Discharge-Based Electron Beam Sources for Plasma-Activated EB-PVD

B. Zimmermann[1], G. Mattausch[1], F. Fietzke[1], B. Scheffel[1], F.-H. Rögner[1],
Chr. Metzner[1]

[1]*Fraunhofer Institute for Organic Electronics, Electron Beam and Plasma Technology FEP,*
Winterbergstraße 28, 01277 Dresden, Germany
Email to corresponding author: burkhard.zimmermann@fep.fraunhofer.de

Discharge-based electron sources are flexible high-power tools for vacuum high-rate coating. They open the applicability of PVD for a wider field of processes due to their potential to enable tailoring film properties or significantly lowering process cost. Therefore, a magnetically enhanced hollow cathode arc plasma source and a discharge-based electron beam gun have been developed and will be presented here. Moreover, aspects of parallel operation for EB-PVD will be discussed.

1. Introduction

Gas discharge-based electron beam (EB) devices represent high-power and low-cost tools for a variety of processes required in vacuum high-rate coating. These include the fields of substrate pre-treatment, electron beam generation for materials evaporation, plasma activation in PVD, and post-treatment steps.

Low-voltage electron beam (LVEB) sources utilizing a hollow-cathode arc (HCA) discharge have been proven as versatile, compact devices for large-volume high-density plasma generation [1]. Due to the high achievable ion current density, the main application is still related to plasma activation of EB-PVD processes allowing for combining high-rate coating with well-defined film properties such as morphology, hardness, and density [2]. However, in the last years, the operational stability and reliability has been significantly improved by the introduction of magnetical enhancement as well as the usage of pulsed power supplies, and its scope has been extended to further application fields, e.g. substrate pre-treatment by sputter etching, hollow cathode-assisted reactive magnetron sputtering [3], or the very fast deposition of amorphous hydrogenated carbon (a-C:H) utilizing the new arcPECVD process [4].

Whereas the HCA plasma activation considerably expands the applicability of EB-PVD processes in various fields due to the adjustability of film parameters and the versatility of the plasma source, the complexity of conventional EB gun technology represents a strong restriction especially in cost-sensitive fields. Therefore, a discharge-based EB gun has been developed, which utilizes a high-voltage glow discharge (HVGD) in front of the cathode to stimulate electron generation by secondary as well as thermionic emission [5].

In this paper, the discharge principles of both discharge-driven electron sources as well as application examples are presented. Furthermore, aspects of parallel operation will be discussed.

2. Hollow cathode arc plasma source

The HCA is an established tool in PVD technology to generate dense plasmas in order to excite or ionize the vapor; in combination with a substrate bias voltage, the energy of the film-forming particles can be tailored and the optimum growth conditions can be met. Furthermore, in reactive processes, the reactivity of the gas is enhanced by ionization and dissociation processes.

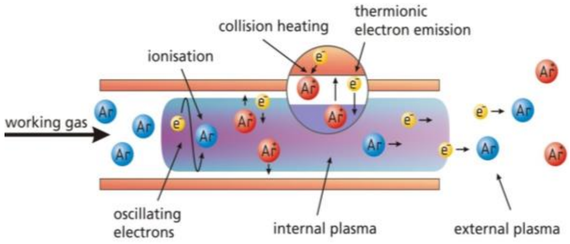

Figure 1: Mechanisms of the arc discharge within the hollow cathode tube

A HCA device consists of a refractory metal tube which is flown through by a working gas, typically argon (see figure 1). After ignition, the internal plasma is established within the tube; ions reaching the plasma sheath are accelerated by the cathode fall voltage onto the cathode tube. The ion bombardment heats up the cathode to high temperatures; consequently, electrons are thermionically emitted and accelerated within the cathode sheath into the plasma in order to sustain it by impact ionization. Electrons traveling through the internal plasma without collision are reflected by the opposite cathode wall, leading to oscillating electrons and enhanced ionization efficiency.

A fraction of electrons from the cathode wall as well as from the internal plasma drifts through the cathode orifice into the process chamber towards the anode, forming the so-called LVEB with energies of several tens of eV and generating the external plasma for technological application.

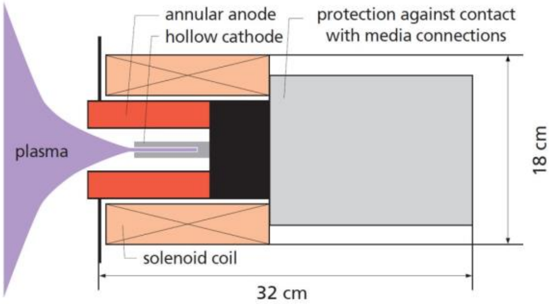

Figure 2: Coaxial arrangement of cathode, anode, and solenoid coil and dimensions of the HCA plasma source

Conventional HCA sources suffer from the drawbacks of high gas throughput (100 sccm and more), time-consuming and unreliable ignition procedures (cathode pre-heating by tungsten spirals with limited liftetime), and an inhomogeneous plasma distribution within the chamber. Therefore, a magnetically enhanced HCA source has been developed, which is operated with a pulsed power supply (figure 2) [3].

