ALD Applications
Room Auditorium - Session AA-TuM

ALD for Solar Energy Materials I & II

Moderators: Han-Bo-Ram Lee, Incheon National University, Wei-Min Li, Jiangsu Leadmicro Nano-Equipment Technology Ltd.

8:30am **AA-TuM-1 Atomic Layer Deposition Enabling Higher Efficiency Solar Cells**, ***Bram Hoex,*** University of New South Wales, Australia **INVITED**

It is well known that atomic layer deposition (ALD) can synthesize materials with atomic-scale precision. In this presentation we will present some of the recent work in our research group at the University of New South Wales and show that ALD has been instrumental in improving the efficiency of both crystalline silicon as well as thin film solar cells. It will be shown that the deposition of alumina layers at both sides of a silicon solar cell can have some unexpected benefits in addition to significantly simplifying the use of batch ALD in high volume production. The ease of the controlled incorporating dopants in ALD films will be shown to be crucial in improving the properties of NiO for application in hole-selective contact for a wide range of solar cells. And finally, we will show that ALD is currently enabling world-record cadmium-free copper zinc tin sulphide (CZTS) solar cells allowing this technology to cross the important 10 % energy conversion efficiency.

9:45am **AA-TuM-6 Atomic Layer Deposition of $Zn_{1-x}Mg_xO$ and $Zn_{1-x}Mg_xO$:Al as Transparent Conducting Films for Chalcopyrite Solar Cells**, ***Poorani Gnanasambandan,*** *R Leturcq, P Lunca-Popa,* Luxembourg Institute of Science and Technology, Luxembourg*; M Sood, S Siebentritt,* Université du Luxembourg, Luxembourg

We investigate atomic layer deposited aluminium-doped zinc magnesium oxide and zinc magnesium oxide films with varying Mg content as transparent conducting films and as electron extraction layers for chalcopyrite solar cells. Transparent conducting materials are quintessential part of a photovoltaic device. One of the main challenges involves the design of materials combining appropriate band alignment, conductivity and transparency, to be adapted to the absorber in the solar cells. Important parameters for optimization are reduced density of interface defects to avoid recombination, and well-controlled work function and doping. Moreover, the developed materials will require low-temperature vapor phase deposition compatible with the cell processing.

Previous studies on the impact of $Zn_{1-x}Mg_xO$:Al as transparent electrodes and Mg doped ZnO thin films for the window layer of CIGS solar cells employed co-sputtering and electrodeposition respectively [1][2][3]. With the advantage of low temperature and highly conformal thin film growth, we study ALD grown $Zn_{1-x}Mg_xO$ and $Zn_{1-x}Mg_xO$:Al (with x varying from 0.1 to 0.4). We elucidate the effect of doping on the band alignment, electrical and optical properties. We examine how the growth conditions can pave the way towards precise control of Mg content in order to adjust the bandgap and possible growth of $Zn_{1-x}Mg_xO$ film stacks with graded Mg doping. By achieving precise control of Mg content, we aim to investigate the effect of ALD grown $Zn_{1-x}Mg_xO$ films on high-bandgap solar cells based on $CuInS_2$ absorbers.

[1]. Kuwahata, Yoshihiro, and Takashi Minemoto. "Impact of Zn1-xMgxO: Al transparent electrode for buffer-less Cu (In, Ga) Se2 solar cells." Renewable energy 65 (2014): 113-116.

[2]. Wang, Mang, et al. "Electrodeposition of Mg doped ZnO thin film for the window layer of CIGS solar cell." Applied Surface Science 382 (2016): 217-224.

