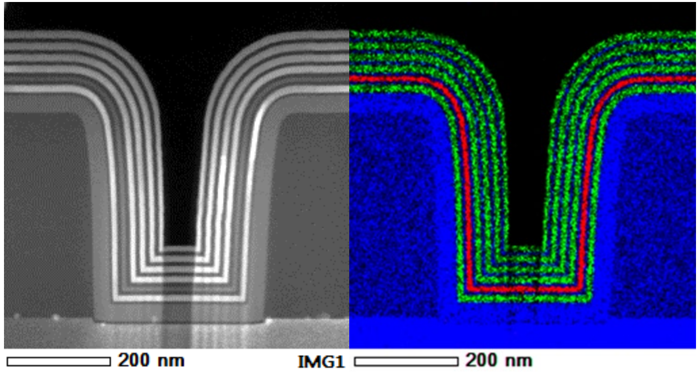


Image: Atomic Limits – TU/e (2021)

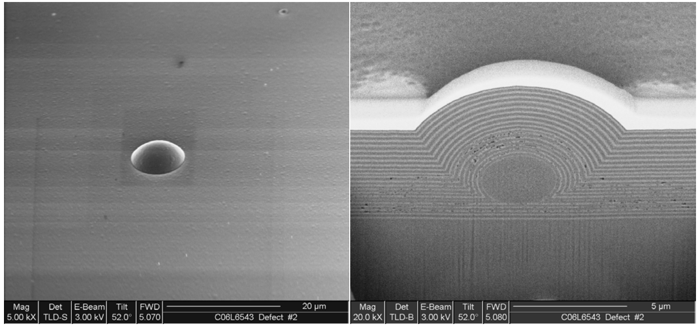
# ALD conformality comes from surface-controlled growth




Film stack in etched trench (Arts *et al.*)



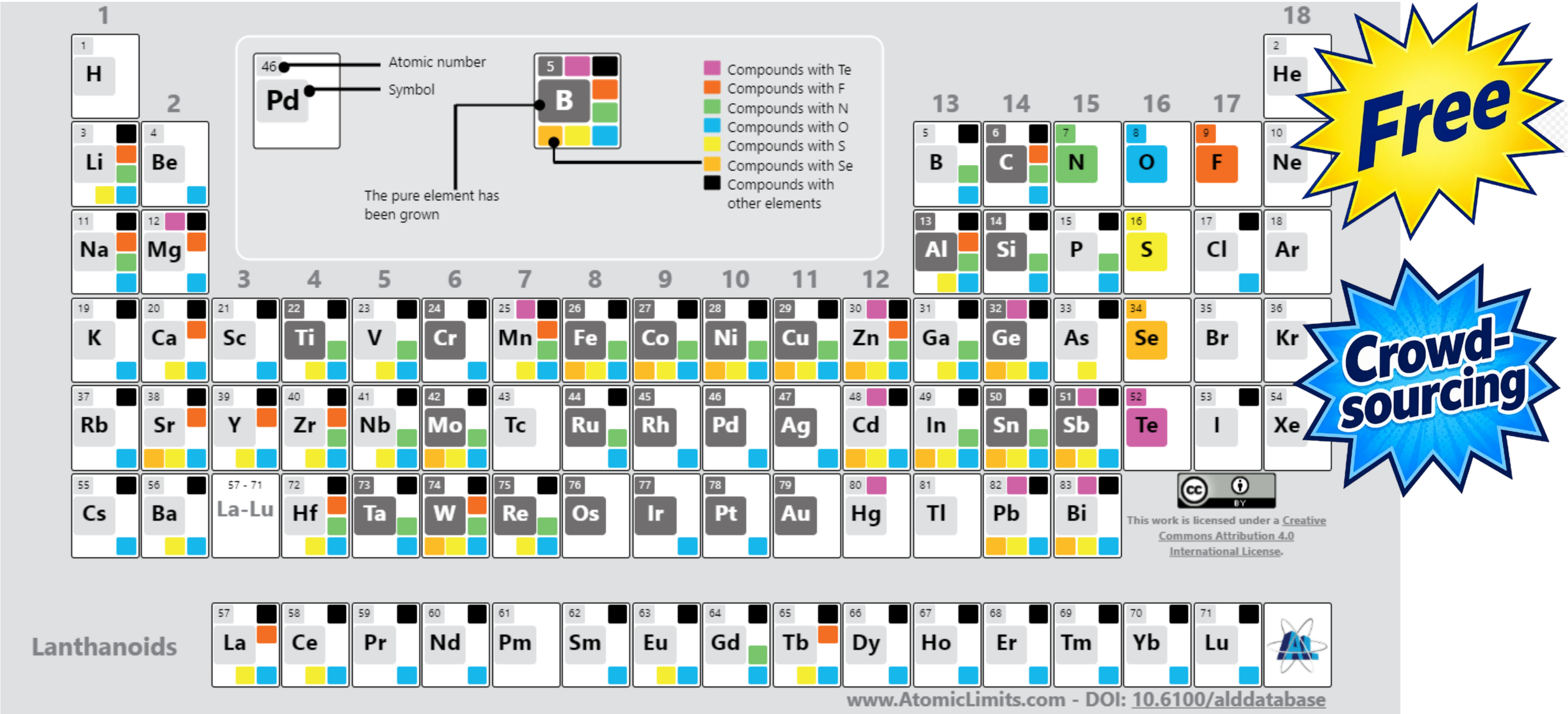

Encapsulated particle on wafer (Jim Wang)





Knoops *et al.*, Handbook of Crystal Growth Vol III, 2nd ed. (2015)
Arts *et al.*, Chem. Mater. 33, 5002 (2021)
Courtesy of Jim Wang (NanoNuvo Corporation, USA – ALD 2008 Bruges)

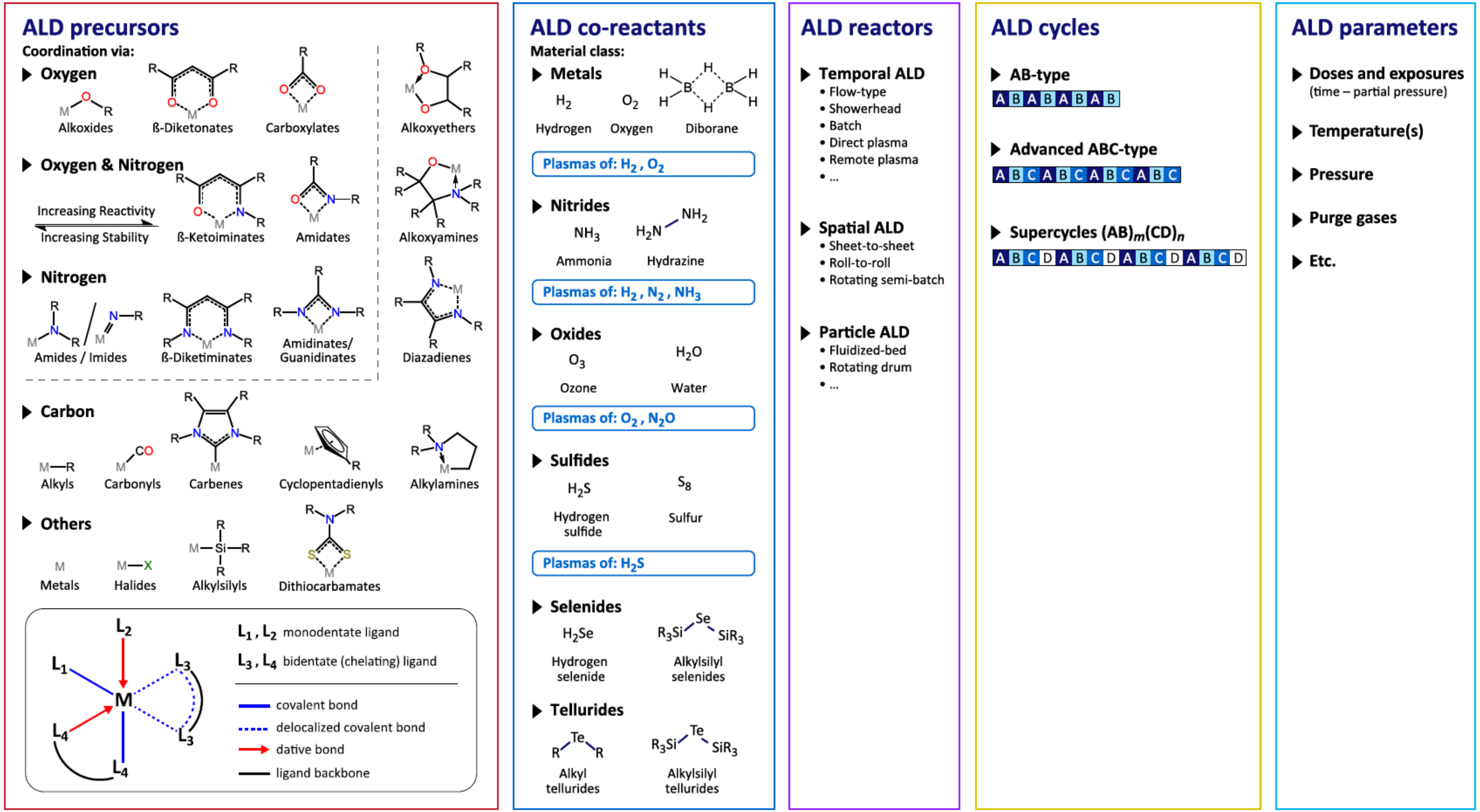
# AtomicLimits ALD Database: all published processes with refs.






**ALD database DOI**: https://doi.org/10.6100/alddatabase
Content management: TU/e team including alumni

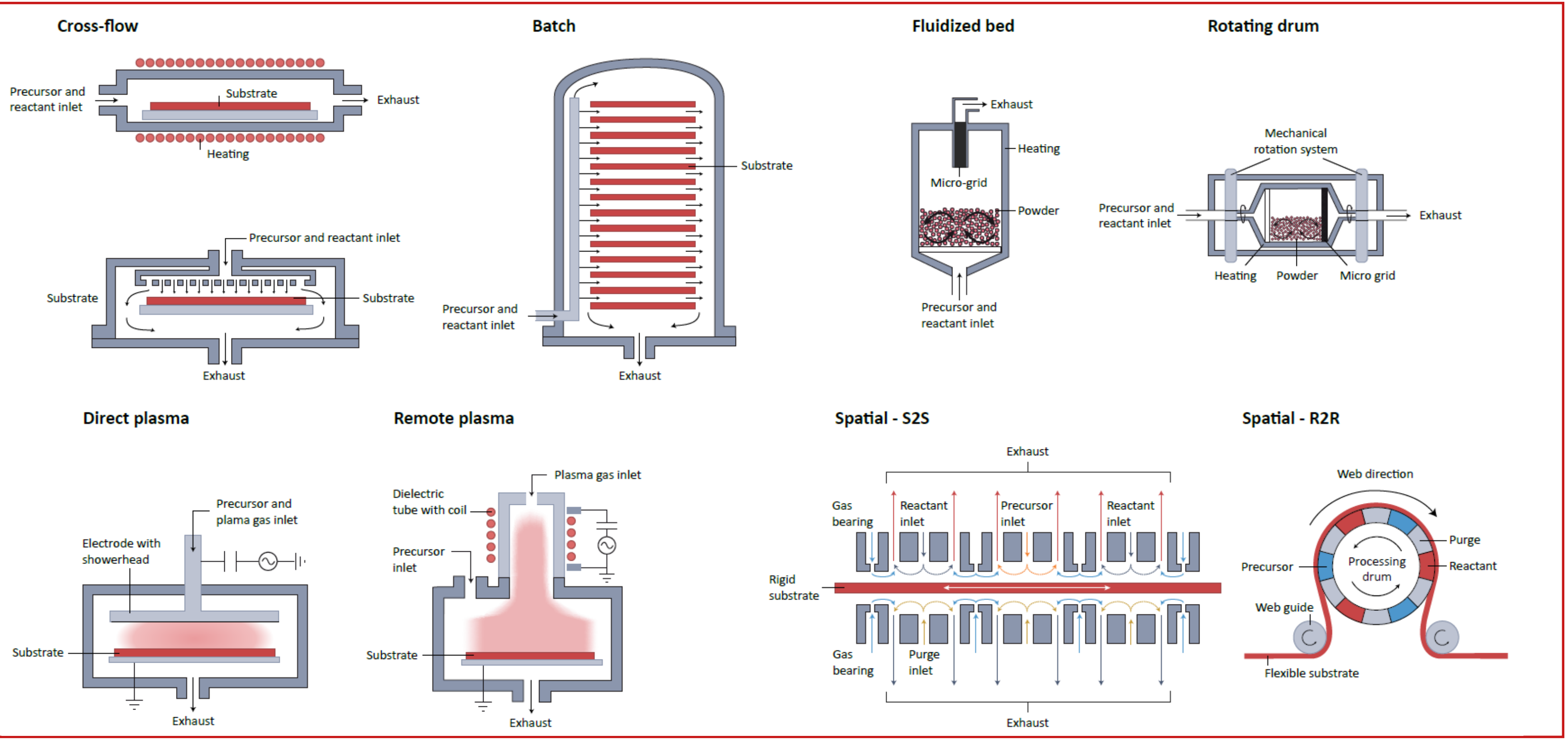
# ALD is simple in principle, but rich in choices






Kessels *et al.*, Nat. Rev. Methods Primers 5, 66 (2025)

# One ALD principle, many reactor architectures

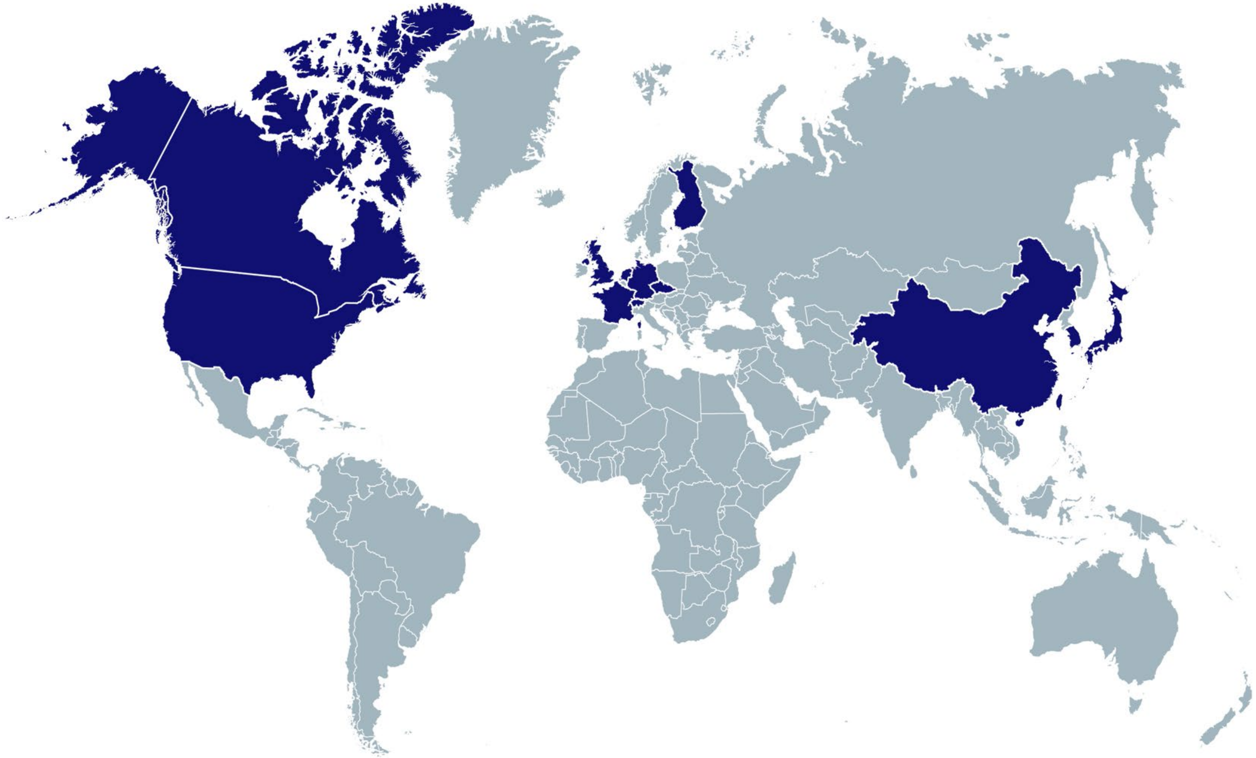





Kessels *et al.*, Nat. Rev. Methods Primers 5, 66 (2025)

