Recent Developments in Vacuum Arc Deposition

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Invited Paper

***Abstract*—Vacuum arc deposition (VAD) has become an established industrial art for producing hard, wear-resistant and decorative coatings. Sophisticated coatings, including tertiary compounds and multilayers, are increasingly used. Filtered VAD and hot electrode vacuum arcs are increasingly investigated to obtain high-quality, macroparticle-free films. Improved filtered sources, including large rectangular filters, have been demonstrated. Besides tool coatings, films for metallizing integrated circuits and protecting magnetic media are being developed.**

***Index Terms*—Cathodic arc, coatings, filtered vacuum arc, thin films, vacuum arc.**

I. Introduction

VACUUM ARC deposition (VAD), a process which uses the cathode or anode vapor plasma produced from a high current discharge in a vacuum environment to deposit coatings and thin films, was first investigated at the end of the 19th century and has been widely industrialized during the past quarter century [1]–[3]. All deposition processes contain the following elements:

1) generation of the depositing material in a mobile form—in VAD, as a metal vapor plasma, usually in the form of a hypersonic jet produced by cathode spots;
2) transport of the depositing material to the substrates—in VAD, magnetic fields are often utilized to collimate the depositing plasma;
3) condensation of the mobile material on the substrate surface to form the coating or film—in VAD, this process is often influenced by negatively biasing the substrate to control the impacting ion energy.

Since its modern industrialization, the use of VAD has grown rapidly, both due to the quality of the coatings produced and the favorable economics of the process. During the last decade, the characterization and understanding of the processes and the sophistication of its application have advanced significantly. The objective of this paper is to review these recent developments.

II. Theoretical Developments

Two theoretical issues which directly impact on VAD are reviewed in the following paragraphs. These are the interaction of vacuum arc generated plasma jets with background gas and the transport of these jets through "macroparticle filters."

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A. Interaction With Background Gas

VAD is rarely conducted under true vacuum conditions. In most industrial depositions, a reactive background gas is added in order to deposit ceramic compounds (e.g., N_2 in the case of TiN deposition), and in some cases an inert gas is added to stabilize the arc. Early work by Drouet and Meunier described the interaction of the plasma jet generated by the cathode spots by a snowplow model [4], and Boxman and Goldsmith established a simple criteria based on equating the momentum flux of the plasma jet with the background gas pressure [5].

The supersonic plasma jet emitted by the cathode spots is decelerated by collisions of its particles with the background gas particles. The critical issue is what happens as the jet velocity approaches the sonic velocity. Kelly *et al.* developed a fluid model to describe the interaction of plasma radially expanding from a discharge between small parallel electrodes into a surrounding gaseous ambient. Using a quasi-time-dependent integration technique, they found a solution in which the jet velocity smoothly passes the sonic point [6]. In contrast, Gidalevich *et al.* consider the mutual interaction of supersonic cathode spot plasma jets and the interaction with a stationary ambient gas. When the relative velocity of the two components is supersonic, Gidalevich proposes that a shock front will form, having a thickness on the order of a mean-free-path [7], [8].

B. Plasma Transport in Filters

The major disadvantage of VAD is the coproduction of liquid droplets or solid debris, known collectively as macroparticles (MPs), at the cathode spot. To alleviate this problem, MP filters have been devised in which the plasma is guided by a magnetic field through a curved duct, which blocks a direct line of sight between the cathode and the substrate, thus preventing most MP transmission. Recent practical advances in this field will be reviewed in Section IV below, while recent theoretical advances are described briefly here.

The major problem with most filtered systems is that the plasma output is low compared to unfiltered sources. Theoretical models have been devised to explain the plasma transport and to help design more efficient filters. Much of the theoretical work is based on the early flux tube [9], rigid rotor [10] and drift [11], [12] models. Usually, the magnetic field is sufficiently strong to guide the electrons, but not the ions. The ions are held radially against the centrifugal force by an electric field generated by their separation from the electrons.