[3]. Inoue, Yukari and Hala, Matej et al. "Optimization of buffer layer/i-layer band alignment" in 42nd IEEE Photovoltaic Specialist Conference (IEEE, New Orleans, 2015), pp. 1

10:00am **AA-TuM-7 Tuning Properties of ALD Oxide and Sulfide Materials for Photovoltaic Applications**, ***Nathanaelle Schneider,*** IPVF-CNRS, France **INVITED**

Atomic layer deposition (ALD) has unique capabilities that makes it very attractive to several applications and in particular photovoltaics (PV).[1] It allows the deposition of pinhole-free thin films, in relative soft conditions (low temperature, low vacuum level), with finely tuned properties (thickness, composition, …) and conformal on high-aspect ratio and/or large surfaces. However, such fine tuning is rarely straightforward and simplistic view of monolayer per monolayer growth is never the case. This is crucial when growing thin films with more than two elements as specific surface reactions can impact the film growth. This is even more pronounced for sulfide multinary compounds due to the higher propensity of cation exchange, greater diffusion rates and unintentional annealing of the labile sulfur.[2]

Such phenomena can be partly controlled by the right choice of precursors,[3] adapting the order of precursor introduction[4,5] or varying the number of successive growth cycles.[6] A deep understanding of the surface chemistry is also necessary and can be achieved by combining modelling (DFT calculations),[7] *in-situ* (such as quartz-crystal microbalance measurements)[2,4,5] and *ex-situ* characterisations (XPS fine profiling, …).[3,6] The aforementioned unique ALD capabilities will be illustrated by examples in PV devices such as $Cu(In,Ga)Se_2$,[6,8] silicon nanowire[5] and perovskite solar cells.[9]

[1] X. Meng, X. Wang, D. Geng, C. Ozgit-Akgun, N. Schneider, and J.W. Elam, Mater. Horiz. **4**, 133 (2017).

[2] H. Le Tulzo, N. Schneider, and F. Donsanti, Materials **13**, 645 (2020).

[3] N. Schneider, M. Frégnaux, M. Bouttemy, F. Donsanti, A. Etcheberry, and D. Lincot, Mater. Today Chem. (2018).

[4] H. Le Tulzo, N. Schneider, D. Lincot, G. Patriarche, and F. Donsanti, J. Vac. Sci. Technol. A **36**, 041502 (2018).

[5] D. Coutancier, S.-T. Zhang, S. Bernardini, O. Fournier, T. Mathieu-Pennober, F. Donsanti, M. Tchernycheva, M. Foldyna, and N. Schneider, submitted.

[6] N. Schneider, L. Duclaux, M. Bouttemy, C. Bugot, F. Donsanti, A. Etcheberry, and N. Naghavi, ACS Appl. Energy Mater. **1**, 7220 (2018).

[7] C. Goehry and N. Schneider, J. Phys. Chem. C **121**, 5871 (2017).

[8] H. Le Tulzo, N. Schneider, D. Lincot, and F. Donsanti, Sol. Energy Mater. Sol. Cells **200**, 109965 (2019).

[9] F.J. Ramos, T. Maindron, S. Béchu, A. Rebai, M. Frégnaux, M. Bouttemy, J. Rousset, P. Schulz, and N. Schneider, Sustain. Energy Fuels **2**, 2468 (2018).

10:30am **AA-TuM-9 Solar Cells Based on Phase-Pure Sb_2S_3 by Atomic Layer Deposition Forming Planar and Coaxial Heterojunctions**, ***Ignacio Minguez Bacho,*** *P Büttner, F Scheler, D Döhler,* Friedrich-Alexander-University Erlangen-Nürnberg, Germany*; C Pointer, E Young,* Lehigh University*; J Bachmann,* Friedrich-Alexander-University Erlangen-Nürnberg, Germany

Interfaces of oxides and heavier chalcogenides layers in thin-film or extremely thin absorber solar cells present defect states at the interface and often a chemical incompatibility which results in dewetting issues. Here, we establish atomic layer deposition (ALD) as a tool to overcome these limitations. ALD allows one to obtain highly pure Sb_2S_3 as light absorber layers, and we exploit this technique to generate an additional interfacial layer consisting of ZnS with thicknesses between 0.2 and 2.0 nm. This ultra-thin layer simultaneously eliminates dewetting, passivates defect states at the interface and slows down interfacial charge recombination. The ability of ALD to generate conformal coatings of porous substrates allows us to generalize the materials system from planar stacks to coaxial heterojunctions based on cylindrical nanostructures. In this system, we optimize length and layer thicknesses in dependence of the carrier diffusion lengths and the light absorption coefficient systematically. We achieve heterojunction solar devices with optimized power conversion efficiency beyond 5.0 %.