# The global equipment landscape for ALD – 60+ companies

Animation

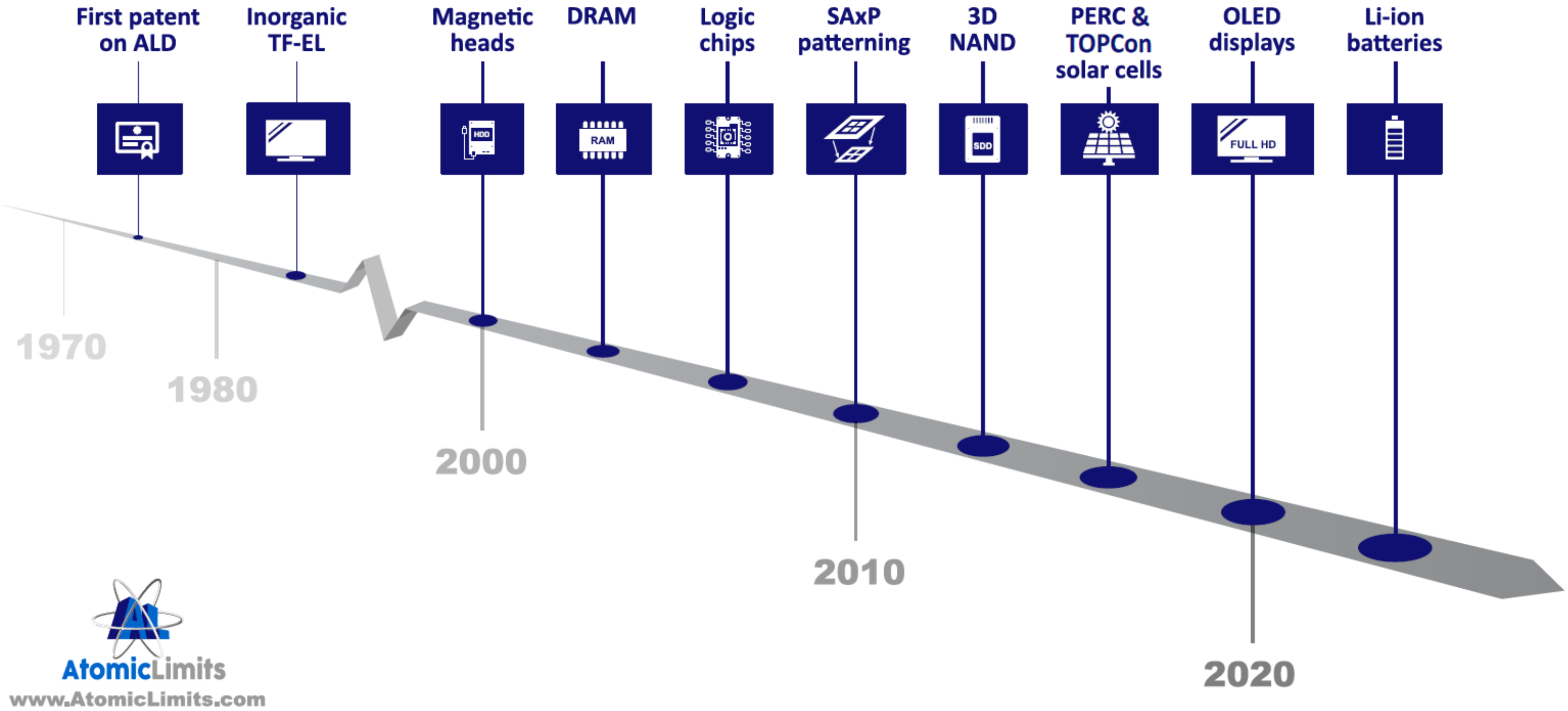




Animation: Atomic Limits – TU/e (2021)

# From invention to industrial impact: 50+ years of ALD

## Applications of ALD in high-volume manufacturing (HVM)






See blog about **ALD timeline** on AtomicLimits.com (2025)

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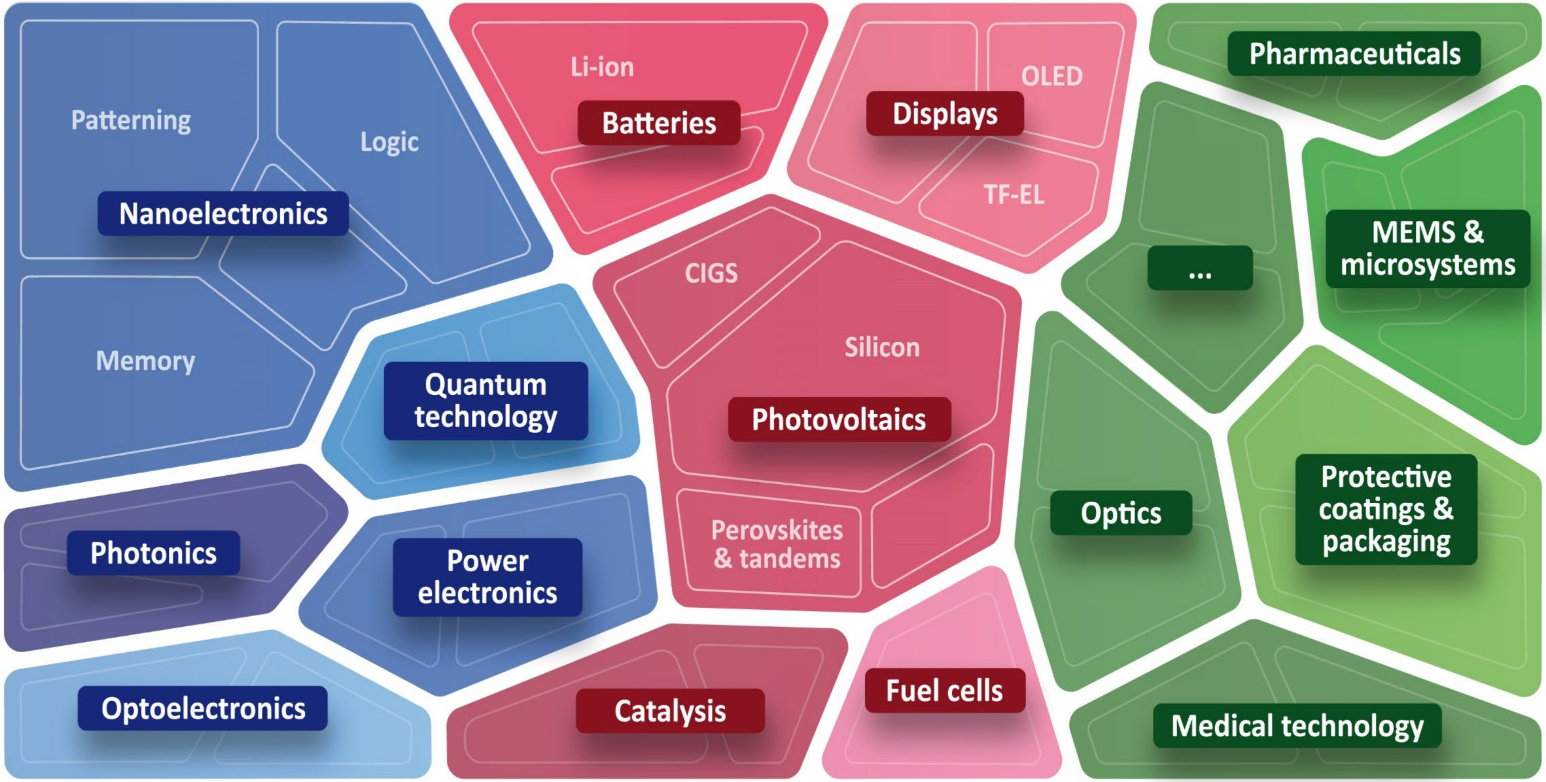
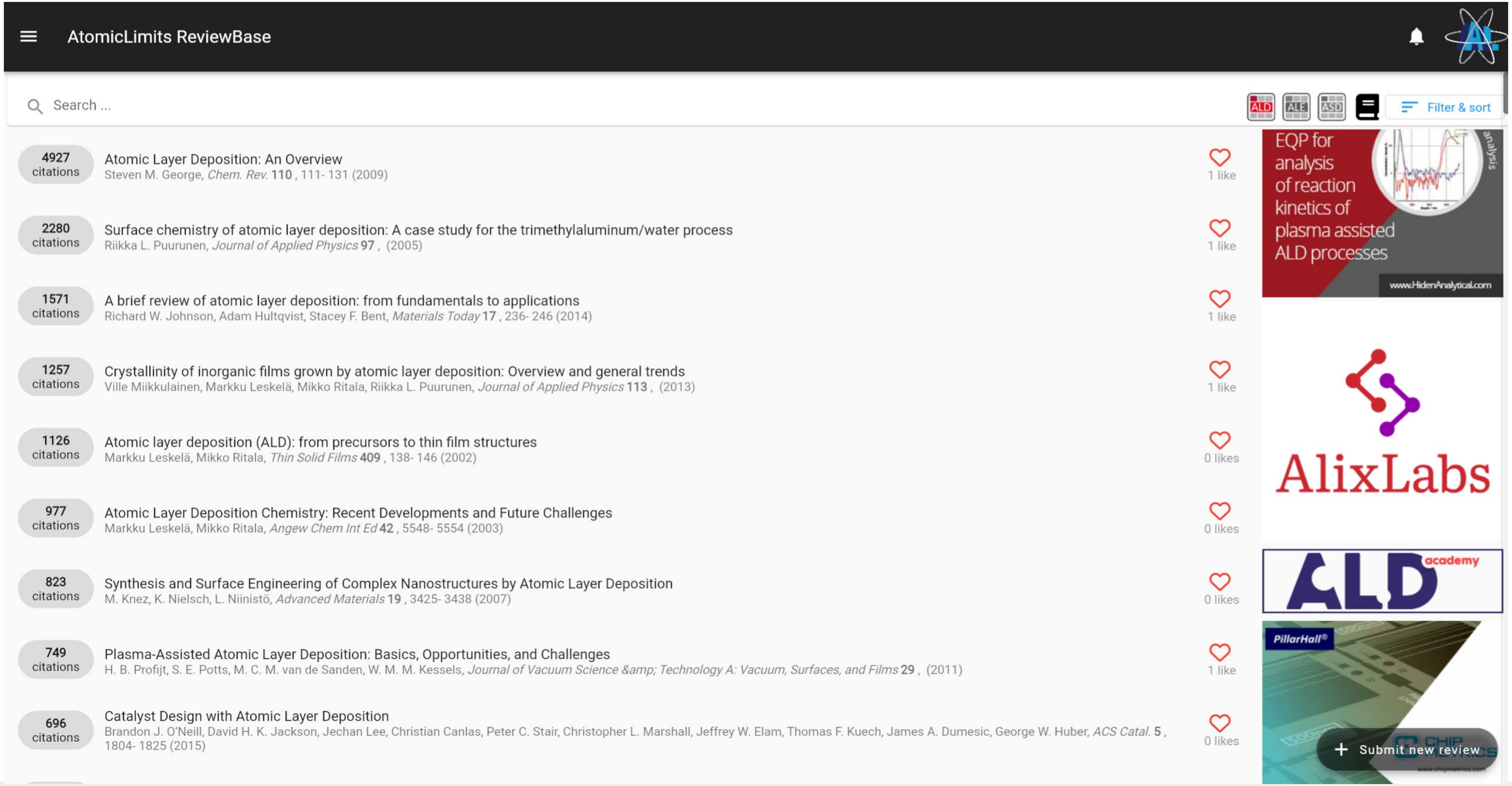
# ALD enables a broad landscape of applications






See blog about **ALD applications** on AtomicLimits.com (2025)
(Image design after image by Jonas Sundqvist at https://www.strem.com)

# AtomicLimits ReviewBase: Browse all review papers






See **ReviewBase** on AtomicLimits.com

# Outline

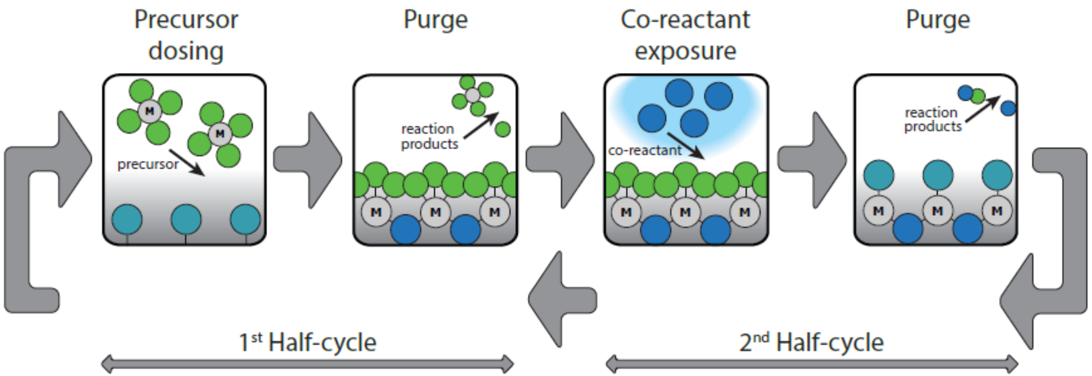
1. Quick introduction to ALD
2. **ALD process development**
3. Towards AI-enabled ALD
4. Challenges in frontier materials

# Blog post & document – AtomicLimits.com

## Atomic Layer Deposition Process Development

### 10 steps to successfully develop, optimize and characterize ALD recipes

Atomic layer deposition (ALD) is a surface-controlled thin film deposition technique that can enable ultimate control over the film thickness, uniformity on large-area substrates and conformality on 3D (nano)structures.[1–5] Each ALD cycle consists at least two half-cycles (but can be more complex), containing a precursor dose step and a co-reactant exposure step, separated by purge or pump steps (see Figure 1). Ideally the same amount of material is deposited in each cycle, due to the self-limiting nature of the reactions of the precursor and co-reactant with the surface groups on the substrate. By carrying out a certain number of ALD cycles, the targeted film thickness can be obtained.