Shi *et al.* [13], [14] used a modified drift model, which took into account radial electric fields generated by biasing the duct wall. Their result agreed well with past experiments and was used to predict the performance of a twist filter. However, their model predicts a radial drift velocity which approaches infinity when the magnetic field approaches zero.

Alterkop *et al.* [15]–[18] developed a two-fluid hydrodynamic model, which treats the electrons and ions self- consistently. The analysis depends on whether the plasma is in electrical contact with the walls, or separated from the walls. Separate approximations are also used for weak and strong magnetic fields and it was found that the plasma output increases slightly with the magnetic field in the weak field case and strongly with the field as the value for magnetic confinement of the ions is approached, as observed experimentally. The model predicts that the plasma beam is displaced from the axis both in the radial direction $\mathbf{G}$, due to centrifugal force, and in the $\mathbf{G} \times \mathbf{B}$ direction.

Beilis *et al.* [19] also used a two fluid hydrodynamic model, in which the inertial electron terms are neglected, in a curved duct with a rectangular cross section. This choice of geometry simplifies the mathematical formulation and may be applicable to recently developed rectangular filters. In addition, they analyzed the sheath, particularly when the duct wall is positively biased, using separate formulations depending on whether the electron Lamor radius is larger or smaller than the sheath thickness. They matched the plasma to the sheath by forcing the radial plasma current to flow through the sheath.

Keidar *et al.* [20] considered the free expansion of a plasma jet entering into a plasma duct, when the initial jet diameter is smaller than the duct diameter. The model predicts that the longitudinal velocity of the plasma is tripled as it expands and propagates. Furthermore, the internal electric field separates the various ionic species.

In comparing with experiments, the theoreticians were usually forced to make some assumptions about the plasma conditions in the experimental filters, or to fit their curves to the experimental results using some free parameter. Verification of theoretical models would be greatly enhanced if experiments were available in which not only the ion current or plasma flux was measured, but also the electron density and temperature within the filter.

III. Hard Coating Designs and Applications

Almost all current vacuum arc coating production uses the unfiltered cathodic arc, primarily for producing hard wear resistant and decorative coatings. During the early industrialization of VAD, gold-colored TiN was the coating of choice. Its high hardness and wear resistance, low diffusion coefficient and good chemical stability enhanced tool performance and lifetime. Furthermore, its bright gold color gave it a marketing advantage—its presence was blatantly obvious, as well as attractive. In addition, it is relatively easy to deposit successfully, due to a large process parameter window.

Today's industrial end-users of coated tools are more sophisticated and specify tools with coatings which perform best in their own application tests. More advanced coatings, including Ti(C,N), (Ti,Al)N, and various multilayer and nano-layer coatings, are occupying increasingly large portions of the cutting tool coating marketplace. Besides being applied to steel tools as it was in the early days, carbide tools are increasingly being coated by VAD. Hydrogen-free diamond-like-carbon (DLC), applied using a vacuum arc on a graphite cathode, is currently being used to coat razor blades and tools intended for nonferrous and not-metallic machining applications.

Non-tool applications are becoming increasing important. These include coatings on medical implants, as well as decorative coatings. The gold- and brass-like colors of TiN and ZrN are particularly important in the latter and are used to coat jewelry, cutlery, spectacle frames, plumbing fixtures, and household hardware (e.g., door knobs), among other uses. In some cases, the color is adjusted by varying the metal ratio in (Ti,Zr)N in order to best imitate a particular gold or brass alloy. Efforts in R&D and marketing are being made to extend the application of VAD to the enormous wear part market. Currently, CrN and CrON coatings are applied to automotive piston rings and TiN and DLC to diesel fuel injectors.

IV. Filtered VAD

The major disadvantage of VAD, in comparison to other physical vapor deposition techniques, is the presence of macroparticles, which degrade the coating quality and preclude the use of VAD in sensitive and lucrative applications in optics and electronics. Filtered VAD (FVAD) uses a magnetic field to bend the plasma jet around an obstacle which blocks macroparticles from reaching the substrate. The paragraphs below review the considerable progress in the design and characterization of filtered vacuum arc plasma sources and the development of coatings that have occurred in recent years.

A. Filtered Source Design and Operation

Figures of Merit for Filter Performance: The need for a macroparticle filter was recognized and addressed by Aksenov and his colleagues in the pioneering Kharkov group, who investigated a number of filter configurations, including the popular toroidal duct design. Generally, the plasma output from filtered sources is less than unfiltered sources operating at the same current. A figure of merit for comparing the plasma output of various sources and for optimizing the performance of a given source is the ratio of the ion current output to the arc current $I_\mathrm{i}/I_\mathrm{arc}$ [21], [22]. The use of this figure of merit takes into account both the plasma transmission of the filter and the coupling of the arc plasma into the filter and, hence, has been called the *system efficiency* [22]. A second useful figure of merit is time averaged ion current output, $\langle I_\mathrm{i} \rangle$, which in a production system determines what coating volume can be produced per unit time, or for a given coating thickness, how many workpieces or what surface area can be coated.

Less well-addressed has been the question of macroparticle filtering effectiveness. It has been acknowledged by several investigators that some macroparticles succeed in reaching the substrate, despite the filter. The MPs are transmitted by some combination of entrainment in the plasma stream [23], electrostatic reflection from the duct walls due to MP charging [19], or mechanical bouncing or splattering upon impact with solid surfaces. Various filter geometries and interior corrugations and baffling have been used to reduce MP transmission, however, no figure of merit for filtering effectiveness has been adopted. The problem is complicated by the diversity of MP sizes, which range up from the limit of detection of the imaging diagnostic to typically tens of μms. In the unfiltered arc, the distribution typically decreases exponentially with MP size. Thus, a long deposition time and, hence, a relatively thick coating, is required to detect the rare large particles, while a thin coating is required to avoid burying the smaller particles. The following figure of merit is proposed: the particle size distribution per unit volume of coating–i.e., the number of particles per MP size interval per unit area of substrate surface, divided by the coating thickness. Determining the above figure of merit will generally require microscopically inspecting a series of coatings with different thickness, using decreasing magnification or resolution (and an increasing observation area) with increasing thickness and counting only particles which are larger than the coating thickness. Depending on the particular requirements of a given application, a limited portion or an integral of the distribution function might be most relevant, e.g., counting only MPs above a certain size, or the fraction of the coating surface area or volume occupied by MPs.

Characterization of Quarter-Torus Systems: The quarter-torus filter has been the most studied configuration. The plasma flux distribution in the quarter-torus has been approximated as a Gaussian, which is generally displaced from the duct axis [24]. Bilek showed that as a first approximation, the plasma flow follows the magnetic field lines. In practical devices, which use finite coils or magnets, fringing fields and asymmetries can significantly deflect the plasma beam from the system axis. Thus, as a first rule for good filtered source performance, magnetic field lines emerging from the active cathode region should arrive at the substrate surface without intersecting the duct walls or other obstacles [25]. In addition, systematic displacements toward the outside of the curve (i.e., in the direction of the 'centrifugal force' $\mathbf{G}$) and out of the plane of symmetry in the $\mathbf{G}\times\mathbf{B}$ direction are also significant [24]. Most of the ion current lost to the wall is in the $\mathbf{G}$ direction [26].

In the quarter-torus filter, the filter transmission generally increases with the strength of the magnetic field. Depending on the coupling of the torus field to the arc electrodes, $I_{\rm i}/I_{\rm arc}$ may likewise increase, but may reach a maximum if the stray field forces the cathode spots away from the center region of the cathode [27]. $I_{\rm i}/I_{\rm arc}$ can also be improved by applying a positive bias to the duct wall, as observed initially by Aksenov [28]. Where it is inconvenient to bias the duct, considerable improvement can be achieved by applying a positive bias to an electrode placed along the interior wall of outermost portion of the duct [29]. The degree of improvement generally declines with increasing field, however. The best $I_{\rm i}/I_{\rm arc}$ reported to date has been 6.3% in a pulsed system [30]. To obtain this $I_{\rm i}/I_{\rm arc}$, the magnetic field was tailored to keep the plasma beam centered in the duct.

The ion energy distribution function (IED) and the charge state distribution of the metallic plasma are affected by its passage through a filter. While Bilek *et al.* [31] state that the IED does not vary significantly as a function of the magnetic field strength B, Shi *et al.* found that the average ion energy increased from 24 to 34 eV when B was raised from 10 to 45 mT [32]. In an experiment where $I_{\rm arc}$ and B varied simultaneously, the mean charge state at first decreased and then increased, with increasing B [33]. The introduction of gas into a filtered arc system generally lowers the directed energy of the cathodic ions and increases their temperature, while low-energy gaseous ions are also produced [34].

New FVAD Configurations: Several FVAD design approaches have been investigated recently, with respect to pulsed versus continuous operation, open versus closed ducts and duct geometries. Pulsed sources have several advantages, including no need for electrode cooling if operated at low duty cycle, and simplicity of construction and the easy retention of the cathode spots on the front cathode surface, thus maximizing coupling of the plasma into the filter duct. In addition, some work indicates that high current pulsed operation produces less MPs, possibly because of vaporization during their passage through the plasma. However, in an industrial environment where production rate is a key question, the cooling requirements for a given production rate are equal or greater than that for a continuous system and the power supplies are generally costlier for a given average arc current.

Open plasma ducts are formed by a free-standing toroidal field coil placed in a large vacuum chamber. The plasma is guided along the magnetic field lines as described earlier, while MPs, for the most part, pass through the mostly empty space between field windings, and thus have only a small probability of bouncing toward the duct exit, reducing MP transmission. Open ducts are most suitable for pulsed systems, where often the pulsed arc current is used to excite the field coil. Continuous open systems are complicated by the need to cool the coil within the vacuum.

In recent years, some new filter geometries have been developed, in addition to the classical quarter-torus [Fig. 1(a)] and others which were reviewed previously [21].

1) The *S-filter* [Fig. 1(b)] is a series connection of two bent ducts, such that the plasma is first bent in one direction and then in the other [35]. This configuration has the advantage that whatever systematic $\mathbf{G}$ or $\mathbf{G}\times\mathbf{B}$ beam displacement is induced in the first bend is approximately cancelled in the second bend.
2) A configuration featuring two bends, as in the S-filter, but in different planes is known as the *twist filter* [36] [Fig.1(c)] when used as an open duct or the *off-plane double bend filter* [37] when deployed as a closed duct.
3) The *Venetian blind filter* [Fig.1(d)] consists of an array of slanted slats, which block a direct path between the cathode and substrate. High current is injected along the slats, producing a magnetic field which guides the plasma between the slats.
4) Axial plasma flow is converted to radial flow [Fig.1(e)] using one of several variants in which a cathode is mounted along the axis of a pair of coils which are energized in the opposite sense, producing a cusp field. Plasma flows from the cathode along the field lines and emerges from the gap between the coils in the radial direction. Plasma, which would have continued past the intercoil gap in the axial direction, can either be reflected back using a biased electrode, or a second cathode can be

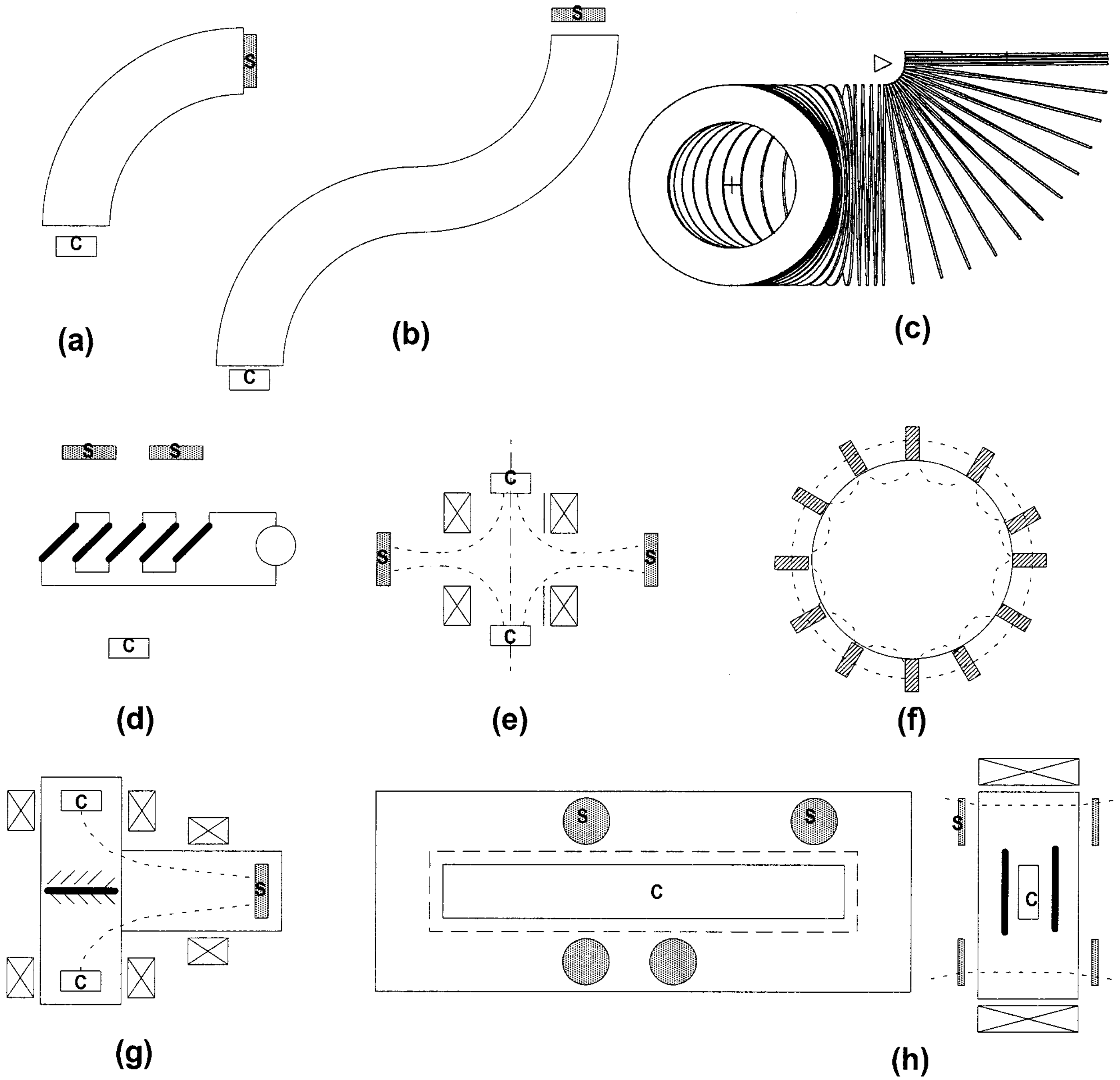

Fig. 1. Filter Configurations: (a) Classic 90° torus, (b) S-filter, (c) Out of plane twist, (d) Venetian blind, (e) Radial flow with cusp field, (f) Magnetic bottle, (g) Double rectangular filtered source, and (h) Dual-flow rectangular source (two views). Dotted lines are magnetic field lines, C-cathode, S-substrate.

symmetrically placed opposite the first cathode, so that the two plasma beams collide forming a stagnation point in the center of the axis [38].

5) While not an MP filter, the *magnetic bottle* [Fig.1(f)] can be used to improve deposition uniformity from FVAD sources. It consists of a cylindrical vacuum vessel with a series of strong magnetic gap fields located around its periphery. The region near the axis has only a weak fringing field. Plasma is injected at one end and expands radially, filling the weak-field region with an approximately uniform plasma while plasma losses at the wall are prevented.
6) Rectangular cathodes have been coupled to filters, to produce a plasma beam with a large cross section area. In the variant shown in Fig.1(g), two sources are mounted on a vacuum chamber and can provide up to 6 A of metal ion current while operating at a total cathode current of 240 A [39]. This source is currently used in industrial production.
7) Another rectangular variant uses the peripheral surface of a rectangular bar as a cathode and produces two rectangular plasma beams [Fig.1(h)]. This source can be used to coat workpieces passing both sides of the source in an inline deposition system. Up to 5 A of ion current was extracted, with an arc current of 300 A [40].

