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Spatial Atomic Layer Deposition for large-area and flexible electronics

Large-area and flexible electronics could potentially benefit tremendously from the advantages of ALD over other deposition techniques. These applications, however, require deposition on large areas and on flexible substrates at deposition rates unattainable with conventional ALD. For this purpose, spatial ALD has been developed, which relies on spatial instead of temporal separation of precursor exposures. In this article, the pivotal role of the Dutch spatial ALD community is highlighted, and several examples of current challenges in spatial ALD are discussed.

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Spatial Atomic Layer Deposition

In the past decade, ALD has become a mainstream deposition technique in the microelectronics industry because of its excellent performance in terms of film quality, thickness control, uniformity and step coverage. For the same reasons, the use of ALD is of interest for other application areas as well, including large-area and flexible electronics. Examples include buffer, transport and conductive layers in thin-film solar cells, thin-film encapsulation and oxide semiconductors in OLED displays, as well as passivation layers and electrolytes for (solid state) Li-ion batteries. Although the examples mentioned are quite diverse, they share common aspects that clearly differentiate them from microelectronics; in order to keep the costs of mass production low and the throughput high, these applications require deposition on large areas or on flexible substrates at high deposition rates. Mainstream ALD equipment is typically designed for processing 300mm wafers, either in single-wafer or in batch mode, where typical deposition rates are in the order of nanometers per minute. It would be very difficult to adapt these tools to handle the very large substrates used in e.g. OLED display manufacturing (up to 1.5m x 1.85m), or roll-to-roll systems that can handle 1.3m wide foils at tens of meters per minute web speeds. Therefore, a different way of doing ALD than the usual time-sequenced, low pressure methods currently employed is required. One possible solution is Spatial Atomic Layer Deposition (SALD) [1].

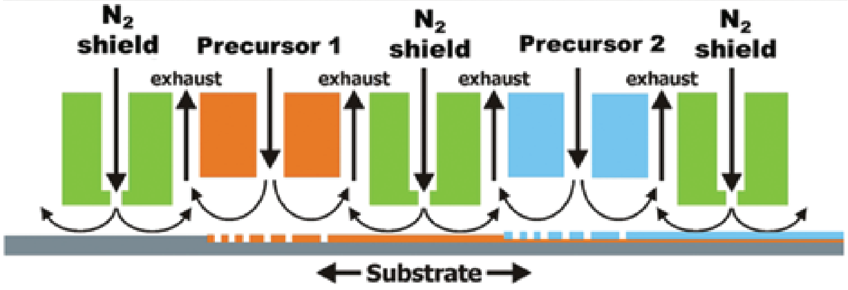

Figure 1 Schematic drawing of TNO's spatial ALD reactor concept, where the two precursor zones are separated by inert gas shields. The two half-reactions will take place subsequently to form an ALD monolayer, by moving the substrate underneath the reactor. Thicker layers are obtained by repeating the movement.

Just as in conventional ALD, in Spatial ALD the substrate is exposed to individual precursors sequentially, but instead of separating precursor steps in time, the precursors are spatially separated in precursor zones through a so-called injector or deposition head. To prevent mixing of precursors and to avoid CVD, these zones are confined by inert ambient zones, for instance a vacuum, a gas shield, or a gas bearing [1]. The ALD cycle is completed by moving the substrate with respect to the injector head from one zone to the other, which is repeated until the desired thickness is achieved. Moving the substrate or injector can be done by e.g. translation or rotation. With the reaction rates generally being very high and without the need for purging a reaction chamber between the precursor steps, the effective deposition rates achieved can be in the nm/s range [2], allowing high-throughput processing and the deposition of relatively thick films. Furthermore, spatial ALD does not require a low-pressure environment and can be operated at atmospheric pressure. The injector head itself can be engineered to very large sizes allowing for deposition on large area substrates.