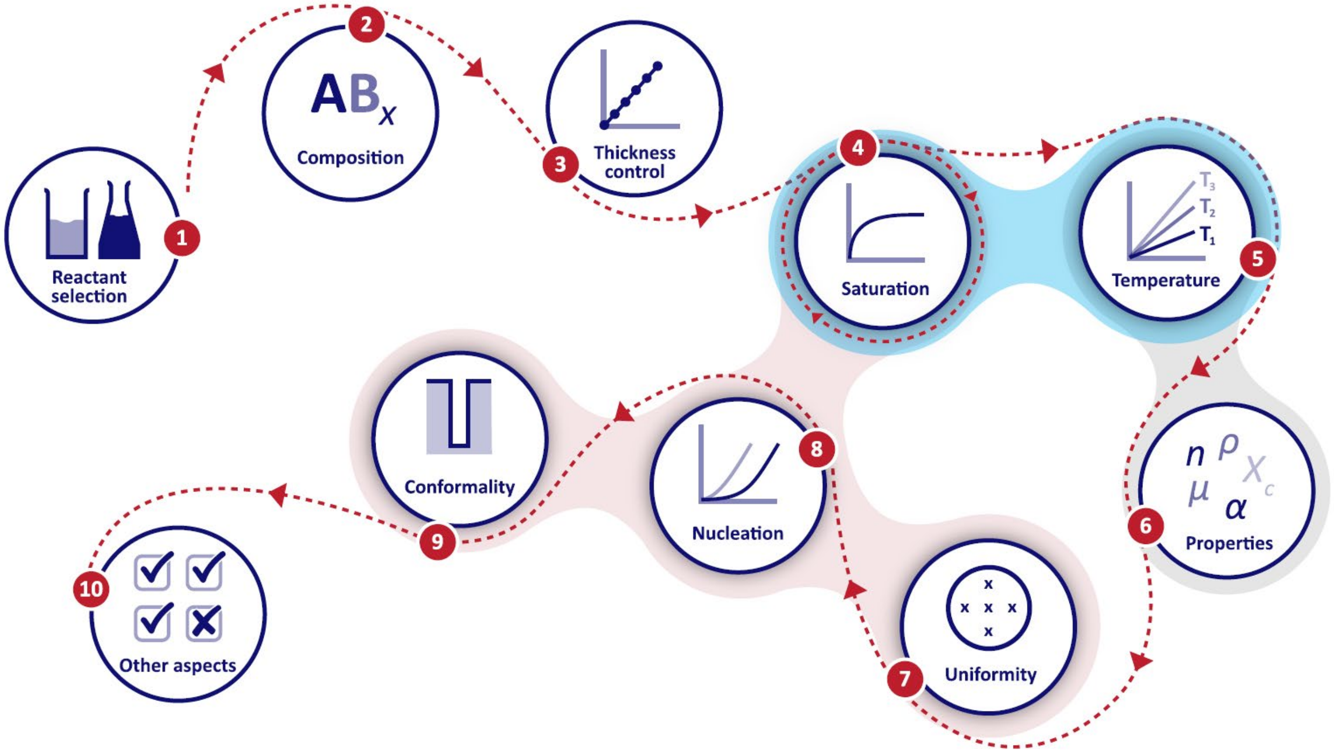





See blog about **ALD process development** on AtomicLimits.com

# ALD process development: a structured 10-step workflow

## Steps ordered roughly chronologically, with iteration often needed



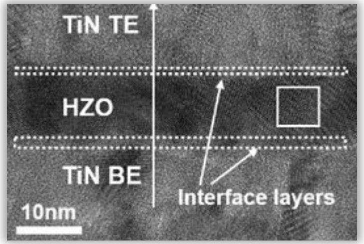


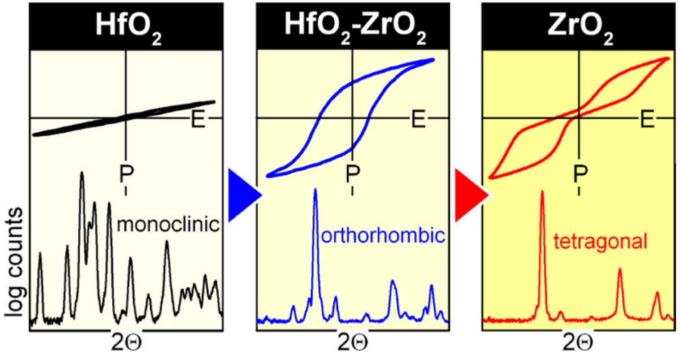

See blog about **ALD process development** on AtomicLimits.com

# ALD process development for $Hf_xZr_{1-x}O_2$ – Illustrative example

## An illustrative case study to demonstrate the process-development workflow

$Hf_xZr_{1-x}O_2$ (HZO) with ferroelectricity

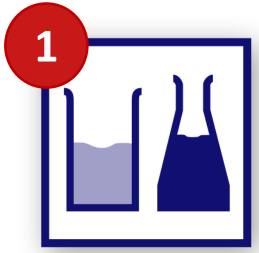


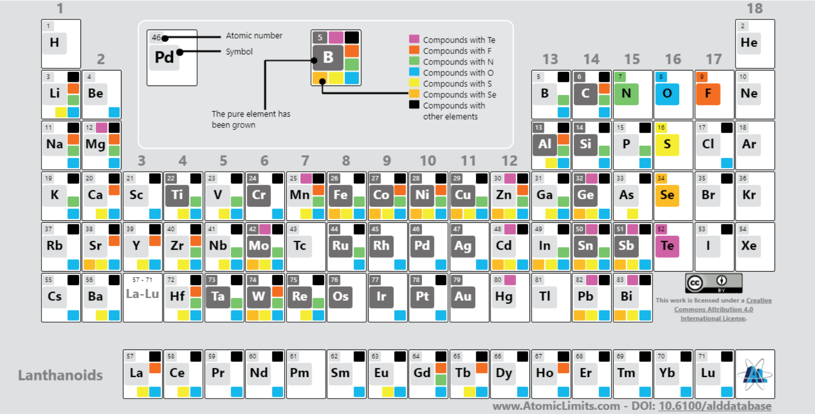

| ALD development step | $HfO_2$ | $ZrO_2$ | $Hf_{1-x}Zr_xO_2$ |
|---|---|---|---|
| 1 Reactant selection | | | |
| 2 Composition | | | |
| 3 Thickness control | | | |
| 4 Saturation | | | |
| 5 Properties | | | |
| 6 Temperature | | | |
| 7 Uniformity | | | |
| 8 Conformality | | | |
| 9 Nucleation | | | |
| 10 Other aspects | | | |

Müller *et al.*, Nano Lett. 12, 4318 (2022)
Schröder *et al.*, Nature Reviews Materials 7, 653 (2022)

# Step 1: Reactant selection - Best choice?



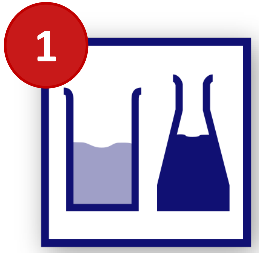

- Right precursor/co-reactant pair is critical
- Must deliver the required elements to the surface
- Need to balance volatility, stability, and reactivity
- Must fit reactor and process constraints
- Literature (ALD database) provides the first shortlist

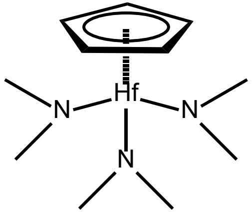


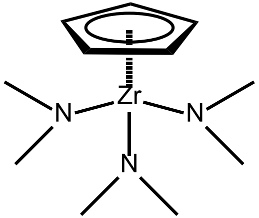



See blog about **ALD process development** on AtomicLimits.com

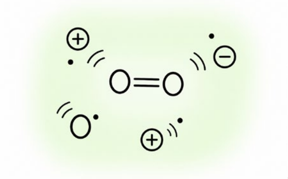
# Step 1: Reactant selection - $Hf_xZr_{1-x}O_2$ case



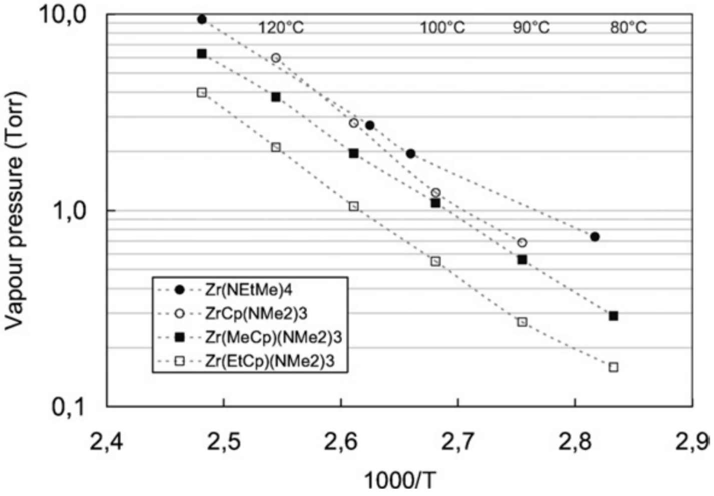



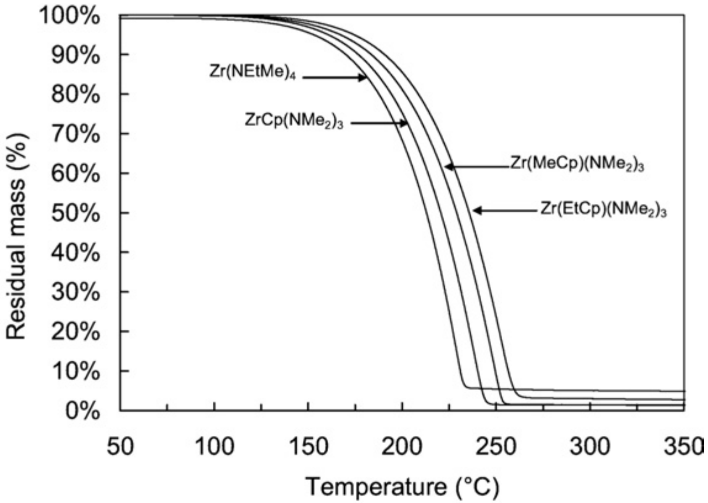




## Vapor pressure for Zr precursors




## Thermogravimetric analysis (TGA)



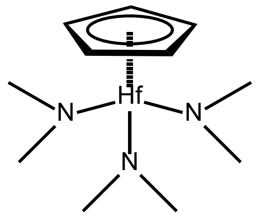


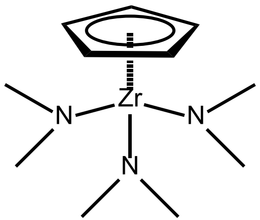

Niinistö *et al*. J. Mater. Chem. 18, 5243 (2008)

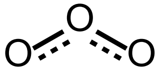
# Step 2: Composition - Desired material?

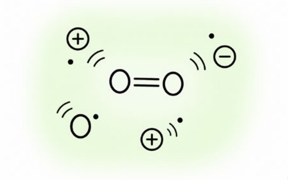
- Check the material after an early test run
- Start with fast, accessible diagnostics
  - Refractive index by ellipsometry
  - Composition by XPS
  - Conductivity for metallic films, etc.
  - …
- If results strongly deviate, revisit reactant selection

# Step 2: Composition - $Hf_xZr_{1-x}O_2$ case

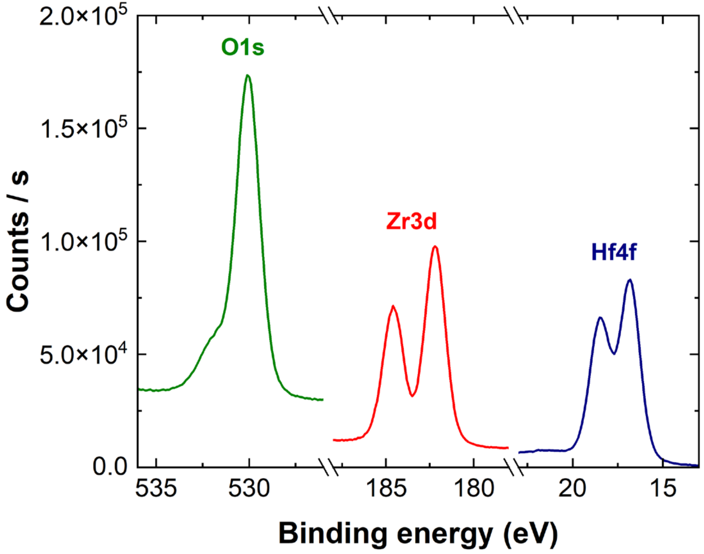


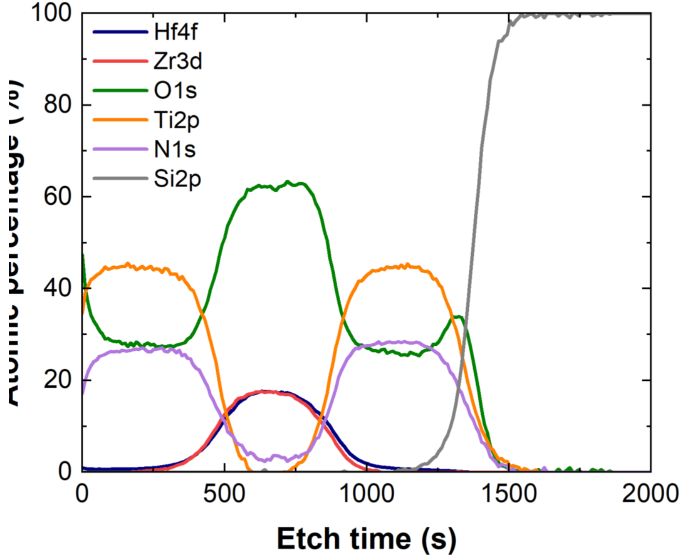




## XPS scans



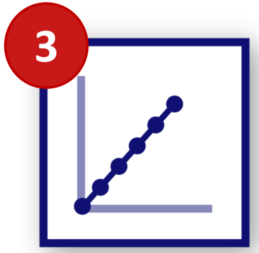

## XPS depth profile (for $TiN/Hf_xZr_{1-x}O_2/TiN$ stack)





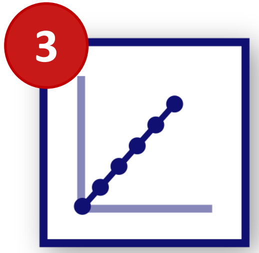

Van der Heijden *et al.*, to be published (2026)

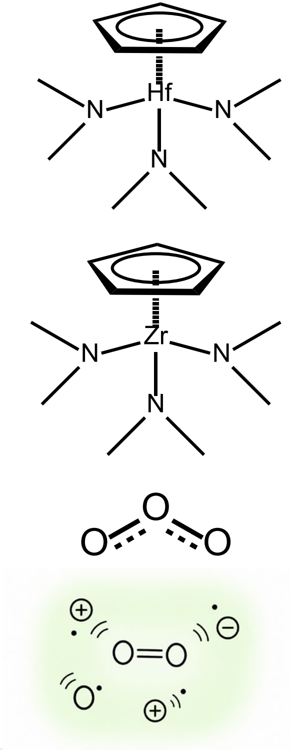
# Step 3: Thickness control – linear in #cycles?



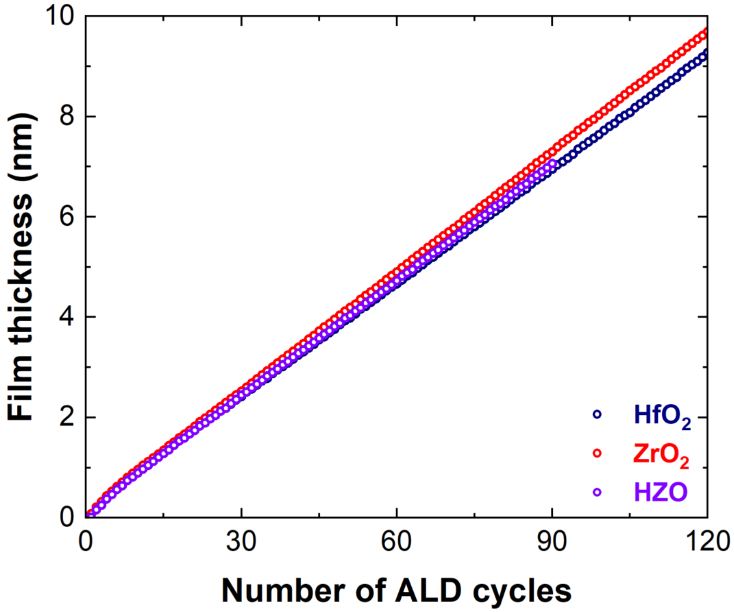

- Verify thickness scales with cycle number
  - *In situ*: follow thickness during growth
  - *Ex situ*: prepare a thickness series
- Linear growth is necessary, but not proof of ALD
- Compare GPC with literature values, should match!
- Pay attention to the substrate temperature

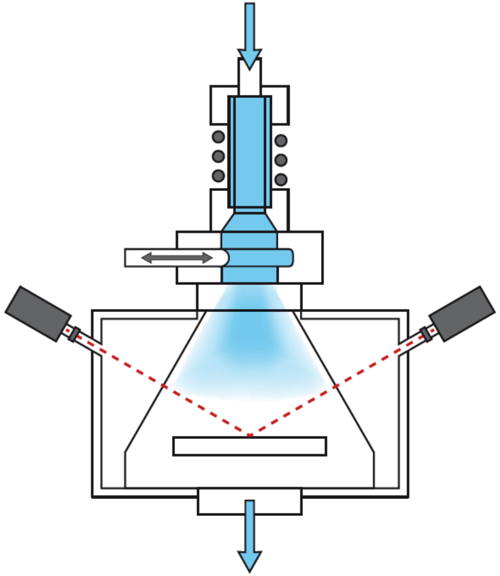
# Step 3: Thickness control – $Hf_xZr_{1-x}O_2$ case




Thickness monitoring by *in situ* ellipsometry



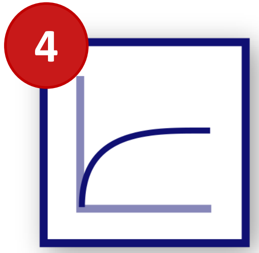





Van der Heijden *et al.*, to be published (2026)

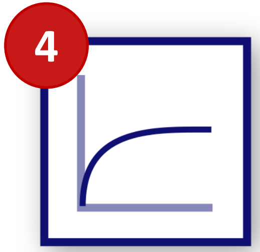
TU/e

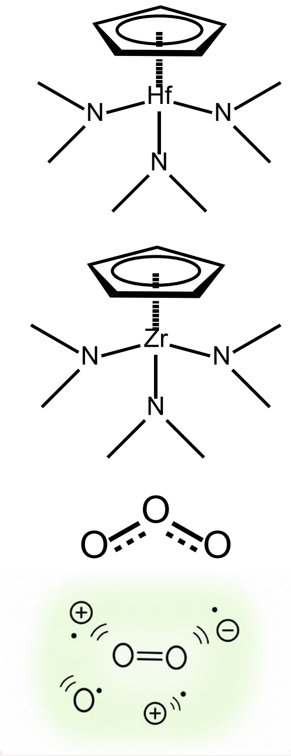
# Step 4: Saturation – Self-limiting growth?