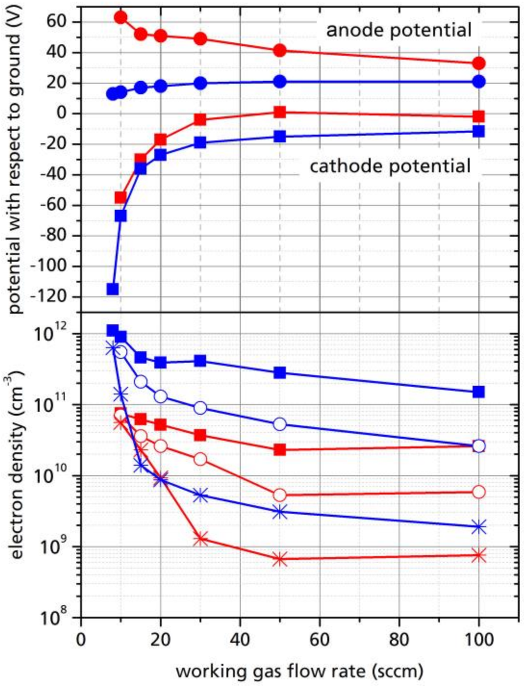

Figure 3: Cathode and anode potentials as well as the electron densities at a distance of 30 cm (squares), 70 cm (circles), and 125 cm (stars) from the HCA source for chamber pressures of 0.1 Pa (red) and 1.2 Pa (blue), respectively, at a discharge current of 100 A.

A solenoid coil is arranged coaxially around the cathode tube and the annular anode. As a consequence, the magnetic field lines are axial within the internal plasma and spread out into the chamber. In combination with high voltage pulses of 1400 V, the HCA discharge ignites within seconds due to short cathode arc spot pulses rotating in the magnetic field resulting in fast heating of the tube and eventually in the desired diffuse arc mode.

During stable operation, the working gas flow rate can be reduced, as the electron gyration around the magnetic field lines ensure sufficient ionization rates within the internal plasma. Furthermore, the cathode potential drops to large negative values elevating the energy of the LVEB electrons, leading to a strong rise of electron density and plasma coverage within the chamber. This is shown in figure 3, which contains results from spatially resolved Langmuir probe as well as optical emission measurements [1]. Typical discharge parameters are arc currents of 50 to 200 A at voltages between 30 and 150 V allowing for plasma densities of more than 10^{12} cm^{-3}.

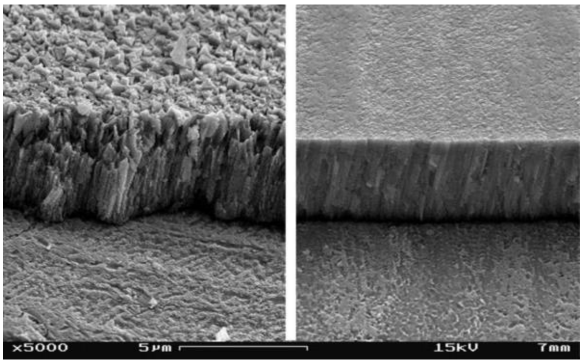

Figure 4: SEM pictures of EB-PVD-deposited YSZ without (left) and with (right) HCA plasma activation.

As an application example, reactive plasma-activated EB-PVD of dense yttria-stabilized zirconia (YSZ) layers on steel for electrolyte layers in solid oxide fuel cells is shown [6]. After HCA plasma sputter etching of the substrates, layers with thicknesses around 5 µm with growth rates of 40 nm/s have been realized by EB evaporation of 8 mol% Y_2O_3-ZrO_2 and additional oxygen injection. During HCA activation, a substrate ion current of up to 25 mA/cm^2 has been measured. The plasma-activated coatings revealed a remarkably denser, smoother microstructure (figure 5), a hardness increase from 5 to 20 GPa, and a texture change of the cubic phase from {111} to {100} in comparison to the sample without plasma.

