

ALD Applications

Room Hall 3D - Session AA2-TuM

Energy: Solar Energy Materials I

Moderators: Wei-Min Li, Jiangsu Leadmicro Nano-Equipment Technology Ltd., Nathanaelle Schneider, CNRS-IPVF

10:45am **AA2-TuM-12 ALD Layers and Interfaces in Next Generation Photovoltaics**, *Mariadriana Creatore*, TU Eindhoven, Netherlands **INVITED**
Innovation in thin film and interface engineering has played an essential role in pushing the conversion efficiency of the most widespread photovoltaic (PV) technology, i.e. crystalline silicon-based (c-Si), towards its thermodynamic limit. In this respect, ultra-thin, conformal, high-purity Al₂O₃ thin films synthesized by ALD are key in c-Si PV manufacturing industry as they passivate the c-Si surface, thereby suppressing a major channel of electron-hole pair recombination.

Recently, we have explored the ALD synthesis of thin films for metal halide perovskite-based photovoltaics. The latter has rapidly reached a conversion efficiency of 26% and, when coupled with c-Si PV in a so-called tandem device, leads to efficiencies already beyond 33%.

In this contribution I will discuss the merits which ALD offers to perovskite-based PV by reviewing our work on NiO-based hole transport layers and discussing in depth the case study of ALD SnO₂. The latter is implemented in perovskite PV R&D and industry as buffer layer, i.e., imparting thermal and environmental stability to the device, while protecting the perovskite absorber and fullerene electron transport layer from the sputtering of the transparent top contact. More recently, ALD SnO₂ is explored as solvent barrier layer in the tunnel recombination junction of perovskite/perovskite tandem PV, to prevent the damage of the wide-gap perovskite absorber when processing the narrow band-gap perovskite cell. Although we can conclude that several ALD merits are already extensively acknowledged by the PV community, studies addressing ALD film growth on challenging substrates such as fullerenes and metal halide perovskites are rarely reported in literature. We are convinced that these studies provide a rationale to implement more efficiently these layers at device level and promote process upscaling. Therefore, this contribution will also highlight the adoption of *in situ* diagnostics, namely spectroscopic ellipsometry and IR spectroscopy, to characterize the ALD SnO₂ growth on two commonly adopted fullerenes, C60 and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM). Our studies show that a substrate-inhibited growth occurs in the case of PCBM and, to a minor extent, in the case of C60, with respect to the traditional c-Si substrate. Moreover, IR spectroscopy highlights the loss of vibrational features of the ester group in PCBM upon SnO₂ growth, whereas C60 is chemically unaffected. We conclude that the delayed film growth and chemical modifications detected on PCBM are responsible for the consistently lower device performance when ALD SnO₂ is grown on PCBM instead of C60.

11:15am **AA2-TuM-14 Utilizing Low-Temperature Ald Technique to Investigate Perovskite Nickelates for Photovoltaic Applications**, *Anjali Choubey*, *H. Hovde Sønsteby*, University of Oslo, Norway; *H. von Wenckstern*, University of Oslo, Germany; *O. Nilsen*, University of Oslo, Norway

Photovoltaic (PV) materials currently in use like silicon are well-established but approaching their theoretical limit, and halide-perovskites, even though developing rapidly, are toxic and unstable under environmental conditions. Developing new materials for PV applications is slowly becoming a crucial necessity. In this study, perovskite nickelates are investigated as an alternative to traditional PV materials. Nickelates are not only stable under environmental conditions, but they can also withstand photobleaching and thermal exposures. Additionally, they are well known for their metal-insulator phase transition, magnetic phase transition, and structural transition [1, 2]. This has made them very lucrative for various electronic, magnetic, dielectric, superconducting, and catalytic applications. However, there is not much data on the use of nickelates for PV applications, and we are trying to bridge that gap.

Some perovskite nickelates have shown bandgap tuning abilities by A-site substitution control [2-4]. In this work, Terbium nickelate (TbNiO₃) and Gadolinium nickelate (GdNiO₃) thin films fabricated using low-temperature atomic layer deposition (ALD) techniques were studied and characterized. X-ray diffraction gave information about the structure and crystallinity, x-ray fluorescence and x-ray photoelectron spectroscopy about the composition, and spectroscopic ellipsometry about the thickness and refractive indices of

the deposited films. These investigations helped gain valuable insight into the extent of the semiconductive nature of these materials and how to exploit them for PV applications.

Reference:

1. Catalano, S., et al., *Reports on Progress in Physics*, 81(4), 046501 (2018).
2. Mercy, A., et al., *Nature Communications*, 8(1), (2017).
3. Chang, L., et al., *Journal of Physics D: Applied Physics*, 49(44), (2016).
4. Chang, L., et al., *ACS Applied Materials & Interfaces*, 11(17), (2019).