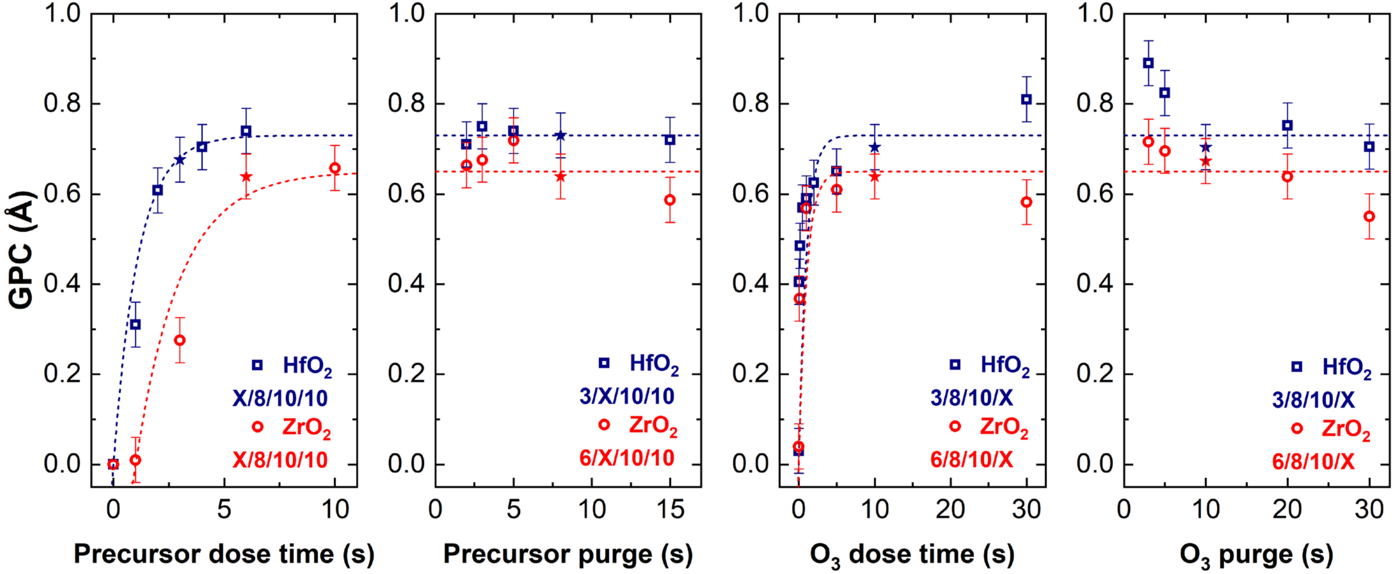



- Start with sufficiently long dosing and purge times
- Vary one timing parameter at a time
- Look for a GPC plateau at long exposures
- Check zero-dose cases: no growth expected
- Confirm saturation for both half-cycles
- Repeat under “extreme” conditions if needed

# Step 4: Saturation – $Hf_xZr_{1-x}O_2$ case



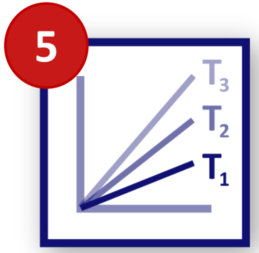



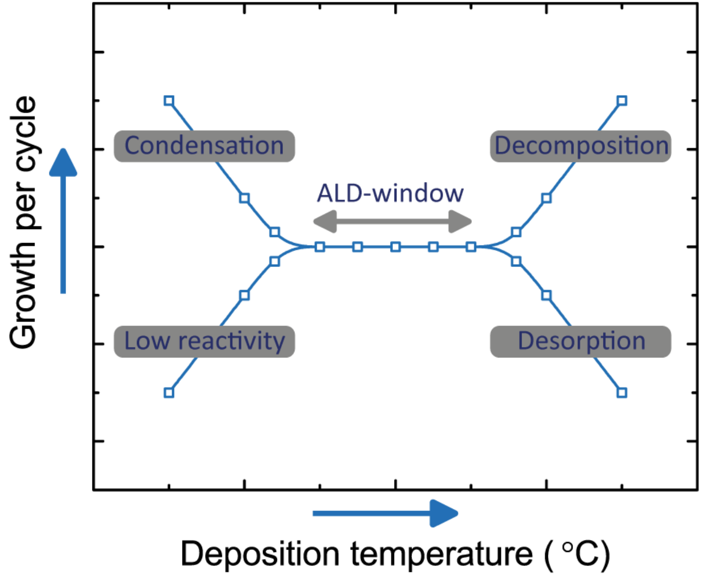



Van der Heijden *et al.*, to be published (2026)

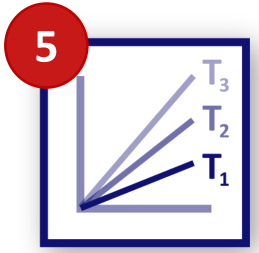
# Step 5: Temperature – Temperature window?

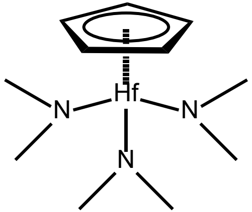


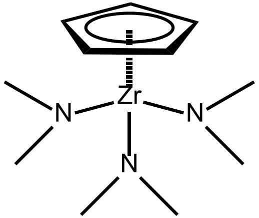

- Vary substrate temperature and measure GPC
- Check if self-limiting growth is maintained
- GPC does not need to be constant across the window
- Compare the temperature window with literature
- Check actual versus set substrate temperature

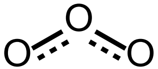


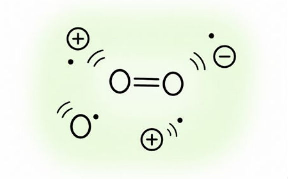
# Step 5: Temperature – $Hf_xZr_{1-x}O_2$ case

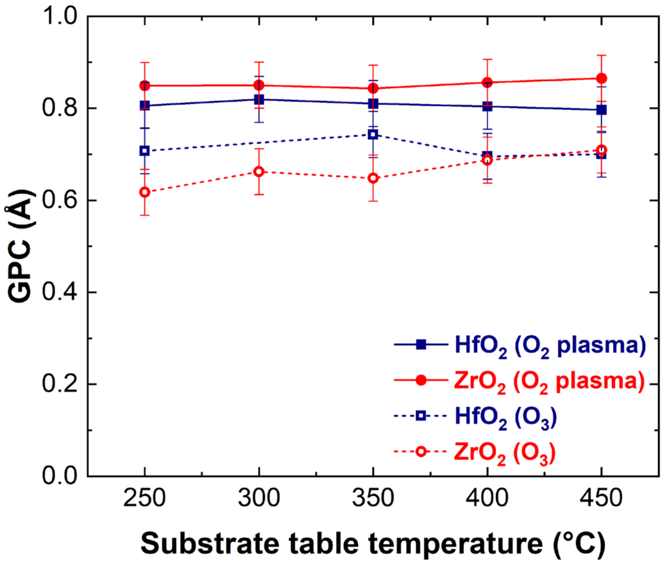


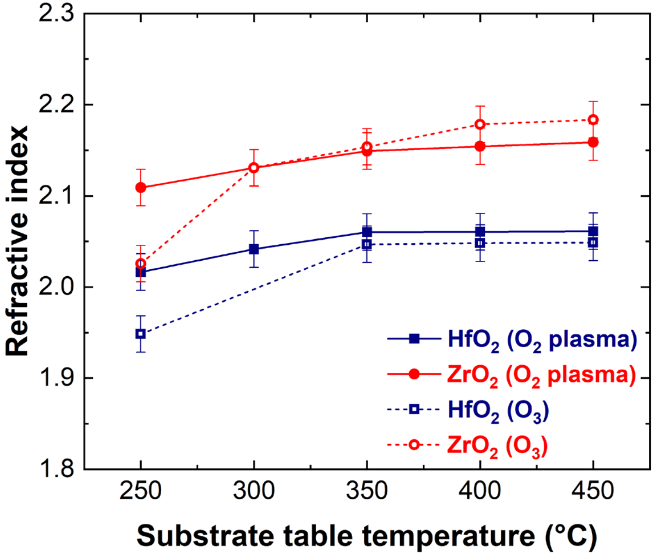







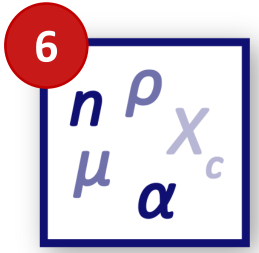


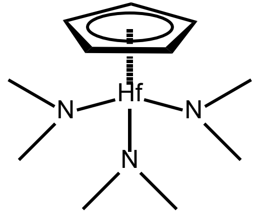

Van der Heijden *et al.*, to be published (2026)

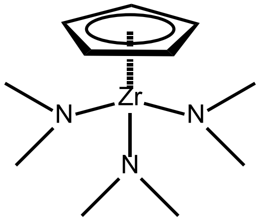
# Step 6: Film properties – Film quality?

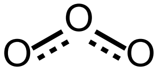
- Characterize the film beyond thickness
- Composition and impurities: XPS, RBS, SIMS, …
- Structure and morphology: XRD, Raman, TEM, AFM, …
- Density and optical properties: XRR, ellipsometry, …
- Electrical properties: 4-point probe, Hall, devices, …
- Verify temperature dependence
- Benchmark against literature

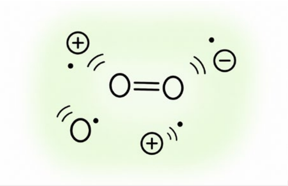
# Step 6: Film properties – $Hf_xZr_{1-x}O_2$ case



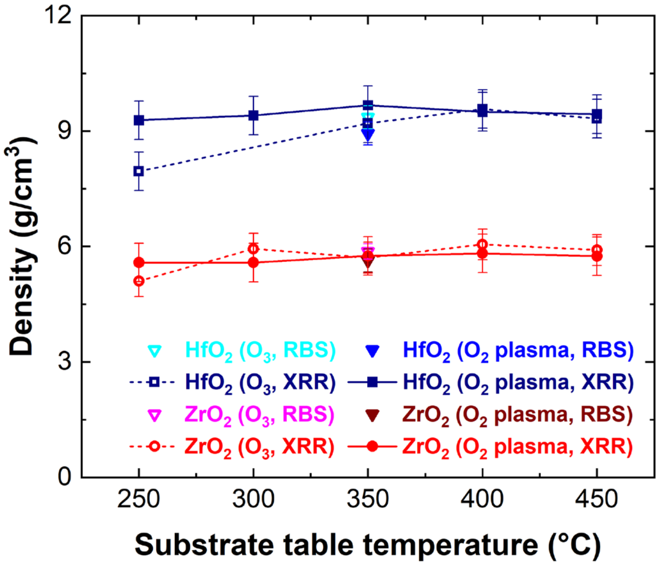





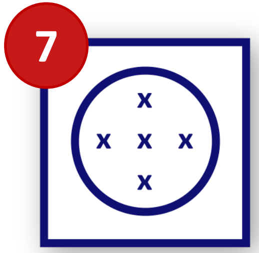
| Sample | M:O ratio | H (at. %) |
|---|---|---|
| $HfO_2$ (ozone) | 0.57 ± 0.04 | 6.2 ± 0.5 |
| $ZrO_2$ (ozone) | 0.49 ± 0.04 | 6.6 ± 0.5 |
| HZO (ozone) | 0.50 ± 0.04 | 4.8 ± 0.5 |
| $HfO_2$ (plasma) | 0.60 ± 0.04 | 8.8 ± 0.5 |
| $ZrO_2$ (plasma) | 0.55 ± 0.04 | 7.8 ± 0.5 |
| HZO (plasma) | 0.53 ± 0.04 | 12.1 ± 0.5 |





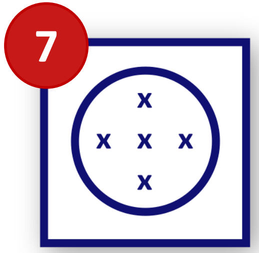

Van der Heijden *et al.*, to be published (2026)

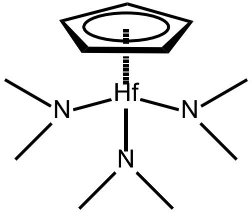
# Step 7: Uniformity – Same film across the substrate?

