Equipment for atmospheric, spatial Atomic Layer Deposition in roll-to-roll processes

R. Knaapen\textsuperscript{1}, P. Poodt\textsuperscript{1}, R. Olieslagers\textsuperscript{1}, A. Lankhorst\textsuperscript{1}, M. van den Boer\textsuperscript{1}, D. van den Berg\textsuperscript{1}, A. van Asten\textsuperscript{1}, F. Roozeboom\textsuperscript{1,2}
\textsuperscript{1}TNO, Eindhoven, Netherlands
\textsuperscript{2}Eindhoven University of Technology, Netherlands

-Raymond.Knaapen@tno.nl-

Abstract
A novel type of reactor has been designed for atmospheric atomic layer deposition (ALD) on flexible substrates. In the reactor, a flexible substrate slowly advances around a fast rotating drum. Gas bearing technology is used to prevent physical contact between the flexible substrate and the drum, and to separate reactants to enable a spatial ALD process. With a substrate speed of 1 m/min and drum rotational speed of 5 Hz, a layer of 100 nm of Al\textsubscript{2}O\textsubscript{3} can be applied in a continuous roll-to-roll process at deposition rates of 1 nm/s. Higher speeds to ~2 nm/s are foreseen to be possible.

1 Introduction
There are many types of thin film deposition techniques, such as Physical Vapour Deposition (PVD), Chemical Vapour Deposition (CVD) and Atomic Layer Deposition (ALD). ALD has a number of unique properties like high conformality over challenging substrate topography, superior layer quality and thickness control down to Ångström level. The deposition rate, however, is very low in conventional, \textit{temporal} ALD reactors (~1 nm/min), which makes cost of ownership high. To achieve high throughput and to reduce costs, a \textit{spatial} version of ALD has been developed. Whereas in conventional ALD, reactants are dosed separated in time using a purge or pump step, in spatial ALD reactants are dosed simultaneously and continuously at different physical locations. The separation of reactants is done by using gas bearing technology. As no purging step is needed in spatial ALD, the process can be operated at much higher speeds, limited by layer deposition chemistry rather than pumping times. Thus, deposition rates exceeding 1 nm/s have been
reported for spatial atmospheric ALD of Al₂O₃ [1]. This has led to the development of high throughput, industrial scale ALD tools for surface passivation of crystalline silicon solar cells.

Because of the increased throughput and decreased cost levels, new application fields are opening up for spatial ALD, such as flexible electronics, including system-in-foil, flexible displays, OLEDs and solar cells. Examples of layers are transparent oxide (semi)conductors (e.g. ZnO) and moisture permeation barriers (e.g. Al₂O₃).

![Spatial ALD principle for deposition of Al₂O₃ on a moving substrate using trimethylaluminium (TMA) and H₂O.](image)

Figure 1: Spatial ALD principle for deposition of Al₂O₃ on a moving substrate using trimethylaluminium (TMA) and H₂O.

2 ALD on flexible substrates

Substrates in existing ALD applications are usually flat and rigid, like silicon wafers or glass plates. Performing spatial ALD on these substrates involves e.g. reciprocating substrates or substrates rotating under a flat ALD injector head.

For deposition on flexible substrates, a new type of atmospheric, spatial ALD reactor has been designed. Instead of a flat ALD injector head, a rotating drum is used to supply the reactant gases to slots at the peripheral surface of the drum, parallel to its rotation axis. A flexible substrate is transported around the drum surface, where gas bearings are used to separate the foil from the drum as well as separate the different reactants, see Fig. 2. The foil being contactless enables the drum to rotate at high speed while the foil itself is slowly advancing, such that every part of the foil surface comes into contact with a predefined number of reactant cycles. Each individual reactant pair cycle will deposit one monolayer of e.g. Al₂O₃. In the current design, the drum has six pairs of reactant slots at its outer surface. Thus, when the drum rotates at a frequency of 5 Hz, the number of reactant pairs per second is approximately 30, for low foil traversing speeds of approximately 1 m/min. When the foil covers 50% of
the drum surface, an approximately 100 nm thick layer can be applied in a single pass continuous roll-to-roll process.

![Diagram](image)

Figure 2: Illustration of a flexible substrate moving clockwise over a counterclockwise rotating drum with three pairs of reactant slots at its outer surface.

3 Reactor design

One of the challenges in the rotating drum design is the bearing of the foil around the drum. This bearing must ensure that the foil does not touch the drum and at the same time must separate the reactant gases. The same approach is used as drawn in Fig. 1, where reactant gases are separated by bearing gas. A reactant gas is first fed through a divider, which ensures a homogeneous concentration distribution over the entire drum width. Next, the reactant gas flows through a restriction before encountering the foil surface. The flow continues to outlet collector channels, which enable exhaust with a low flow restriction.

Another challenge is the gas supply to and exhaust from the rotating drum, and interrupting the chemical reaction at all parts of the drum surface that are not covered with foil. Both issues are addressed using a contactless gas feed-through at the flat sides of the drum. This feed-through uses a similar gas separation method as used around the drum in the ALD deposition region. Each reactant is supplied to a circular channel in the feed-through disc. Next to the reactant channel there is an exhaust channel and a gas bearing channel, see also Fig. 3. The reactant will flow into the drum through a channel that is aligned to the reactant channel in the feed-through disc. To block the reactant supply at the drum part that is not covered with foil, the circular channel is interrupted over the corresponding rotation angle. In the interrupted part, a flush gas is supplied to remove reactants from the reaction chambers. Only a small part of each reactant will directly leak to the exhaust, through
the gas bearing gap, where it will admix with bearing gas. Reactants are exhausted through a scrubber to be neutralized. Furthermore, the drum is heated from the centre by a radiation heater. Typically, the process temperature will be around 120 °C.

Figure 3: Foil bearing around the drum and contactless gas feed-through to the drum.

Figure 4: Prototype design of a spatial ALD roll-to-roll reactor with 300 mm diameter drum, which will be incorporated in a roll-to-roll line.

References: