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# Processes in a plasma arc installation for vacuum coating depositions Part 1. Plasma generation

Vladimir I. Gorokhovskiy

*Mirus International Inc., 801 Sheppard Avenue West, North York, Ont. M3H 2T3 (Canada)*

Vladimir P. Polistchok and Ivan M. Yartsev

*Institute for High Temperatures, Russian Academy of Sciences 127412, Izhorskaya 13/19, Moscow (Russian Federation)*

## Abstract

This paper presents the results of research on energy and mass transfer at the surface of evaporated electrodes in low pressure arcs. It is shown that, when the electrode temperature is sufficiently high, the arcs may have diffusive electrode spots. The parameters of this discharge are determined by the cathode material and the temperature conditions on the electrode. The characteristics of the cathode material are described by the atom–electron ratio  $\zeta$ , which is determined by the ratio of flows of thermally evaporated atoms to electrons from the cathode. The conditions of existence and the primary characteristics of vacuum arcs with a cathode diffusive spot are analysed. The characteristics of discharge on an evaporating hot anode and a.c. arc are investigated. The parameters for process optimization and the coating deposition equipment are discussed.

## 1. Introduction

At present various types of vacuum plasma arc installations (PAIs) are widely used for coating deposition and surface treatment. The plasma-generated medium in a PAI is produced by ionization of the thermally evaporated atoms from electrodes in a low pressure or “vacuum” arc (VA). These discharges can generate highly ionized supersonic plasma jets from the atoms of electrode material [1, 2]. These jets produce coatings from the metallic alloys and compounds. If complex chemical coatings are required (nitrides, carbides, oxides) then the reactive gases ( $N_2$ ,  $O_2$ ,  $CO_2$  and others) must be placed in the VA region. PAIs have several advantages as compared with other vacuum plasma installations. Principally, the working medium has a high degree of ionization and chemical activity; furthermore, it is possible to use external electromagnetic fields to control the parameters of the working medium. High values of ion energy ( $10^2$ – $10^3$  eV) and ion concentration produce effective ion treatment of the substrate surface. They also form transition layers and synthesize coatings with unique properties [3].

There are two distinct types of VA. If the mean temperature of the consumption electrode (cathode)  $T_c$  is not sufficiently high (less than  $10^3$  K), then non-stationary random moving cathode spots with a current density  $j_c > 10^4$  A cm<sup>-2</sup> [1, 2] will occasionally occur. These spots are a source of cathode material vapour. By

using VA on the cold cathodes (cold-cathode arc (CCA)), effective methods have been realized that deposit high quality coatings with various compositions and purposes [4]. Plasma jets in CCA usually contain macroparticles of the electrode material with a maximum size of 1–10  $\mu$ m. Special electromagnetic macroparticle filters (EMFs) have recently been developed to clear the plasma flow from these particles and to improve the technological characteristics of PAIs [5]. Alternative techniques using hot evaporated electrodes (HEEs) have been developed for PAIs. In these installations the mean electrode temperature is high enough (greater than  $1.52 \times 10^3$  K) to allow the use of a new type of VA with a stationary diffusive spot (SDS) [6–11]. In this case the current density of the electrode is low ( $10$ – $10^2$  A cm<sup>-2</sup>) and evaporation takes place on a specific area of the working surface on the electrode. Plasma flows in VA on HEEs do not contain macroparticles and the plasma parameters of these flows may be changed within wide limits. Compared with PAIs, with CCA the problems of arc stabilization on the electrode surface and discharge ignition are significantly simplified.

Depending on the polarity of connection of the power supply two types of arc on HEEs may be distinguished: the arc with hot evaporated anode (HEA) or hot evaporated cathode (HEC). For these cases electrode heating may be produced by the discharge itself or by means of an auxiliary power source. In general obtaining VA on an HEA is more straightforward than VA on an HEC.

As a rule the degree of working medium ionization in the latter case is higher. Besides the HEA-arc operating demands, the application of a special second electrode (cathode) is necessary. It must have a comparatively high temperature to provide the required arc current. The installations with VA on an HEA allow the realization of plasma vapour deposition processes giving high quality single-layer and multilayer coatings based on nitrides and oxides. For example, VA on an HEA from metals and semiconductors heated by electron beam is used in the Balzers installation [12].

The purpose of this work was the study of processes occurring in the technological PAI. Special attention was given to VA on HEEs. In the first part of this work the phenomena connected with plasma generation in the arc discharge are discussed. In the second part (*Surf. Coat. Technol.*, 61 (1993) 108) the working medium processes of the PAI are presented

## 2. Experimental details

In general, the electrode material in VA on HEEs is melted and placed in a special crucible. To investigate VA on HEEs two types of installation have been used. In the first type of installation (PAI-1) used, the heat-insulated crucible is heated by an electron beam heater (EBH). The EBH power was less than 1.5 kW. The temperature of the HEE significantly determines the characteristics of VA. This temperature was measured by optical pyrometer with an error of 2%. Physical investigations of d.c. VA with cathode and anode HEE polarities and a.c. VA were completed. For the HEE anode the second electrode was thermionic tungsten cathode. Rare earth metals with high electron emission (Gd and La) and Cu, Mg, Cr and Ti were used as the material for the HEEs. The experimental methods and main results are described in refs. 7-9 and 13.