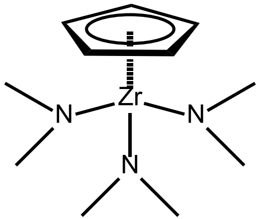



- Check uniformity on the largest substrate that fits
- Poor uniformity may indicate:
  - Incomplete saturation
  - Precursor decomposition
  - Gas-flow or reactor limitations
  - Influence of reaction products
  - Non-ideal ALD behavior
- Check both thickness and property uniformity

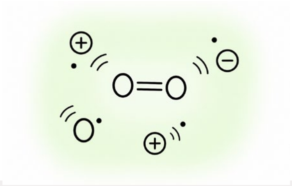
# Step 7: Uniformity – $Hf_xZr_{1-x}O_2$ case



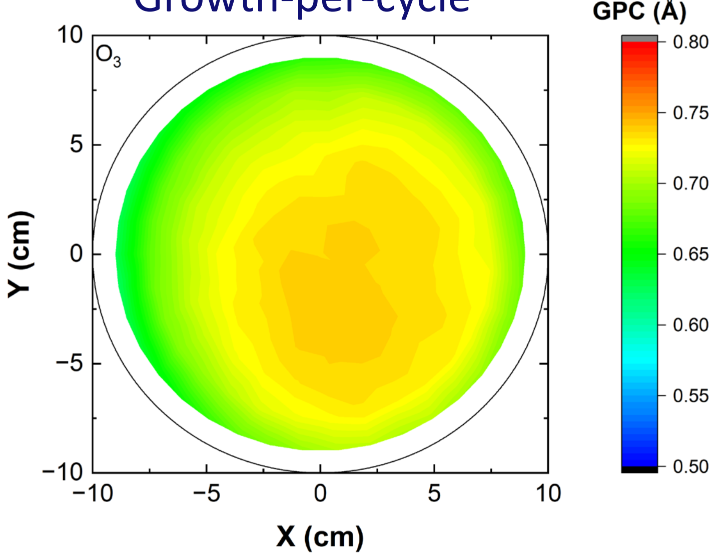





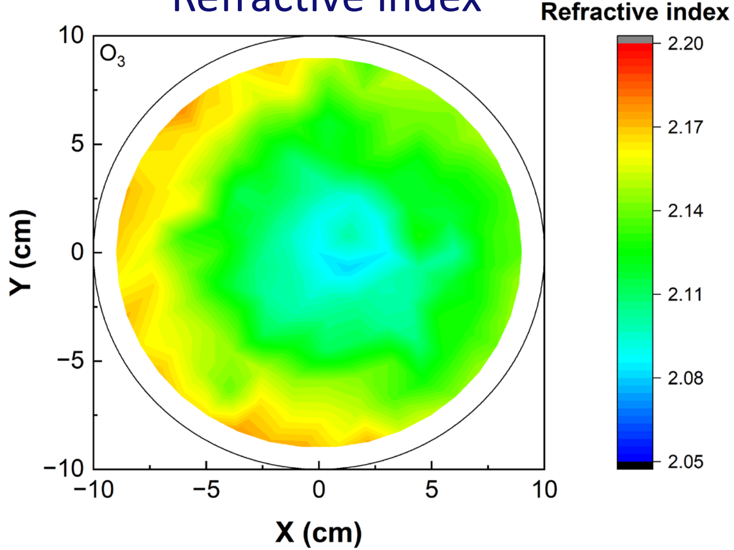



## $Hf_xZr_{1-x}O_2$ with $O_3$

### Growth-per-cycle




### Refractive index

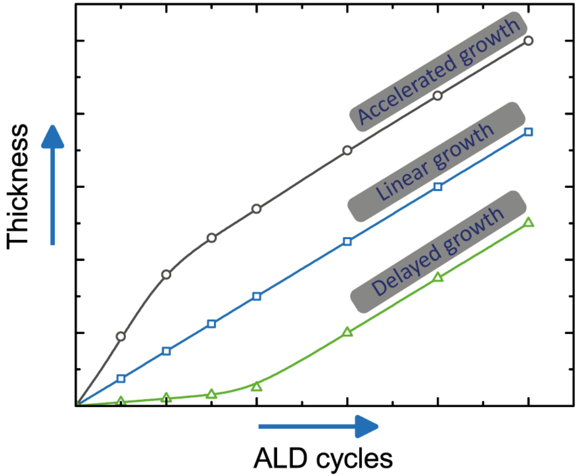




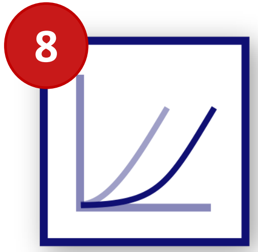

Van der Heijden *et al.*, to be published (2026)

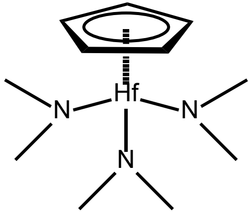
# Step 8: Nucleation – How does the film initially grow?

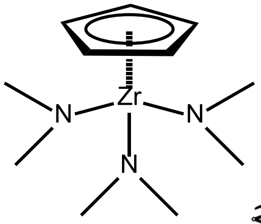
- Check growth versus cycle number on relevant substrates
- Pay attention to surface pretreatment and temperature
- Test all relevant surfaces in the final application
- Assess nucleation, island growth, and film closure
- Use early-growth tools such as ellipsometry, AFM, TEM or XPS

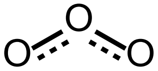


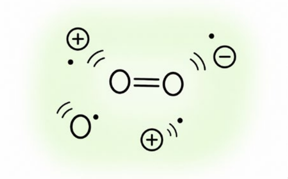
# Step 8: Nucleation – $Hf_xZr_{1-x}O_2$ case



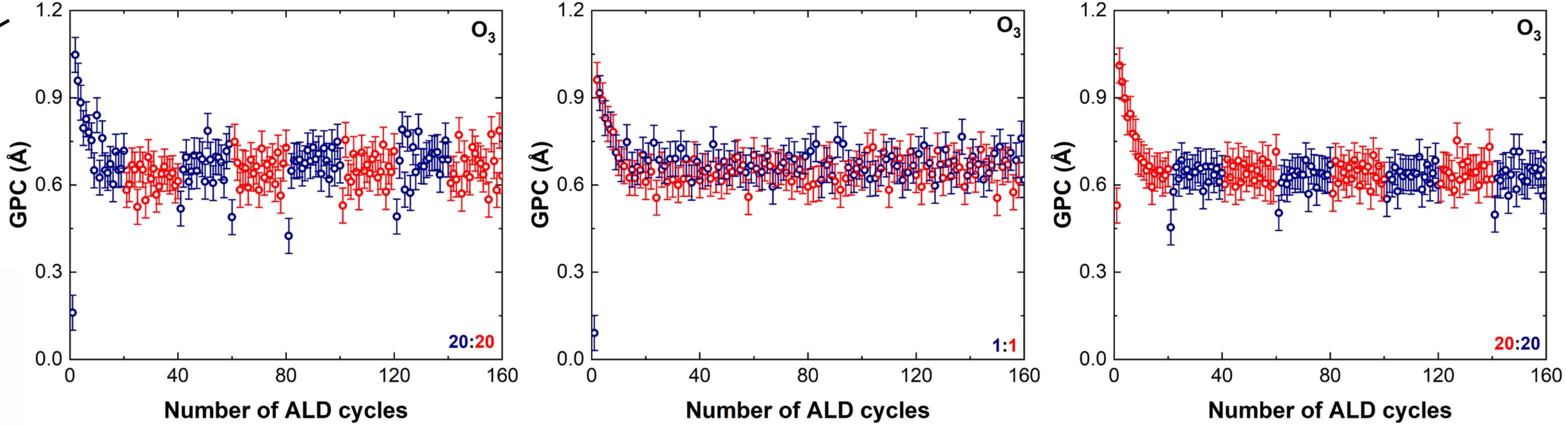




$Hf_xZr_{1-x}O_2$ with $O_3$

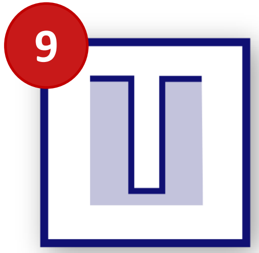
20 c. $HfO_2$ : 20 c. $ZrO_2$ | 1 c. $HfO_2$ : 1 c. $ZrO_2$ | 20 c. $ZrO_2$ : 20 c. $HfO_2$



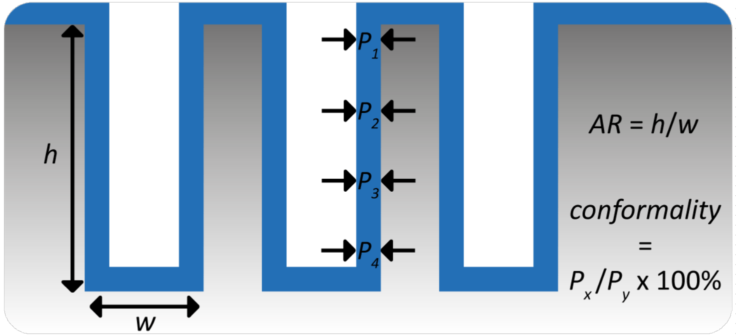



Van der Heijden *et al.*, to be published (2026)

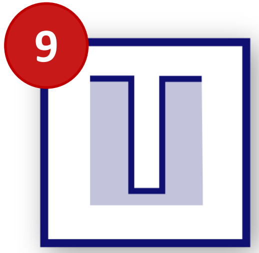
# Step 9: Conformality – Same film in 3D substrates?

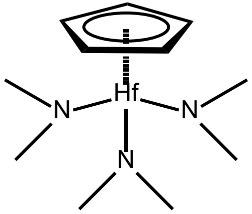


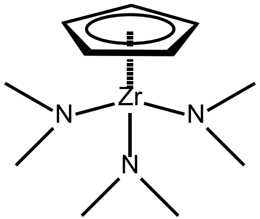

- Test conformality in relevant 3D structures
  - Trenches, pores, particles, test structures, …
- Check thickness at different positions
- Check property conformality where possible too
  - Composition, etch rate, electrical behavior, …
- Use structures with different aspect ratios
- Limited conformality can reveal transport limits or insufficient dosing

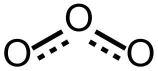


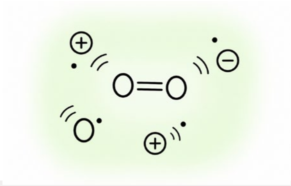
# Step 9: Conformality – $Hf_xZr_{1-x}O_2$ case



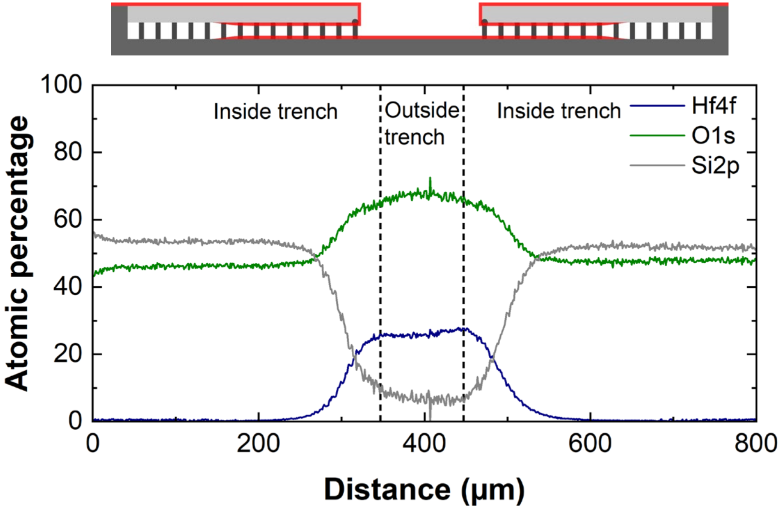



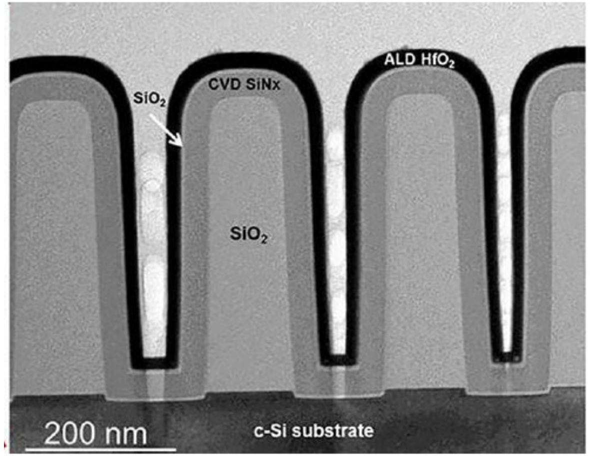



## $HfO_2$ in Chipmetrics PillarHall structure




## $HfO_2$ in etched trenches




Van der Heijden *et al.*, to be published (2026)
Sharma *et al.*, J. Vac. Sci. Technol. 35, 01B130 (2017)

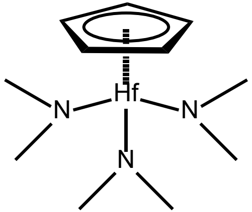


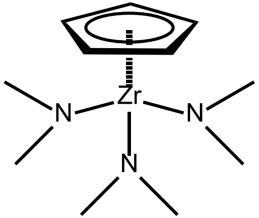
TU/e

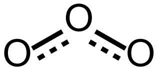
# Step 10: Other aspects – What else is important?

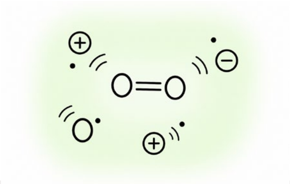

- Stability: check film changes over time and environment
- Reproducibility: repeat the same recipe many times
- Reactor history: consider wall conditioning and memory effects
- Precursor use: avoid excessive precursor consumption
- Integration: ensure compatibility with the process flow and device stack
- Throughput and scalability: match the needs of the application
- …

# Step 10: Other aspects – $Hf_xZr_{1-x}O_2$ case

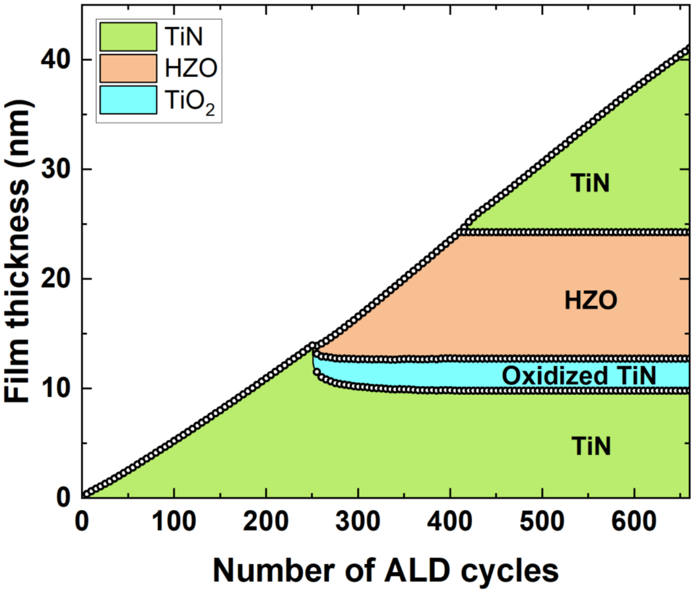



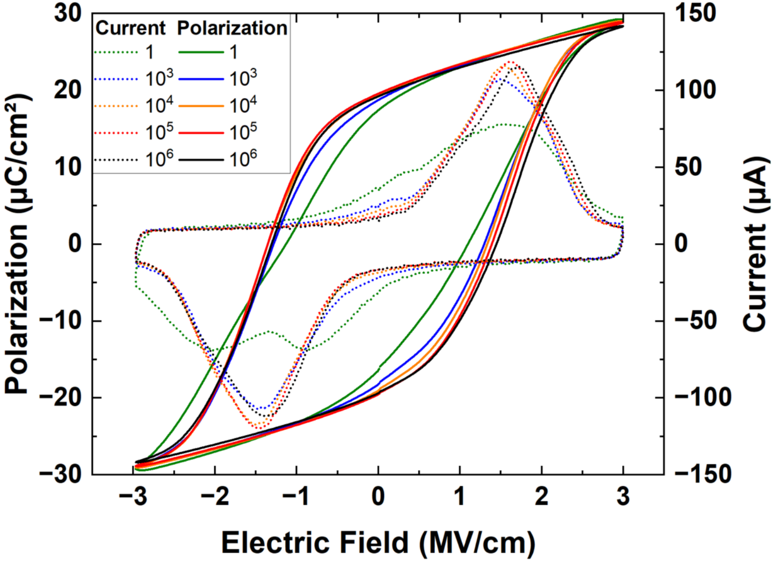



*In situ* ellipsometry during ALD of stack




Current and polarization






Van der Heijden *et al.*, to be published (2026)

# ALD process development for $Hf_xZr_{1-x}O_2$ – Illustrative example

| ALD development step | $HfO_2$ | $ZrO_2$ | $Hf_{1-x}Zr_xO_2$ |
|---|---|---|---|
| 1 Reactant selection | 🟢 | 🟢 | 🟢 |
| 2 Composition | 🟢 | 🟢 | 🟢 |
| 3 Thickness control | 🟢 | 🟢 | 🟢 |
| 4 Saturation | 🟢 | 🟢 | |
| 5 Properties | 🟢 | 🟢 | 🟢 |
| 6 Temperature | 🟢 | 🟢 | 🟢 |
| 7 Uniformity | 🟡 | 🟡 | 🟡 |
| 8 Conformality | 🟠 | 🟠 | 🔴 |
| 9 Nucleation | 🟢 | 🟢 | 🟢 |
| 10 Other aspects | 🟠 | 🟠 | 🟠 |