B. Optical and Electronic Coatings

Thin films for optical, magnetic, and electronic devices have the most stringent requirements for uniformity and in particular macroparticles can devastate device performance. FVAD coatings are currently being studied or developed for a variety of applications in this area.

Both optical and magnetic devices frequently require hard protective coatings. FVAD DLC, in particular, is being investigated as a protective film for hard disks and disk reading heads, as well as optical devices. In addition, FVAD is being investigated as a means of applying magnetic thin films on disks.

Transparent conducting oxide films are used as electrodes in solar cells and flat panel displays. Amorphous SnO_2 films have been prepared with FVAD at rates of up to 10 nm/s. When transverse current injection or rapid thermal annealing is applied together with FVAD, the best reported conductivities have been obtained [41], [42].

Amorphous Si films, needed for solar cells and thin film transistors (especially in active matrix liquid crystal displays) have also been prepared by FVAD. To date, however, the FVAD films have had inferior characteristics compared with those prepared by glow discharge chemical vapor deposition [43]–[45].

In ultra-large scale integrated circuits, preparation of the "back-end" metallization layer which provides interconnects between the various circuits is costlier than preparation of the active devices. With increasing miniaturization and speed requirements, the interconnects are in the form of lateral trenches and vertical vias having large depth to width ratios and are difficult to prepare with conventional vacuum deposition techniques. FVAD has been proven to have the capability of providing conformal barrier layers and filling trenches [46], [47]. An industrial scale deposition system for Cu metallization is currently being developed [48].

V. Arc Coating Technique Variations

Most VAD is conducted with an arc running in a cathode spot mode, with the cathode spot plasma jets impinging (directly or after filtration) on substrates located relatively far from the cathode, as described above. A few interesting VAD alternatives have been studied, however. Pulsed vacuum arc deposition, with the substrate located very close to the cathode and typically operated as the anode, can be used to obtain extremely high instantaneous deposition rates and interesting metallurgical effects by utilizing the heat flux to heat the substrate surface [49]. Recently, the properties of TiN coatings applied using this technique to steel substrates was studied [50]. It was found that with 500 A peak current pulses on a 15-mm diameter cathode, the coatings became less porous and had a preferred (111) orientation when the background N_2 pressure was increased from 200 to 1000 Pa.

A hot electrode vacuum arc mode can be generated by thermally isolating one of the electrodes and vaporizing it. While a few materials (e.g., C, Cr) sublimate from the solid state, in most cases, a refractory crucible electrode must be constructed, which holds a more volatile liquid material which is evaporated. Under certain conditions, the arc attaches diffusely to the electrodes, avoiding macroparticle formation. However, this method shares the disadvantages of thermal evaporation:

1) The liquid melt can interact with the hot crucible, contaminating the former and damaging the latter.
2) Most alloys cannot be evaporated from a single source.
3) The crucible must be oriented with the free liquid surface on top.

Recently, a new hot electrode variant, the Hot Refractory Anode Vacuum Arc, has been studied in which material emitted from cathode spots is deflected or re-evaporated from a hot refractory anode [51]. A MP-free deposition zone is obtained with better through-put than typical FVAD apparatus, while apparently avoiding the difficulties inherent with other hot-electrode vacuum arcs.

VI. Conclusion

The understanding of the interaction of vacuum arc plasma jets with background gases and magnetic fields has advanced significantly. The traditional use of VAD for producing hard coatings has expanded to new applications and become more sophisticated in applying new coating materials. Considerable progress has been achieved in improving macroparticle filtering techniques and electronic and optical applications are being developed. Further development is needed, particularly in correlating experiments and theories.

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