11:30am **AA2-TuM-15 Atomic Layer Deposition of Defect-Engineered TiO_x and TaO_x Protective Coatings for Photoelectrochemistry**, *Tim Rieth*, *O. Bienek*, *J. Kühne*, *I. Sharp*, Walter Schottky Institut, Technische Universität München, Germany

Photoelectrochemical (PEC) energy conversion provides a viable route to chemical fuel production from solar light. In this approach, charge carriers generated within a semiconductor light absorber immersed in an electrolyte are used to drive electrochemical reactions. Several different PEC design approaches exist, ranging from single photoelectrodes to tandem configurations of photoanodes and -cathodes to buried photovoltaic junctions. However, common to each of these designs is a central materials challenge of achieving simultaneous stability, efficiency, and scalability. Furthermore, the coupling of the semiconducting photo absorber and catalytic components demands sophisticated solid state interface engineering. In this talk, we focus on the stabilization of this semiconductor/electrolyte interface with functional conformal coatings grown by plasma-enhanced atomic layer deposition (PE-ALD). We investigate defect-engineered electron-selective titanium oxide (TiO_x) protection layers for efficient InP photocathodes and identify ALD pathways to optimize interfacial and bulk charge transport, as well as to tune the driving force for photovoltage generation [1, 2]. Furthermore, we use PE-ALD to deposit ultra-thin tantalum oxide (TaO_x) protective coatings showing improved chemical stability compared to TiO_x in alkaline and acidic environments. We extend the utilization of TiO_x and TaO_x protective coatings to buried photovoltaic junctions and evaluate the respective suitability in terms of optical transparency, conductivity and stability, achieving unassisted solar-water-splitting efficiencies of 15%. Spectroscopic methods, including X-ray photoelectron spectroscopy (XPS), photothermal deflection spectroscopy (PDS), and spectroscopic ellipsometry (SE), are employed to characterize and understand the intentionally introduced defects and correlate electronic properties to the PEC performance. Overall, the presented TiO_x and TaO_x protective coatings represent an important step toward efficient and stable PEC devices, while the demonstrated ALD-based defect engineering generally enables electronic property control of ultra-thin layers.

[1] Bienek, O. et al. Engineering Defects and Interfaces of Atomic Layer Deposited TiO_x Protective Coatings for Efficient III-V Semiconductor Photocathodes. *ACS Photonics* **10**, 3985-3997 (2023). <https://doi.org/10.1021/acsp Photonics.3c00818>

[2] Bienek, O. et al. Suppressing substrate oxidation during plasma-enhanced atomic layer deposition on semiconductor surfaces. *Applied Physics Letters* **124**, 071601 (2024). <https://doi.org/10.1063/5.0182200>

11:45am **AA2-TuM-16 Influence of Atomic Layer Deposition Tin Oxide Properties on the Performance of Perovskite Solar Cells**, *Bhavya Rakheja*, *A. Hultqvist*, Uppsala University, Angstrom Laboratory, Sweden; *T. Törndahl*, Uppsala University, Sweden

Presently, the photovoltaic industry is dominated by single-junction silicon solar cells. As this technology is reaching its theoretical power conversion efficiency limit, the need to develop multi-junction solar cell structures, capable of producing more power per unit area, is rising. Multi-junction solar cells employ a combination of single-junction solar cells, each capable of absorbing complementary parts of the solar cell spectrum. Perovskite solar cells (PSCs) have witnessed tremendous success and their efficiency has improved from merely 3.8% in 2009 to more than 26% in a single-junction solar cell [1]. Moreover, PSCs have also demonstrated great efficiencies when used in tandem configurations with silicon and Copper indium gallium (di)selenide (CIGS) solar cells [1]. All top efficiency PSCs in a tandem structure employ tin oxide (SnO_x), grown using the Atomic layer deposition (ALD) technique, as an electron selective buffer layer. *The material properties of SnO_x are crucial for facilitating good electron transport while mitigating losses.*

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The perovskite layer in a PSC is sandwiched between electron and hole-selective layers. These layers facilitate the transport of the generated charge carriers to their respective contacts. Good energetic alignment between perovskite and charge transport layers is pivotal in maximizing the solar cell's power output. As it has been established in several studies, the bandgap of ALD SnO_x is sensitive to the deposition temperature [2]; making it critical to optimize the band alignment between ALD SnO_x and adjacent layers. Moreover, other process parameters such as the choice of co-reactant can also affect the number of defects and; consequently, impact the power losses in a PSC [3]. In this study, we conduct in-situ quartz crystal microbalance studies and also investigate how the material, optical, and electronic properties of ALD SnO_x evolve as a function of deposition parameters, and their influence on solar cell's device parameters. In particular, we examine the effect of two co-reactants (H₂O and H₂O₂) and their respective doses. By fine-tuning ALD SnO_x deposition parameters and studying their influence on the device performance, one can establish an improved understanding toward the incorporation of ALD SnO_x in PSCs.

References:

- [1] "Best Research-Cell Efficiency Chart." A. Available: <https://www.nrel.gov/pv/cell-efficiency.html>
- [2] M. N. Mullings, C. Hägglund, and S. F. Bent, *Journal of Vacuum Science & Technology A*, vol. 31, no. 6, p. 061503, Jul. 2013, doi: 10.1116/1.4812717.
- [3] T. Hu *et al.*, *Advanced Materials*, vol. 29, no. 27, p. 1606656, 2017, doi: 10.1002/adma.201606656.

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