0.70 Å/cycle for $HfO_2$
0.65 Å/cycle for $ZrO_2$

XPS, RBS, ellipsometry, XRD, AFM, TEM data available

Check conformality
Thickness/composition

$TiN/Hf_xZr_{1-x}O_2/TiN$
Device performance

# Challenge: the hidden complexity behind an ALD recipe

## The recipe is only part of the process; many hidden factors can shape the outcome

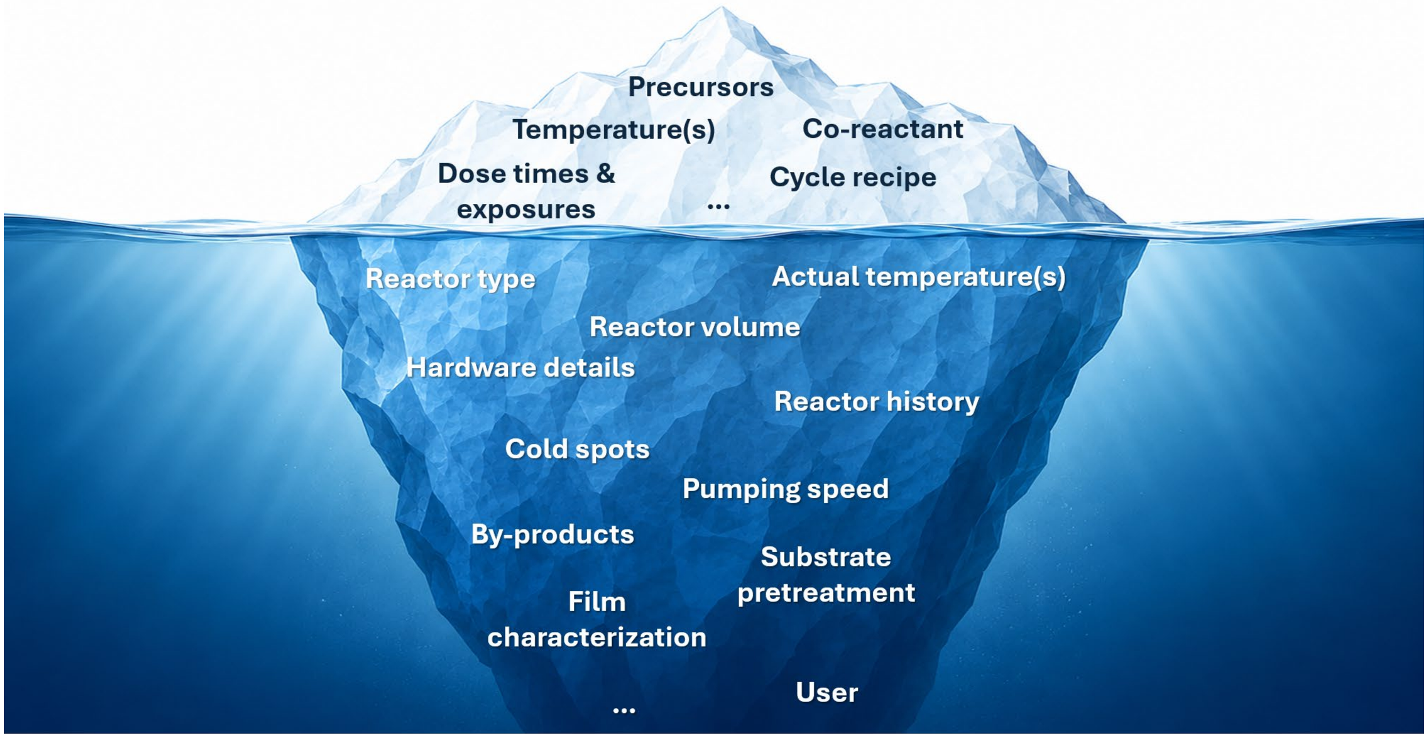





Image: Atomic Limits – TU/e (2026)

# Outline

# AI for ALD is emerging as a rapidly growing research theme

## Literature spanning modeling, optimization, design, extraction, and agentic workflows*

| Group | Authors | Title | Reference |
|---|---|---|---|
| Christofides | Ding *et al.* | Machine learning-based modeling and operation for ALD of $SiO_2$ thin-films using data from a multiscale CFD simulation | Chem. Eng. Res. Des. 151, 131 (2019) |
| | Ding *et al.* | Machine learning-based modeling and operation of plasma-enhanced atomic layer deposition of hafnium oxide thin films | Comput. Chem. Eng. 144, 107148 (2021) |
| | Tom *et al.* | Machine learning-based run-to-run control of a spatial thermal atomic layer etching reactor | Comput. Chem. Eng. 168, 108044 (2022) |
| | Tom *et al.* | Machine Learning Modeling and Run-to-Run Control of an Area-Selective Atomic Layer Deposition Spatial Reactor | Coatings 14, 38 (2024) |
| Yanguas-Gil & Elam | Paulson *et al.* | Intelligent Agents for the Optimization of Atomic Layer Deposition | ACS Appl. Mater. Inter. 13, 17022 (2021) |
| | Yanguas-Gil *et al.* | Machine learning and atomic layer deposition: Predicting saturation times from reactor growth profiles using artificial neural networks | J. Vac. Sci. Technol. A 40, 82408 (2022) |
| | Yanguas-Gil *et al.* | Surrogate models to optimize plasma-assisted atomic layer deposition in high aspect ratio features | Phys. Plasmas 32, 73507 (2025) |
| | Yanguas-Gil *et al.* | Design and performance of AI agents interfacing with an atomic layer deposition tool | Rev. Sci. Instrum. 97, 053903 (2026) |
| Banerjee | Arunachalam *et al.* | Machine learning approach to thickness prediction from in situ spectroscopic ellipsometry data for atomic layer deposition processes | J. Vac. Sci. Technol. A 40, 12405 (2022) |
| Shong | Van *et al.* | Machine learning-based exploration of molecular design descriptors for area-selective atomic layer deposition (AS-ALD) precursors | J. Mod. Model 30, 10 (2024) |
| | Dogan *et al.* | Bayesian Machine Learning for Efficient Minimization of Defects in ALD Passivation Layers | ACS Appl. Mater. Inter. 13, 54503 (2021) |
| | Kim *et al.* | Machine learning-based analysis of the physio-chemical properties for the predictive thickness control of atomic layer deposition | IFAC-PapersOnLine 55, 626 (2022) |
| | Chiu *et al.* | Ligand Engineering to Machine learning : Optimizing Ru ALD for Ultrathin Film Deposition | Proc. IITC 2024 |
| | Häussermann *et al.* | Leveraging Bayesian Optimization Software for Atomic Layer Deposition: Single-Objective Optimization of $TiO_2$ Layers | Materials 17, 5019 (2024) |
| | Seo *et al.* | Deriving optimal atomic layer deposition process conditions using machine learning | J. Ind. Inf. Integr. 47, 100879 (2025) |
| | Gu *et al.* | AI-Driven Inverse Design of Complex Oxide Thin Films for Semiconductor Devices: A Case Study of Hf-Zr-O | Chem. Mater. 38, 5285 (2026) |
| Takoudis | Navabi *et al.* | Artificial intelligence in atomic layer deposition | J. Vac. Sci. Technol. A 43, 60801 (2025) |
| | Ramachandran *et al.* | Advancing atomic layer deposition through artificial intelligence: Generative modeling and predictive analytics for $TiO_2$ growth rate prediction from TDMAT/$O_3$ | Surf. Coat. Technol. 526, 133341 (2026) |
| Bent & Tonner-Zech | Westermayr *et al.* | Opportunities and Challenges of Machine Learning in Atomic Layer Deposition | Chem. Mater. 38, 2142 (2026) |
| D'Souza, Karasulu Mackus & Kessels | Sadruddin *et al.* | Designing an agentic AI workflow for structured information extraction from scientific text: A case study on atomic layer deposition of ZnO and IGZO | J. Vac. Sci. Technol. A 44, 32413 (2026) |
| | Sadruddin *et al.* | SCHEMA-MINERpro: Agentic AI for Ontology Grounding Over LLM-Discovered Scientific Schemas in a Human-in-the-Loop Workflow | Semantic Web 17, 1 (2026) |
| | D'Souza *et al.* | Publishing FAIR and machine-actionable reviews in materials science: The case for symbolic knowledge in neuro-symbolic artificial intelligence | J. Vac. Sci. Technol. A 44, 32408 (2026) |

*Not a complete literature survey.

# A vision for AI-enabled ALD: closed-loop AI-workflow

## From target properties to recipe prediction, experiments, and model learning

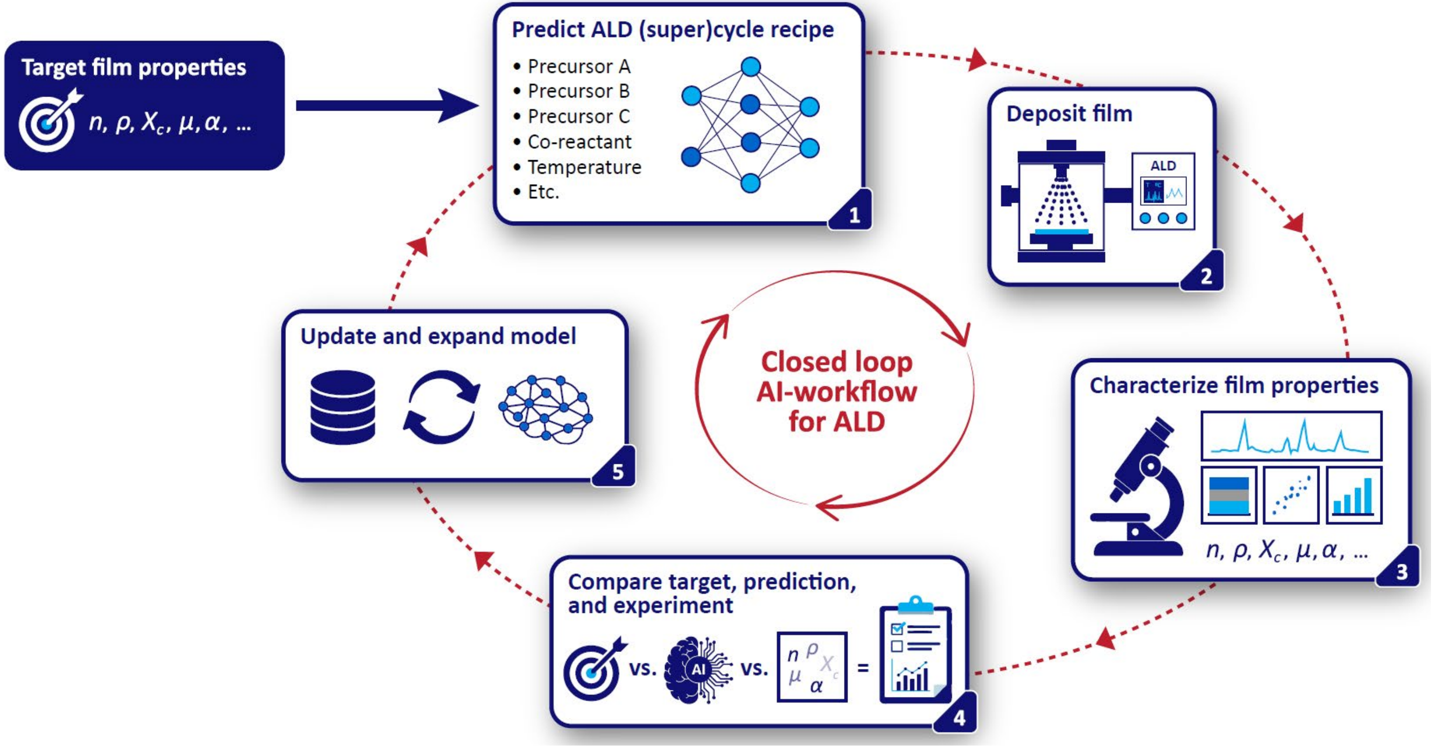





Image: Atomic Limits – TU/e (2026)

# What an AI-enabled ALD workflow needs from the data layer

## The AI loop needs structured ALD knowledge, context, and feedback

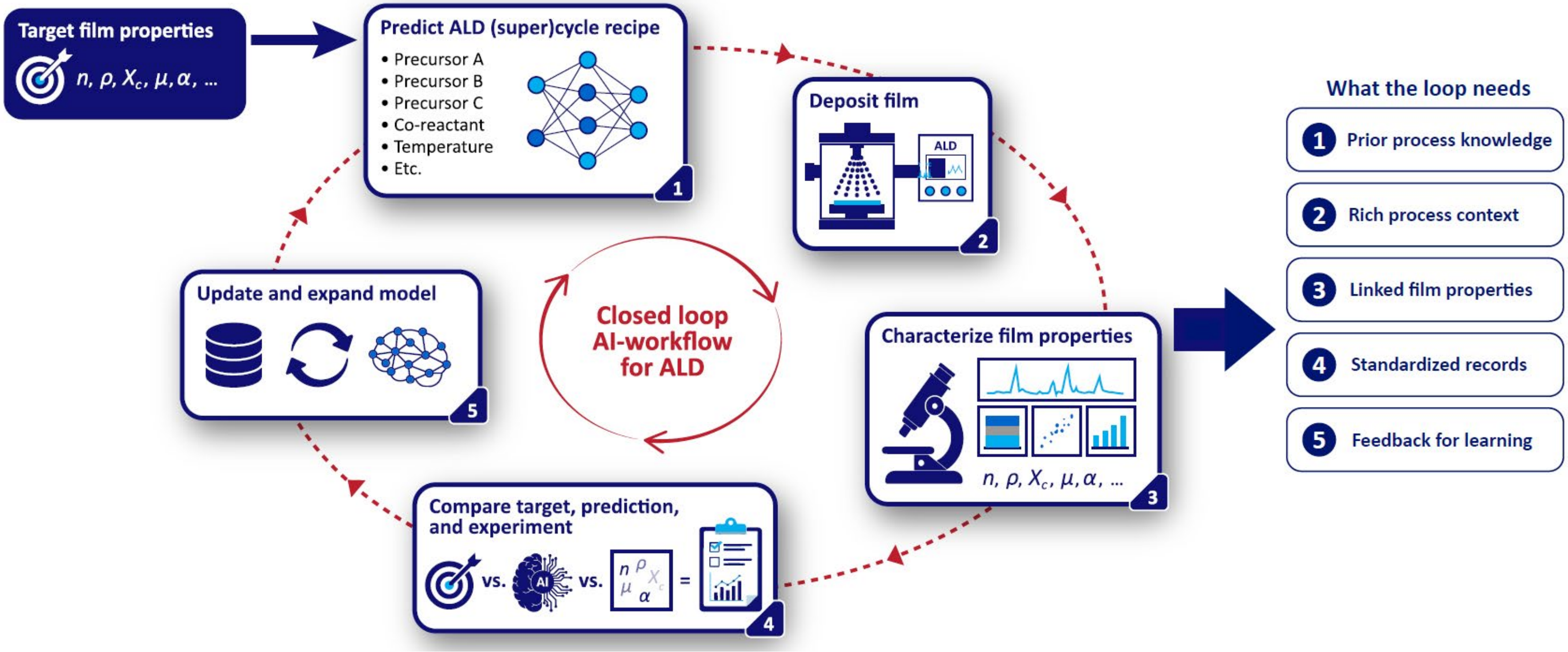
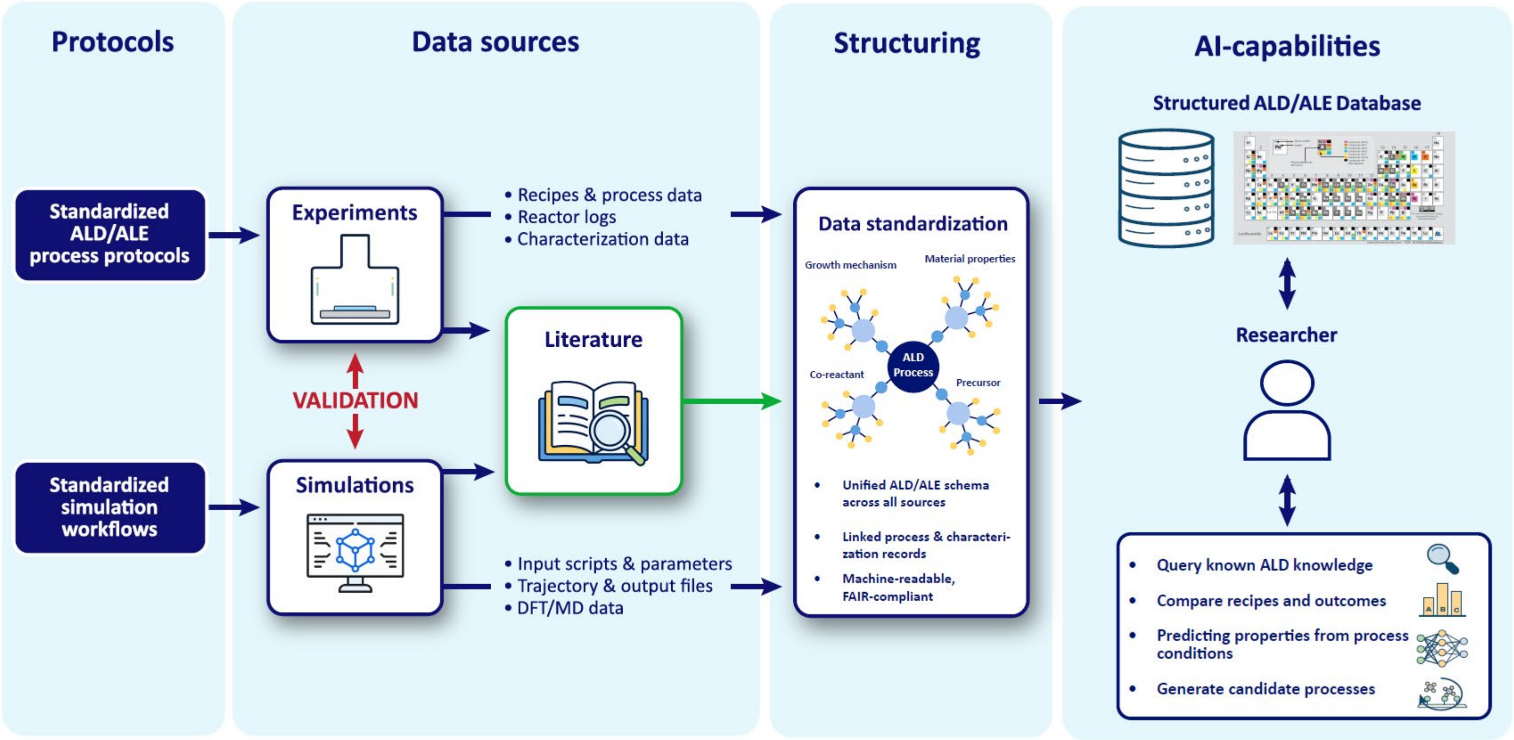





