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High rate low pressure plasma-enhanced chemical vapor deposition for barrier and optical coatings

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ABSTRACT

Two types of plasma-enhanced chemical vapor deposition (PECVD) technologies were developed. The main benefits of these DC-magnetron based PECVD (magPECVD) and hollow cathode arc driven PECVD (arcPECVD) are their low process pressure in the range of 0.1 to 5 Pa and the high deposition rate up to 400 nm m/min for magPECVD and 3000 nm m/min for arcPECVD. Both processes were used in combination with inline sputtered layers to produce layer stacks in roll-to-roll coaters on polymer webs. Optical layer stacks made with these technologies showed nearly no layer stress. The mechanical robustness of permeation barrier layers was increased by implementation of interlayers made by these PECVD methods. Necessary hardware was developed for industrial applications with web widths of 1.7 m for magPECVD and 2.85 m for arcPECVD.

1. Introduction

Plasma-enhanced chemical vapor deposition (PECVD) is a well known coating technology being used world-wide especially for batch processing. Unfortunately commonly used rf or microwave driven PECVD needs relatively high process pressure in the range of 10 to 100 Pa. This high process pressure makes it difficult to combine these PECVD methods with other PVD technologies like evaporation or sputtering within one vacuum coating machine to be run simultaneously. Otherwise such a combination of PECVD with other coating technologies would be beneficial for many purposes like build up of permeation barrier stacks on polymer substrates. Unlike thermal chemical vapor deposition, PECVD is a low temperature process. Therefore this process can be used to deposit layers on thermally sensitive substrates like polymers or thin metal foils. The PECVD process runs in vacuum and thus runs in a clean and well defined environment. There is a broad choice of precursors available to deposit different layer materials. In combination with different reactive gases also chemical compounds can be formed during layer growth. A wide range of process variables exists to tune the process and the resulting layers. For instance plasma power, plasma frequency and process pressure can be adjusted. To overcome the limitations of common PECVD technologies regarding process pressure two different types of PECVD were developed and will be introduced.

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2. Magnetron driven PECVD — magPECVD

Magnetron sputtering is a well established and worldwide known PVD technology to produce single layers and multi layer stacks of high quality. One of the major advantages of magnetron sputtering is the superior layer thickness homogeneity over large substrate width. The technology is used for architectural glass coating and also for coating of polymer webs with coating width up to 4.5 m. Long term stability of reactive processes at high deposition rate is achieved using a closed loop control of the reactive gas inflow [1]. When equipped with an additional precursor inlet as shown in Fig. 1, a (dual) magnetron system may also be used as plasma source for a PECVD process.

One important feature of this technology is the low process pressure in the range of 0.3 to 3 Pa. The ratio between precursor to reactive gas flow rate can be adjusted to modify the layer composition instantly during the coating process (see later in this paper). The deposition rate of this magnetron driven PECVD (magPECVD) depends on this ratio and on the precursor flow as well. Dynamic deposition rates up to 400 nm m/min were achieved by using HMDSO (hexamethyldisiloxane) as precursor using precursor rich conditions. A deposition rate of 200 nm m/min was proven to be long term stable over 8 h. This could be shown with a device made by Fraunhofer Institute for Electron Beam and Plasma Technology (FEP) in Dresden, Germany, for an industrial customer for a web width of 1.7 m. An additional benefit is the usage of one piece of hardware with an option to run two processes. With the use of the precursor one drives a PECVD device. Without the precursor the device may be used like a common reactive sputtering system.

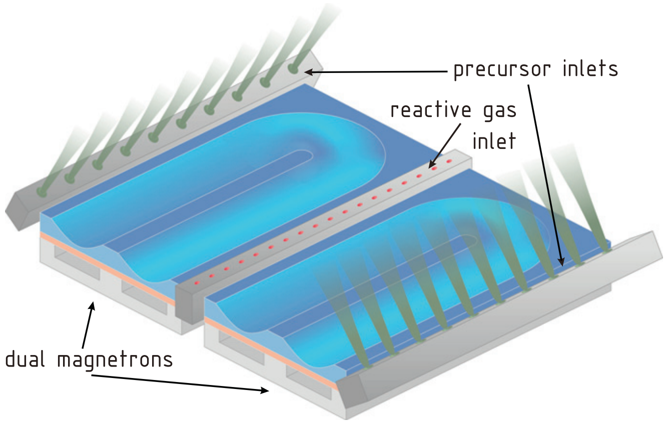

Fig. 1. Typical arrangement of magPECVD device, consisting of a dual magnetron system, a precursor inlet and a reactive gas inlet.

3. Hollow cathode arc PECVD — arcPECVD

A very intensive plasma can be delivered by hollow cathode arc discharges [2]. Respective plasma sources are used within the plasma-enhanced reactive deposition of polymer webs with alumina for permeation barrier coatings for the packaging industry. Such plasma systems were equipped with precursor inlet to setup a device for high rate PECVD deposition. A typical arrangement of hollow cathodes for PECVD deposition is shown in Fig. 2. The feasibility of the so called arcPECVD process has been proven in a pilot-type roll-to-roll coater for 650 mm web width. Four modular hollow cathode arc sources were used. Such plasma sources have already been installed into several industrial web coaters up to 2.85 m web width for plasma-enhanced deposition of alumina (see device photo in Fig. 3). Therefore, a scale up of the arcPECVD process from the pilot coater level (650 mm) to wider webs (2.85 m and beyond) seems feasible without any compromise in deposition rate or process stability.