Spatial ALD in the Netherlands

Although the spatial ALD concept was already described in an early ALD patent

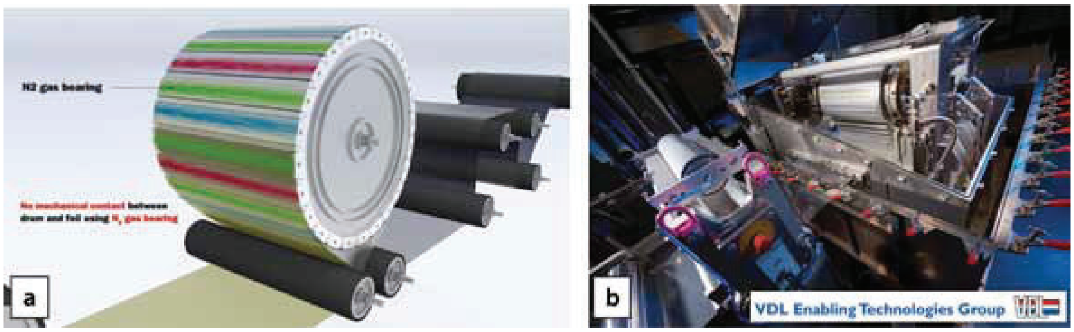

Figure 2 a) Schematic drawing of TNO's atmospheric pressure roll-to-roll spatial ALD concept, where the substrate foil floats without contact on the gases flowing outwards from a drum-shaped SALD injector. A film is deposited on the the foil by moving the foil and rotating the drum. b) Photograph of a 50cm wide roll-to-roll SALD tool built by VDL ETG for applying ZnOS buffer layers on flexible CIGS solar cells [15].

by Antson and Suntola in 1977 [3], the concept was independently reinvented and further developed by several groups in the past years [1]. At present, a growing number of groups and companies worldwide are developing spatial ALD technology for a wide range of applications. The Netherlands has always been a hot spot for spatial ALD R&D. TNO (via its open innovation institutes Holst Centre and Solliance) has been active in R&D programs on spatial ALD since 2009 [4,5]. This resulted in numerous collaborations with industry and academia. Some highlights include the spin-off companies SoLayTec [6] (2010; SALD equipment for c-Si solar cells) and SALDtech [7] (2018; SALD equipment for OLED displays), collaborations with Dutch equipment companies VDL and Meyer Burger (both roll-to-roll SALD equipment) and Smit Ovens (sheet-to-sheet equipment), and collaborations between TNO and TU/e on more fundamental aspects of spatial ALD. Other Dutch spatial ALD activities include Levitech (SALD equipment for c-Si passivation; spin-off from ASM International), SALD (a recent spin-off from SoLayTec) and TU Delft (SALD on powders) [8].

Current challenges in spatial ALD R&D

Whereas the first applications of spatial ALD were based on simple oxides such as Al_2O_3, TiO_2 and ZnO, and on substrates of relatively small sizes (up to 300mm), today the use of spatial ALD for increasingly more demanding applications and complex materials is being explored. In many cases, the literature shows that the added value of introducing ALD layers has already been demonstrated on lab-scale for conventional ALD, but the scale-up towards industrial manufacturing still has a long way to go. One example are OLED displays where quaternary oxides like indium-gallium-zinc oxide (IGZO) have to be deposited with less than 2% variation in stoichiometry and thickness on more than $2m^2$ glass plates at takt times of around 60s [9]. Other examples include perovskite solar cells, where ALD layers have to be deposited in a roll-to-roll mode on very sensitive substrates and at low deposition temperatures [10], and roll-to-roll ALD in highly porous substrates, such as separator membrane foils [11].

Depositing compound materials like IGZO and LiPON involves much more complex reactions than e.g. Al_2O_3. Developing SALD processes for these materials requires careful selection of suitable precursors and an in-depth investigation into the reaction kinetics. The same holds for SALD in high aspect ratio 3D and porous substrates, where the precursor reactivity and diffusivity mainly determine how deep and how fast pores can be coated [12]. Furthermore, temperature limitations imposed by the substrate, e.g. in case of polymer materials, are common in flexible electronics. For this and other reasons, the development of (atmospheric pressure) plasma-assisted SALD is actively being pursued [13].

Next to the evolution towards more complex processes and materials, also patterned deposition is gaining more attention. Because of the excellent step coverage of ALD, patterned deposition through the use of shadow masks is not feasible as it will lead to undesirable deposition under the mask. Therefore, SALD-based maskless direct patterned deposition techniques, including area selective deposition, are being developed [14].

Finally, new SALD equipment concepts are being developed that can be scaled to the very large substrate sizes and high throughputs used in industry, while keeping the investment and operational costs low. The performance of a spatial ALD tool is determined by a complex interplay of gas flows and pressures, precursor-substrate interactions, diffusive and convective transport of precursors and reaction products, substrate movement and plasma radical generation and recombination (in case of plasma SALD). Modeling, in particular fluid dynamics modeling, is an essential tool in developing and designing new SALD reactor concepts and new models are currently being developed.