Image: Atomic Limits – TU/e (2026)

# Making the ALD database AI-ready

## Building the data layer needed for AI-enabled ALD

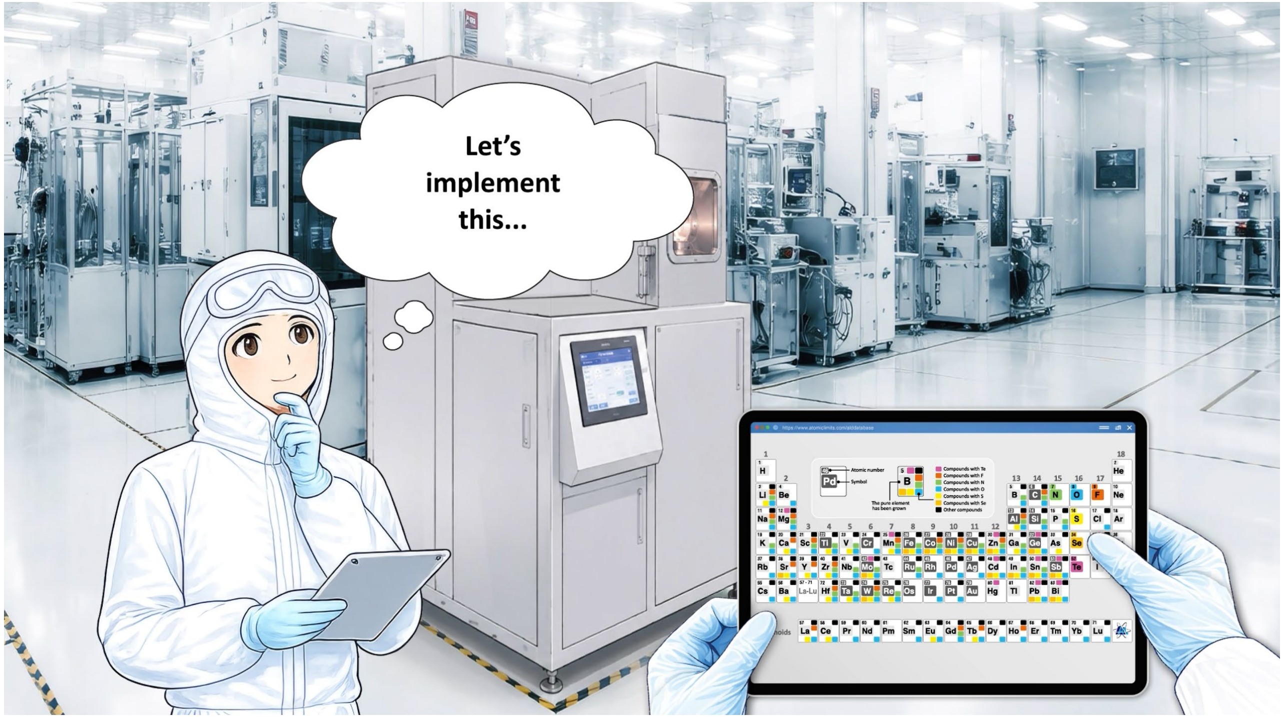



Sadruddin *et al.*, Semantic Web 17, 1 (2026)
Sadruddin *et al.*, J. Vac. Sci. Technol. A 44, 32413 (2026)
D'Souza *et al.*, J. Vac. Sci. Technol. A 44, 32408 (2026)

# Mock-up: an AI-ready ALD database in action

## From today's ALD database toward a research tool for AI-enabled ALD

Video

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# Outlook: from AI assistance to AI agents to self-driving labs

## From supporting decisions, to coordinating research tasks, to driving experimental loops

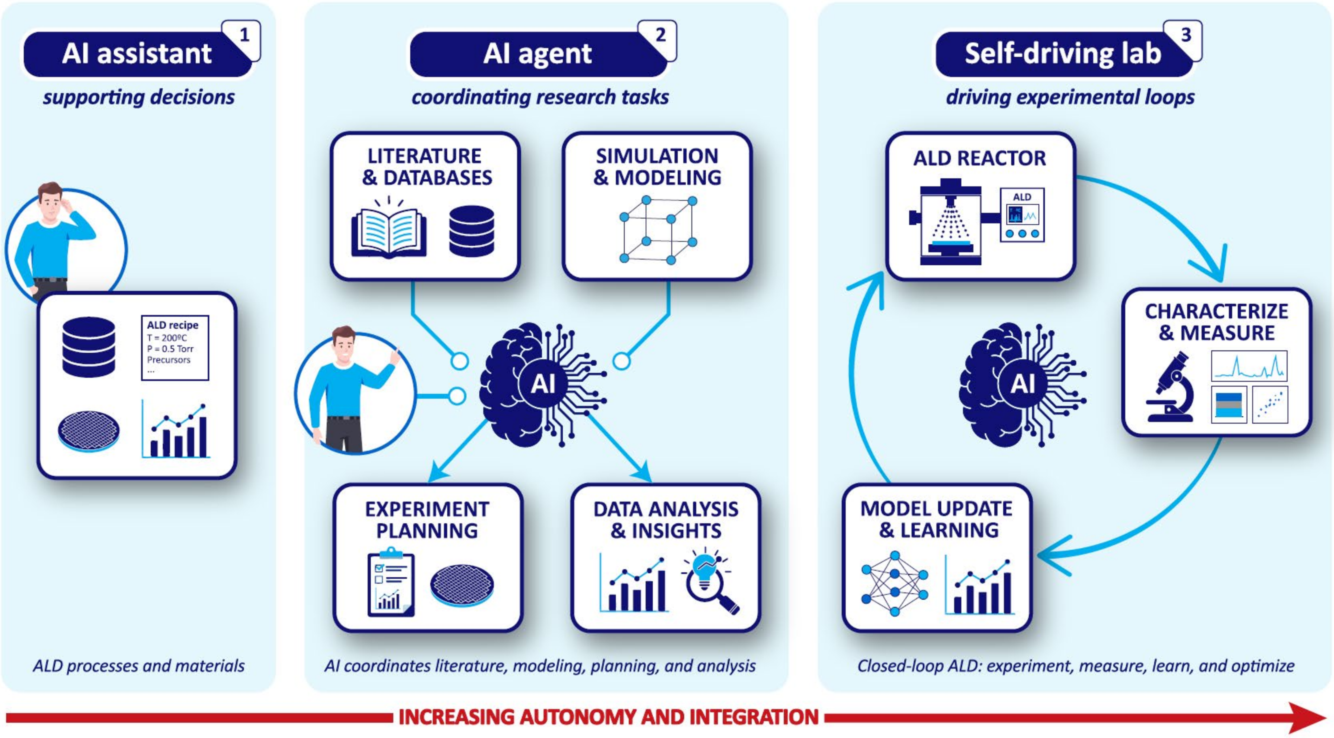





Image: Atomic Limits – TU/e (2026)

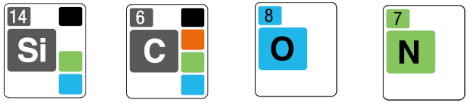
# Outline

1. Quick introduction to ALD
2. ALD process development
3. Towards AI-enabled ALD
4. **Challenges in frontier materials**

# Materials with a difficult ALD chemistry

## Representative material classes where robust self-limiting chemistry remains challenging

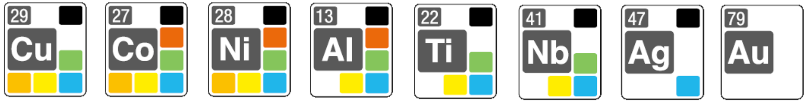
### Foundational Si- and C-based materials




Examples: a-Si, a-C, $SiN_x$, SiCN, low-k, etc.

Limited thermal chemistries, often $O_3$ or plasma based (if any), conformality, low-temperature, …

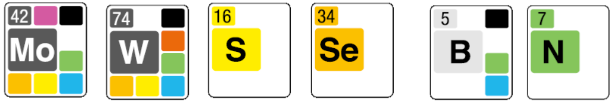
### Metals and their alloys




Examples: Cu, Co, Ni, Al, Ti, Ta, Nb, Au, Ag alloys, etc.

Limited thermal chemistries; reduction to metallic state, contamination, nucleation, film closure, …

### Low-dimensional materials including 2D transition metal chalcogenides




Examples: $MoS_2$, $WSe_2$, h-BN, graphene, etc.

Stoichiometry, layer control, crystallinity, phase, …

### Alkali-containing materials


Examples: battery materials, Na/K-based films, etc.

Air-sensitive precursors, high mobility, hygroscopic films, and non-classical ALD chemistry…

Ovanesyan *et al.*, J. Vac. Sci. Technol. A 37, 060904 (2019)
Hagen *et al.*, Appl. Phys. Rev. 6, 041309 (2019)
Mattinen *et al.*, Adv. Mater. Interfaces 8, 2001677 (2021)
Madadi *et al.*, ACS Appl. Mater. Interfaces 13, 56793 (2021)



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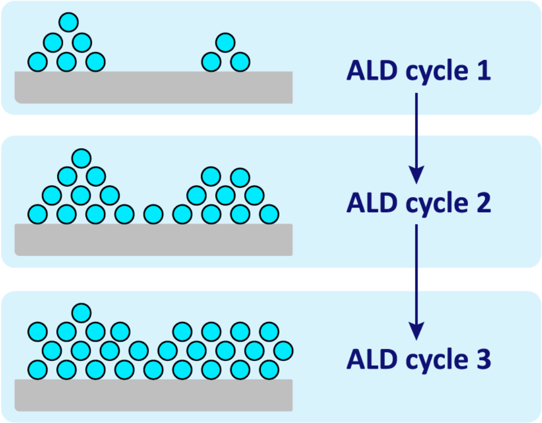
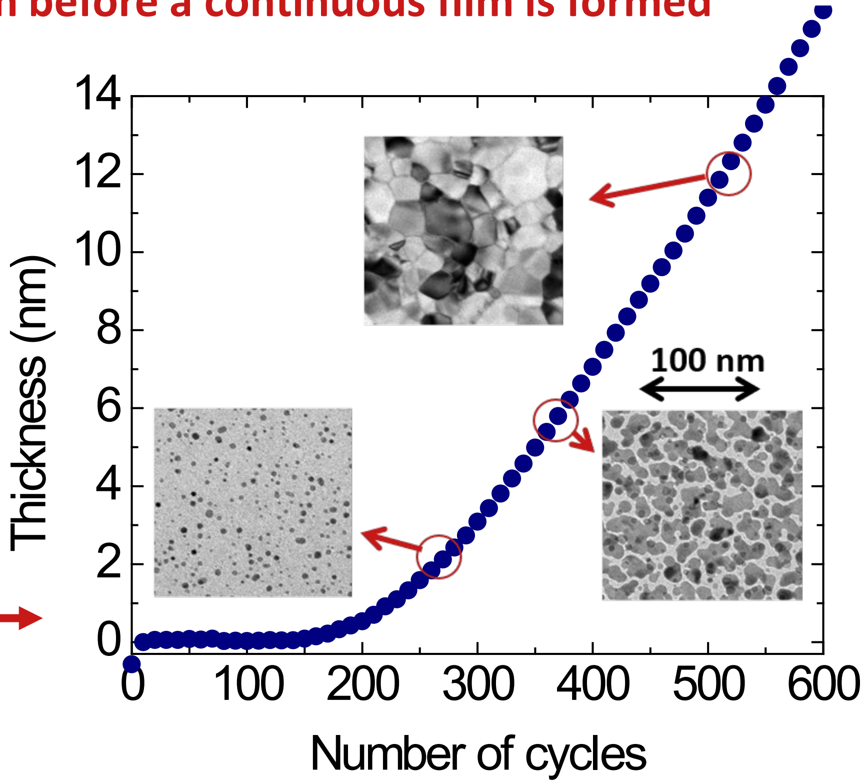
# Nucleation and film closure for ALD metal films

**Metal ALD often starts with island growth before a continuous film is formed**




"Classic example": Pt ALD on oxide surfaces

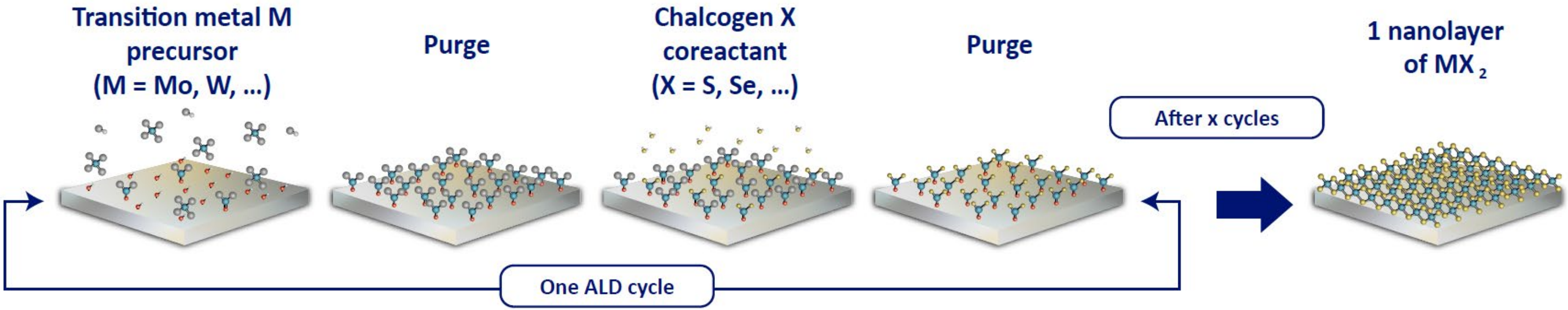
Also highly relevant for other Pt-group metals and coinage metals such as Ag and Au