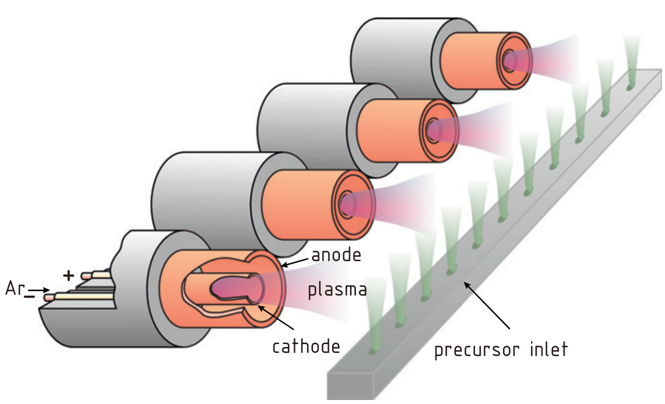

Fig. 2. Typical setup of arcPECVD device, consisting of one or more hollow cathode arc sources and a precursor inlet.

The dynamic deposition rate of an arcPECVD process was investigated more in detail. There is a linear dependency between the precursor flow rate and the deposition rate as seen in Fig. 4. A maximum of 2700 nm m/min at a flow rate of 1000 sccm HMDSO was reached. The variation in deposition rate at HMDSO flow of 500 and 750 sccm is due to change in plasma power and/or additional oxygen flow. The arcPECVD process follows the same "one hardware–two processes"-principle as the magPECVD process. Beside the arcPECVD by itself one can also use the hardware for plasma enhancement of reactive or non-reactive evaporation. This approach is typically used in the high speed alumina coating of polymer webs for the packaging industry [3].

4. Comparison of PECVD methods

Some commonly used PECVD methods were listed in the introduction. They are used very often in batch coaters like systems for wafer coating or coating of eye glasses. But their use in roll-to-roll machines is not very common. A main drawback of these PECVD methods, such as hf or microwave PECVD, is their high process pressure up to 100 Pa (e.g. [4]). In contrast to that magPECVD and arcPECVD need only a pressure in the range of 0.1 to 5 Pa. Due to the lower pressure the combination with other PVD technologies like evaporation or sputtering is easily possible. Within a multi chamber machine PVD and PECVD modules could be run simultaneously with only low requirements regarding gas separation between the coating chambers. Another advantage is the higher deposition rate of magPECVD and arcPECVD in comparison to the others. These deposition rates fit very well to the one which typical PVD methods reach. An inline combination of sputtering and magPECVD or a combination of evaporation and arcPECVD is possible.

	MagPECVD	ArcPECVD	HF-PECVD	MW-PECVD
Typical frequency	10–50 kHz bipolar pulsed	DC	13.56 MHz	2.45 GHz
Process pressure	0.3–3 Pa	0.1–5 Pa	1–20 Pa	5–100 Pa
Deposition rates	20–400 nm m/min	500–3000 nm m/min	10–200 nm m/min	10–100 nm m/min
Remarks	Industrially proven for wide webs, units commercially available		Pressure range different to PVD	

5. Application examples

5.1. Layer composition

A typical feature of PECVD processes is the variability in atmosphere composition. The ratio between the needed precursor flow and an optional reactive gas flow can be adjusted to initiate a change in the layer composition as shown in Fig. 5. Layers made by magPECVD with different ratios between the flow of precursor HMDSO and reactive gas oxygen were investigated by X-ray photoelectron spectroscopy (XPS). With an increase of the HMDSO fraction more and more carbon can be found in the layer. An increased organic modification of the silica layer is observed. With a change of the layer composition other layer properties are changed as well. Fig. 6 shows the elastic modulus and the hardness of such layers in dependence of the precursor to reactive

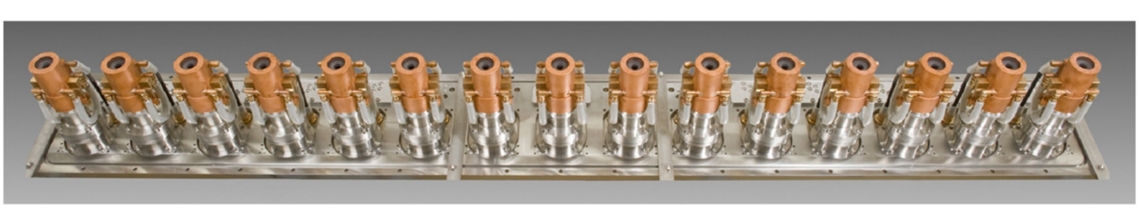

Fig. 3. Arrangement of a number of hollow cathode arc (hca) sources to apply plasma enhancement over a width of 2.85 m; hca-sources are mounted on a rectangular flange right to be installed in web coating machines.