3. Discharge-based EB gun with hybrid cathode

Electron beam sources provide the highest coating rates for industrial-scale processes and deposite layers of excellent uniformity and purity. Conventional electron beam sources employ heated refractory metal cathodes as thermionic emitters. This principle causes a certain sophistication of the source design and complexity of the vacuum as well as electrical supply system. In order to overcome this drawback, electron sources with cold cathodes and greatly simplified control and supply schematic have been developed [7]: a HVGD is sustained in the cathode compartment of the EB gun providing ions, which are accelerated onto the cooled cathode releasing electrons by secondary electron emission when hitting the surface. The electrons are accelerated across the cathode sheath and form the beam (figure 5 left).

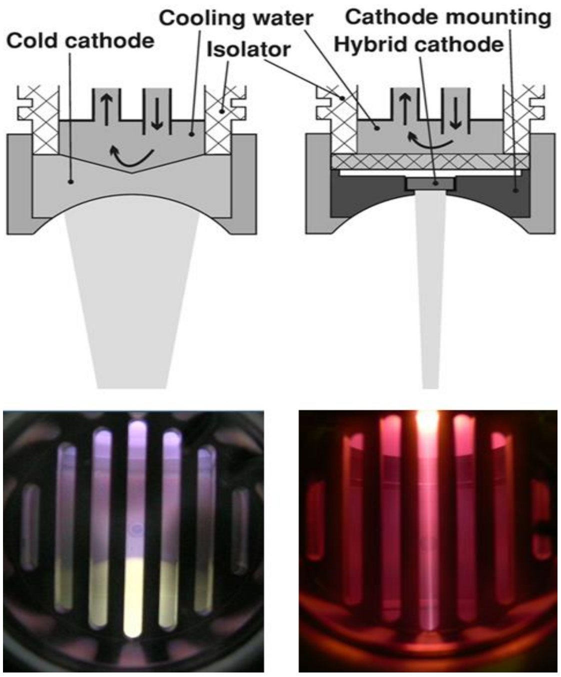

Figure 5: Schematic of the internal setup of a cold cathode (top left) and of a hybrid cathode (top right) and the respective HVGD plasmas and electron beams (bottom)

The HVGD works at a pressure of around 2…5 Pa. The working gas is introduced into the cathode compartment and pumped away by the process chamber vacuum system through the beam guiding tube. Consequently, differential evacuation of the gun can be omitted. Moreoever, since no auxiliary cathode heating system is utilized, the cathode arrangement is remarkably more compact and simplier to maintain, the high voltage power supply does not require heating power supplies floating on high voltage, and the number of high voltage cables is reduced from 3 to 1. However, drawbacks of this lucrative principle are the elevated gas load in the process, arcing issues in the presence of reactive gases, and the larger beam spot reducing the beam power density at the process site.

In order to combine the excellent beam qualitiy of conventional EB guns with the advantageous cold cathode principle, a plasma-based EB source with hybrid cathode has been developed (figure 5 right). A thermally insulated block cathode typically made from lanthanum hexaboride (LaB_6) is mounted with a graphite holder into the cathode compartment. Ions from the HVGD impinge the cathode resulting in secondary electron emission as well as cathode heating. As soon as the latter provides enough power, thermionic electron emission contributes to the beam electron generation; the beam current is controlled by the working gas flow rate at constant voltage.

A simple model considering the power balance of ion heating, radiation cooling, heat conductance and electron emission cooling, as well as the current balance of ions and electrons from thermal and secondary electron emission, allows for a heuristic description of the hybrid cathode working principle. By additionally estimating the ionization in the bulk and in the sheath of the HVGD by the emitted electrons, a qualitative correlation of the total discharge current and the working pressure or gas flow rate could be found and fitted to aquired data (figure 6).

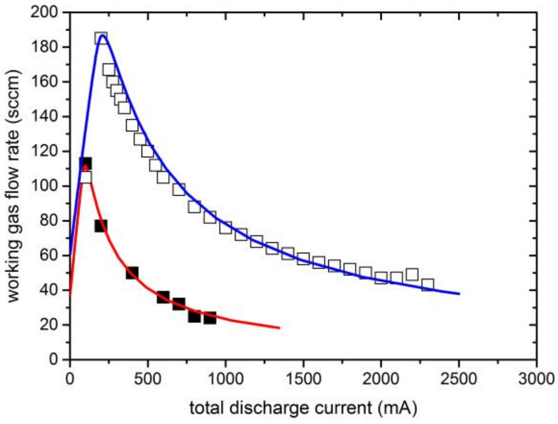

Figure 6: Working gas flow characteristics for hybrid cathodes operated at 30 kV with hydrogen (closed squares) and at 40 kV with helium (open squares), with the respective fits originating from balance considerations

At low currents, the secondary electron emission dominates and the working gas flow rate as well as the cathode temperature increase significantly. With increasing current and ion bombardment, the cathode reaches thermionic emission temperature.