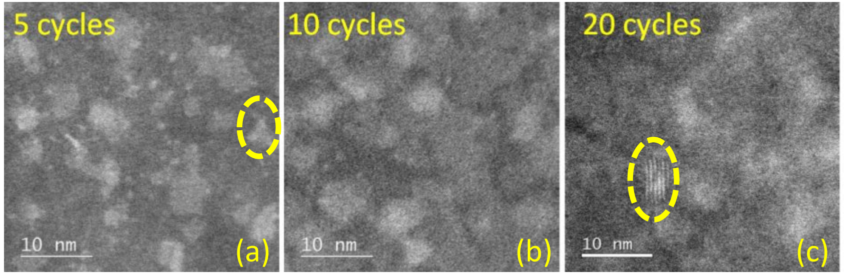



Mackus *et al.*, Chem. Mater. 25, 1905 (2013)
Hagen *et al.*, Appl. Phys. Rev. 6, 041309 (2019)

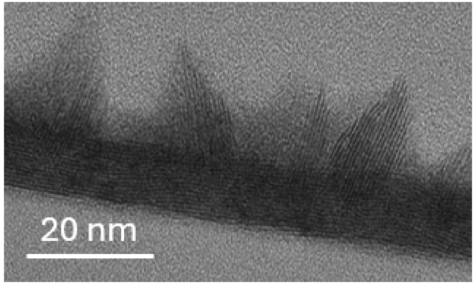
# Layer control and crystallinity for ALD 2D materials

## Low-temperature & conformal — but large-grain, in-plane oriented layers seems hard




Grain size is limited to the tens-of-nm range




Out-of-plane-oriented structures

Sharma *et al*., Nanoscale 10, 8615 (2018)
Mattinen *et al*., Adv. Mater. Interfaces 8, 2001677 (2021)

# ALD challenges in frontier materials

## From making the material to achieving the required film functionality

### Preparing materials with difficult ALD chemistry

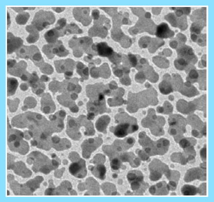
*Can the material itself be grown?*

Intrinsic chemistry challenges:
precursors · reactants · impurities


### Forming closed films, especially at ultralow thickness

*Can a continuous layer form early enough?*

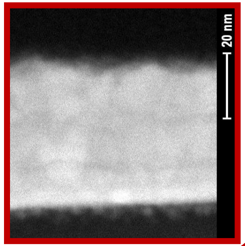
Nucleation challenges:
nucleation · coalescence · closure



### Controlling elemental mixing beyond binaries

*Can elements be mixed at the atomic scale?*

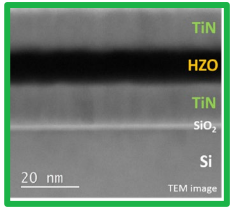
Process route challenges:
mixing · interfaces · phase purity




### Achieving highly crystalline films

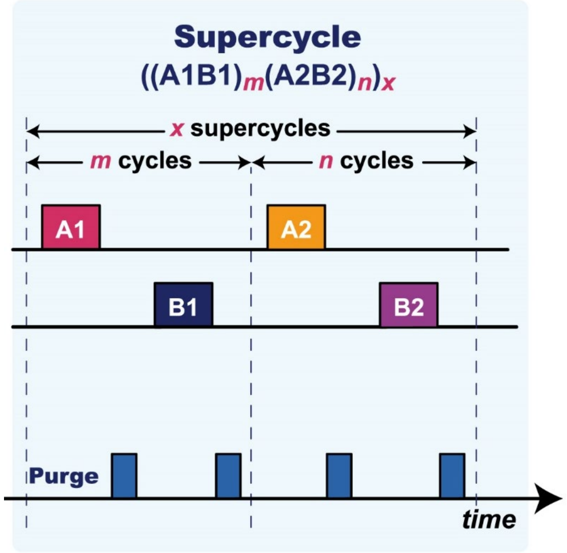
*Can a process deliver the right phase?*

Process route challenges:
phase · orientation · grains

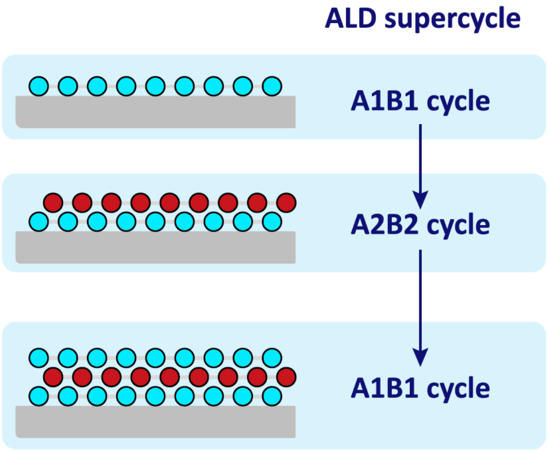


# ALD challenge: elemental mixing beyond binary compounds

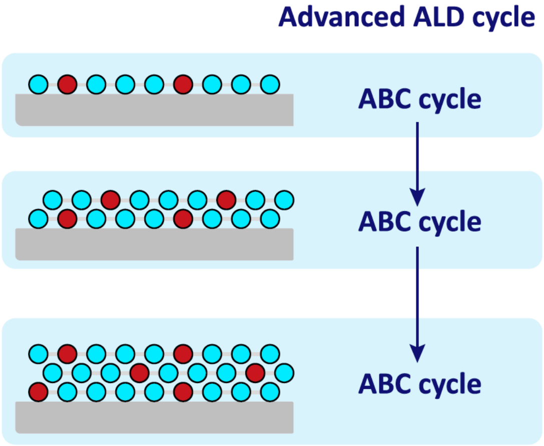
**Average composition is not enough: the atoms also need to be mixed in the right way**




**Supercycles can result in nanolaminate-like films**




**Advanced (super)cycles can promote more homogeneous mixing**



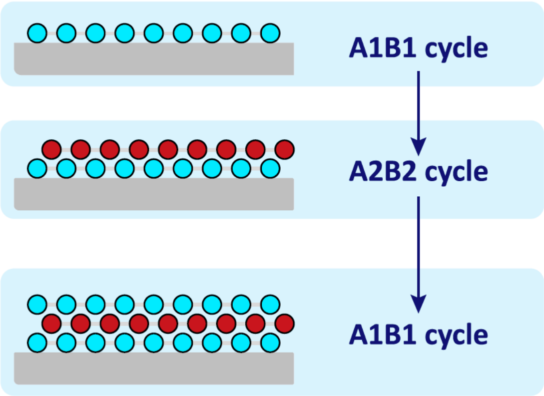


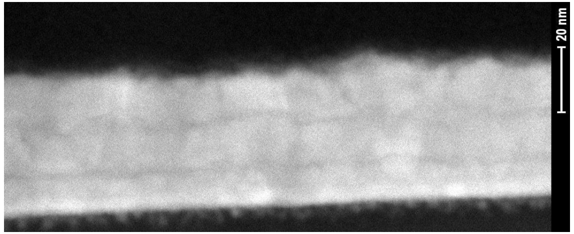

Mackus *et al.*, Chem. Mater. 31, 1142 (2019)

# ALD of nanolaminate films vs. elemental mixing

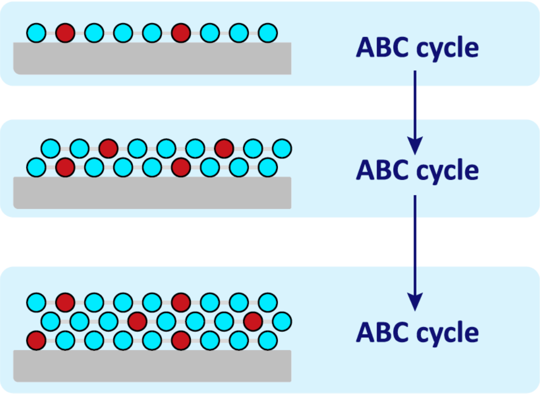
## Better distributed dopants or mixed elements through smart designed ALD processes

**ALD supercycle**




**Advanced ALD cycle**



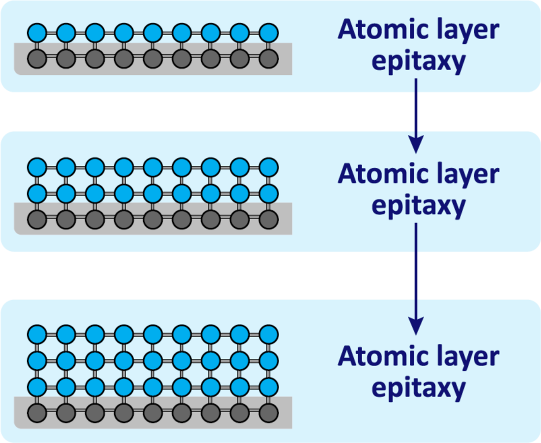

Approaches: Use of inhibitors, bulky precursors, modified sub-cycle sequence, co-dosing, atomic layer modulation, etc.

Kim *et al*., J. Vac. Sci. Technol. A 28, 1111 (2010)
Yanguas-Gil *et al*., Chem. Mater. 23, 4295 (2011)
Wu *et al*., Chem. Mater. 25, 4619 (2013)
Trinh *et al*., ACS Nano 20, 17273 (2026)

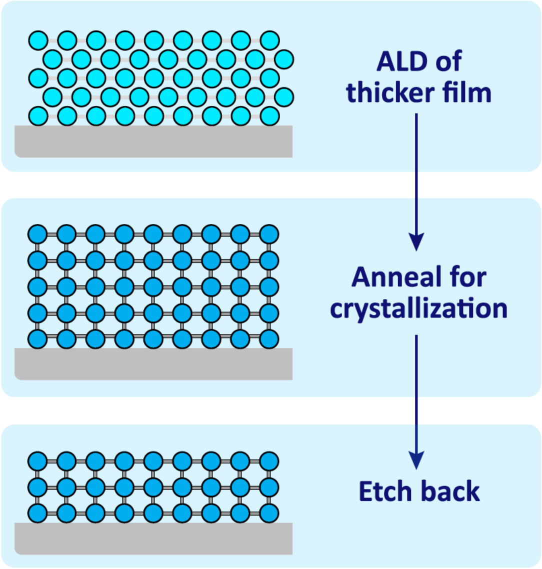
# ALD challenge: routes toward ultrathin crystalline films

**Sometimes the solution is not a better recipe, but a smarter process route**

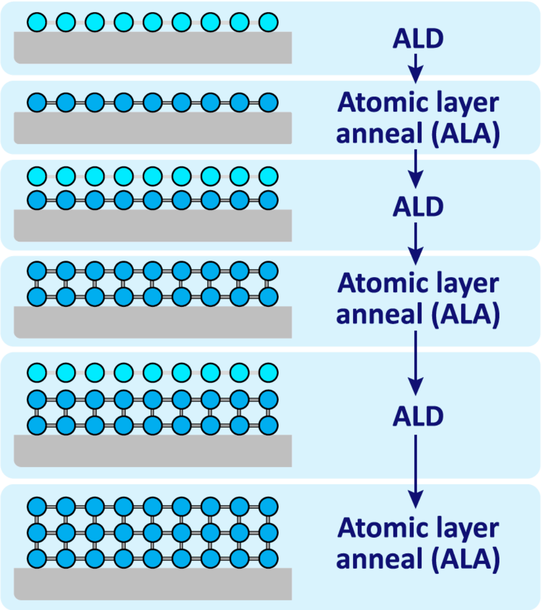
### Template-assisted crystalline growth




### Grow thicker → crystallize → etch back



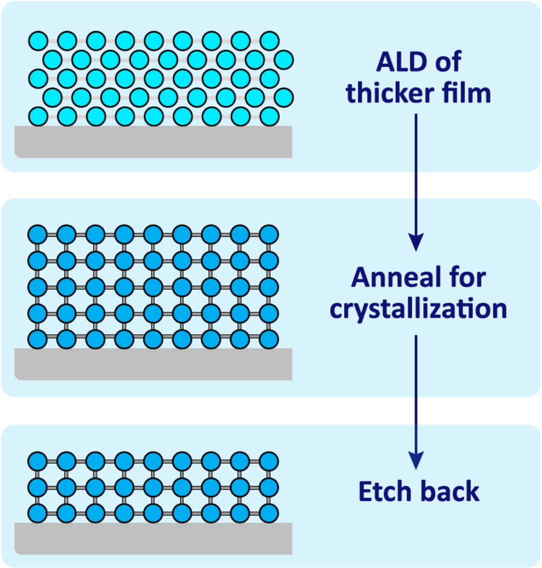

### Combining ALD with periodic activation/annealing



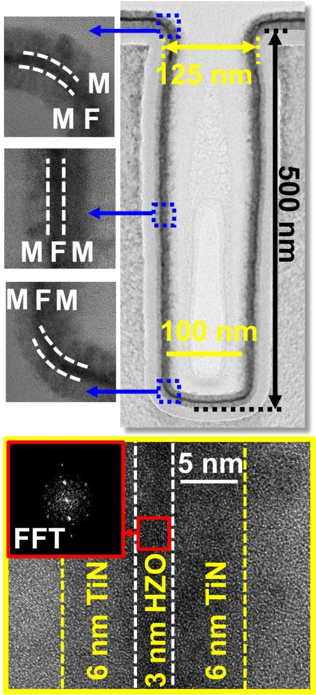

Goodman and Pessa, J. Appl. Phys. 60, R65 (1986)
McBriarty, blog post on www.AtomicLimits.com (2022)
Shih *et al.*, Sci. Rep. 7, 39717 (2017)

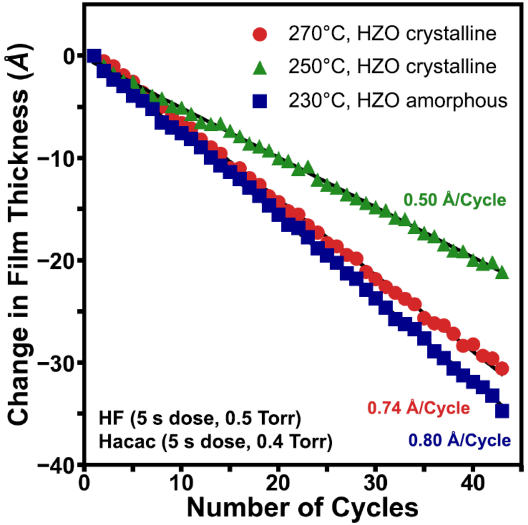
# ALD of ultrathin crystalline films using etch back

## Combining ALD and ALE might be alternative for template-assisted crystalline growth




Example: ultrathin $Hf_xZr_{1-x}O_2$ (HZO) for 3D ferroelectric capacitors



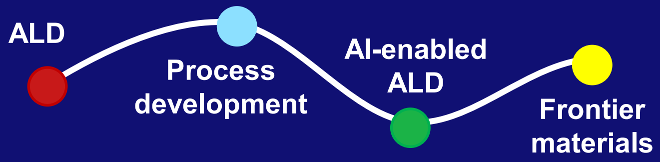




ALE of amorphous and crystalline $Hf_xZr_{1-x}O_2$



Feng *et al.*, VLSI symposium (2026)
Hoffmann *et al.*, Appl. Phys. Lett. 120, 122901 (2022)
Colleran *et al.*, Chem. Mater. 37, 5935 (2025)

# Connecting the story




- ALD is a powerful atomic-scale processing method for materials synthesis
- Developing robust ALD processes (including equipment, precursors, etc.) remains a central challenge for the community.
- Structured workflows, protocols, and databases make process development more systematic
- These structured workflows and data are also essential for AI-enabled ALD
- AI can help us address remaining challenges in frontier materials — together with related approaches such as ALE and ASD

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PMP
Plasma & Materials Processing

PMP principal investigators:
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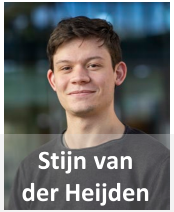
Julian Held – Ye Wang – Erwin Kessels

## FluFe project:

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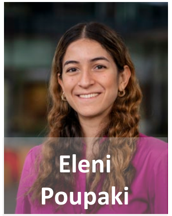
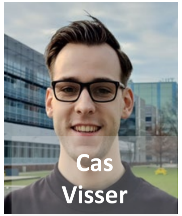
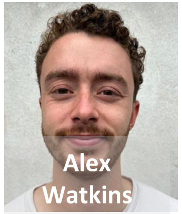
## AWASES project:





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