10:45am **AA-TuM-10 Metal Oxide Infilling of Quantum Dot Thin Films: Charge Separation, Stabilization, and Solar Cell Formation**, ***Fatemeh Hashemi,*** *R Crisp, J Alkemade, G Grimaldi, N Kirkwood, L Siebbeles, J van Ommen, A Houtepen,* Delft University of Technology, Netherlands

Colloidal semiconductor quantum dot (QD) thin films have various applications in photovoltaic devices and as light emitters. This is due to the controlled variation in their band-gap and ease of fabrication. However, these thin films suffer from instability due to their inherent inclination towards oxidative and photothermal degradation. Thus, to increase the stability of these materials for fabricating QD-based electronic devices, encapsulation or pore infilling processes are necessary. The pore infilling process has been shown to also enhance the conductivity and carrier mobility in the QD thin films. The encapsulation process should provide protection against oxidation without hindering the electron transport properties or causing sintering of the QDs. ALD is an ideal candidate for such a process as is provides excellent control over the growth in the sub-nanometer scale and results in conformal coating even in low temperatures.

We perform ALD of two different metal oxides in atmospheric pressure for the infilling and capping of QD thin films. We examine their effects on the stability and carrier mobility of the coated QD films. InP QDs are chosen because of their potential as light emitters (i.e. phosphors) across the visible spectrum. This is due to their favorable bandgap and high photoluminescence quantum yield. Furthermore, InP QDs exhibit multiple exciton generation allowing for higher solar cell efficiency than traditional materials. However, carrier mobility in QD films was too low for practical applications until now.

We compare the results of amorphous TiO_2 with crystalline ZnO films deposited with different thicknesses. The effects of the capping film thickness of QD film on stability and carrier mobility are studied with spectrophotometry and time-resolved microwave conductivity measurements. We show that the inorganic matrix reduces the size of tunnel barriers within the QD thin film hence increasing the carrier mobility through the film without causing sintering of the QDs. Furthermore, our results confirm that the stability of QD thin films is strongly improved when the inorganic ALD coating is applied. The ALD encapsulation process would open up the possibility of fabricating robust InP QD thin films for many optoelectronic devices.

11:00am **AA-TuM-11 ALD of Al_2O3 on Perovskite Solar Cells: Role of Active Interfacial Engineering**, *S Ghosh, N Mahuli,* ***Shaibal Sarkar,*** Indian Institute of Technology Bombay, India

Atomic Layer Deposition of ultrathin Al_2O_3 on hybrid perovskite solar cells drew significant attention due to the considerable improvement in the overall device stability. In our laboratory, with intermittent current-voltagemeasurements, the coated devices show the value of T80>7500 hours under ambient conditions. Subsequently, these coated devices are found highly stable when measured in a cyclic manner for 7 days, replicating the real-life day-night sequences. Such *encapsulation* is found very effective as an oxygen barrier-layer and water-impermeable membrane; hence contribute to the overall stability of these devices.

In this presentation, I would like to emphasize on our experimental findings, subsequently supported by device simulation, which undoubtedly reveals that the perovskite-spiroOMeTAD interfacial band-structure play a detrimental role in initiating the degradation processes in the pristine devices (device structure). We try to provide a comprehensive insight depicting an apparently non-trivial active phenomenon resulted due to the ALD grown Al_2O_3 layer that supposedly be a passive component of the entire device stack. Favored electronic modification of the spiro-OMeTAD/perovskite interface resulted due to the Al_2O_3 ALD provides better charge extraction and lesser ionic accumulation, unlike the unencapsulated devices, and hence offers better performance stability. Our study indicates that essentially the ionic accumulation triggers the device degradation that is eventually followed by materials degradation.

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