Conclusions

Since its re-invention more than ten years ago, spatial ALD has been a subject of growing activities and importance in both the R&D community and industry worldwide. It can be expected that spatial

ALD will be adopted by more and more industries in the coming years, which requires continuous research and development, from a fundamental and from an applied research point of view. The SALD community in the Netherlands has played a pivotal role in the development of SALD because of its unique SALD knowledge value chain; ranging from fundamental research at the TU/e via applied research at TNO, up to the many equipment companies who build and sell SALD equipment to industry. This has resulted in several successes so far, like SALD for c-Si passivation and roll-to-roll SALD for barrier foils. If the Dutch SALD community keeps working together and even further strengthens their ties, it can maintain its worldwide leading position and the future for Dutch SALD will be bright and sunny.

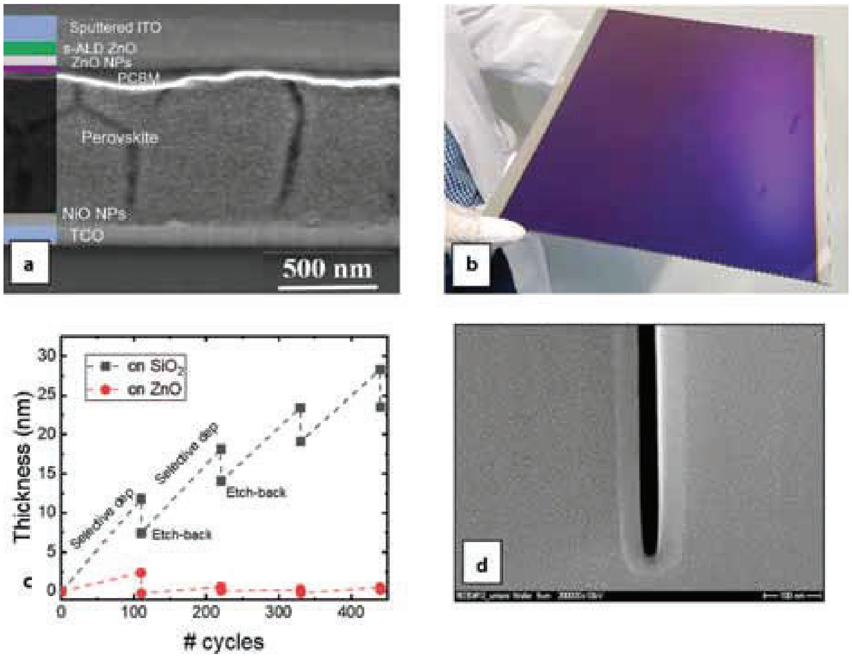

Figure 3 Examples of results of advanced spatial ALD R&D conducted at TNO. a) Cross section of a perovskite solar cell with a 45nm ZnO buffer layer [16] b) InGaZnO film (100nm) deposited by plasma-assisted spatial ALD on a 30cm x 40cm glass plate with a thickness non-uniformity of 1%. c) Area selective spatial ALD of SiO_2 on SiO_2 (growth area) vs. ZnO (non-growth area). Interleaved etch steps are used to selectively deposit thick films [14]. d) SEM cross-section of the bottom of a 9µm deep and 90nm wide pore (aspect ratio 100:1), coated with SALD Al_2O_3 with 100% step coverage

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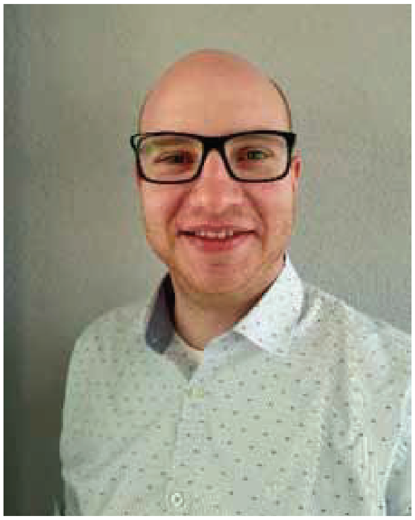

Paul is leading the spatial ALD R&D activities at TNO - Holst Centre, developing spatial ALD technology for applications in large-area and flexible electronics. He is also part-time associate professor at the TU/e and CTO of TNO's spin-off company SALDtech.