As described by the Richardson-Dushman equation, the thermionic electron emission density is very sensitive to small temperature changes. As a consequence, only little additional ion heating leads to considerably enhanced electron emission and eventually ionization rate in the HVGD, resulting in a reduced need of working gas flow rate again. An increase of the gas flow rate occasionally observed at high beam currents and lower acceleration voltages can be attributed to space charge-limited electron emission [5]; however, the gas load remains far below typical values for cold cathodes. Generally, calculations revealed an ion current fraction of less than 5%, decreasing with increasing discharge current or acceleration voltage.

Compact hybrid cathode EB guns with a power up to 120 kW (at 40 kV) have been realized and applied e.g. for the metallization of polymers with copper in FEP's roll-to-roll coater novoFlex® 600 or for deposition of aluminum alloys onto metal strips.

4. Combined operation for plasma-activated EB-PVD processes

After having developed and characterized both the HCA plasma source and the HVGD hybrid cathode EB gun, present work is being dedicated to the combined operation for plasma-activated EB-PVD. Being mounted on a process chamber, the single devices are coupled by the pressure and the ionization of the process gases.

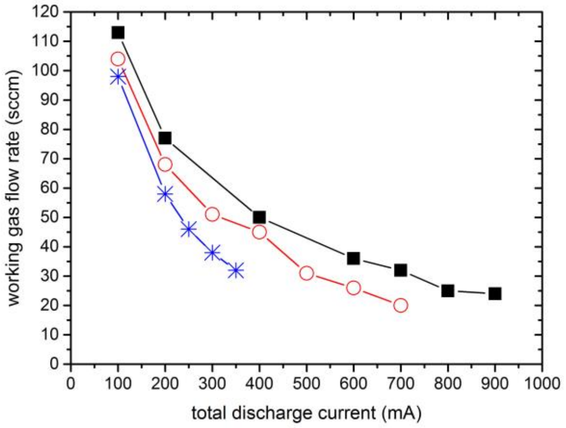

Figure 7: Working gas flow characteristics for argon chamber pressures of < 0.1 Pa (black squares), 0.2 Pa (red circles), and 0.5 Pa (blue stars), recorded with a laboratory model of a 30 kV/30 kW hybrid cathode gun at 30 kV with the working gas hydrogen.

The HCA source has been shown to work properly in a wide pressure range from < 0.1 Pa to 100 Pa. However, conventional EB guns generally exhibit stricter pressure limits, which is true for the hybrid cathode, too (figure 7). The higher the process gas pressure within the chamber, the higher its partial pressure in the cathode compartment contributing to electron emission stimulation. Therefore, the HVGD working gas flow rate decreases additionally to the decrease at increasing beam current. As a result, operating the gun concurrently at high chamber pressures and high EB power is still a challenge, as the HVGD working gas flow rate tends to zero and limits the controllability of the beam. Despite the fact that this issue does not represent a restriction for non-reactive or low-pressure reactive PVD, promising measures to extend the allowed process pressure range including a revised design of thermal cathode insulation and electrical potential distribution within the cathode compartment are under investigation.

5. Conclusions

The HCA discharge has been shown to be an efficient plasma source particularly with magnetic field and at reduced gas flow rates. It is a flexible tool for substrate pretreatment, plasma-assisted PVD and CVD in order to achieve unique coating properties, which has been shown for plasma-activated EB-PVD of dense YSZ coatings.

The hybrid cathode EB gun benefits from its simple mechanical and electrical setup resulting in remarkably reduced cost as well as in compact design in comparison to conventional EB guns. Consequently, it has the potential to cover application fields, where the EB technology has not been suitable or economically reasonable so far.

6. References

[1] B. Zimmermann, F. Fietzke, W. Möller, *Surf. Coat. Technol.* **205** (2011) S393.

[2] H. Morgner, M. Neumann, S. Straach, M. Krug, *Surf. Coat. Technol.* **108-109** (1998) 513.

[3] F. Fietzke, B. Zimmermann, *Surf. Coat. Technol.* **205** (2010) 1491.

[4] B. Zimmermann, F. Fietzke, H. Klostermann, J. Lehmann, F. Munnik, W. Möller, *Surf. Coat. Technol.* **212** (2012) 67.

[5] P. Feinäugle, G. Mattausch, S. Schmidt, F.-H. Rögner, *54th SVC Ann. Tech. Conf. Proc*. (2011) 202.

[6] G. Mattausch, B. Scheffel, O. Zywitzki, Chr. Metzner, F.-H. Rögner, *55th SVC Ann. Tech. Conf. Proc*. (2012) 179.

[7] S. V. Denbnovetsky, V. G. Melnyk, I. V. Melnyk, *IEEE Transact. Plasma Sci.* **31** (2003) 987.