The second type of installation with HEEs (PAI-2) is shown in Fig. 1(a). This figure shows a pilot installation used to produce the superhard coating Thermion-500/SHC. The evaporated material was placed in a water-cooled copper crucible and heated by a gas discharge electron gun. Focusing and scanning of the electron beam on the cathode surface was provided by computer-controlled magnetic lenses. VA with an HEC was burned in an opposing magnetic field which was created by a pair of magnetic coils placed in a coaxial electron beam. The water-cooled enclosure of these coils serves as an anode and, simultaneously, acts as an EMF with an opposing magnetic field for arc plasma flow from the crucible cathode (when the coil enclosure is placed between the operating surface of the crucible cathode and substrates). The substrate was placed on a carousel-shaped substrate holder. An autopolarized bias

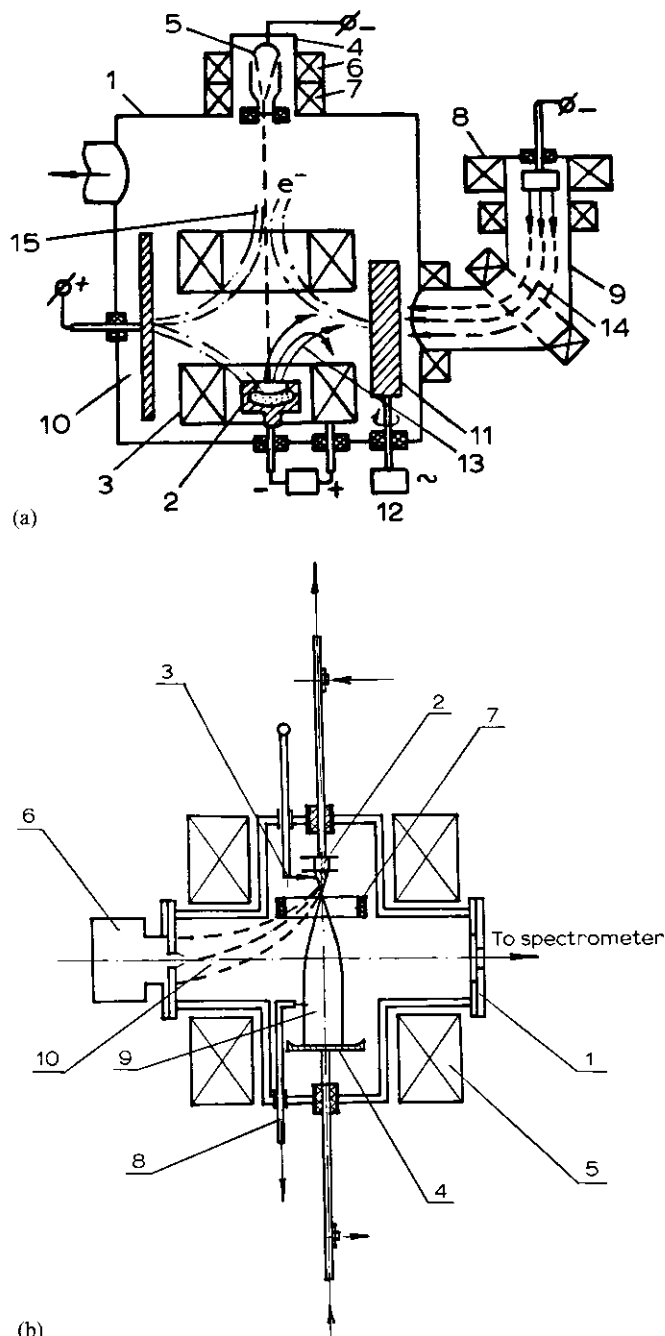


Fig. 1. Experimental PAI. (a) The pilot installation for Thermion-500 SHC: 1, vacuum chamber; 2, water-cooling crucible with operating substance; 3, deviating magnet coil; 4, electron beam gun; 5, gun cathode; 6, focusing lens; 7, scanning lens; 8, plasma source with cold cathode; 9, angular macroparticles filter; 10, auxiliary anode ionize; 11, substrate holder; 12, high frequency generator; 13, column of VA with hot cathode; 14, column of VA with cold cathode; 15, force lines of opposing magnetic field. (b) Installation for investigation of VA with a cold cathode in a longitudinal and opposing magnetic field: 1, vacuum chamber; 2, cathode; 3, ignition electrode; 4, anode; 5, magnetic coils; 6, mass spectrometer; 7, inductive sensor; 8, Langmuir probe; 9, VA column in a longitudinal magnetic field; 10, position of the VA column in an opposing magnetic field.

potential with a frequency of 1.76 MHz was applied to the substrate holder. In addition, a cathodic plasma arc source with angular-shaped EMF was used. A supplementary anode was installed near the substrate holder in order to create an auxiliary gas arc. In PAI-2 VA on an HEC from titanium and carbon were studied.

Investigations of VA on a cold cathode were accomplished in PAI-3, where a copper rod cathode of small diameter (for some experiments) and copper disc anode were used (Fig. 1(b)). Precise cathode construction localized the cathode spot. The magnetic system of PAI-3 produced both a longitudinal and an opposing magnetic field. Condensation probes were used to measure the density of mass flow; the plasma conductivity was determined by an induction coil. Plasma composition was studied using mass and optical spectrometers positioned on the side walls of the vacuum chamber between magnetic coils. The procedures and main results of measurements have been described in refs. 14 and 15. For all installations, the plasma parameters (electron temperature  $T_e$ , ion saturation current  $I_{is}$  and plasma potential) were determined using Langmuir probes. The vacuum chambers were equipped with a system to control the leakage of reactive gas mixtures. As was shown elsewhere [7, 8, 13] the atom–electron ratio  $\xi$  determines the characteristics of VA with an HEC. The value  $\xi$  is the ratio of flows of thermal evaporated atoms to electrons.

Figure 2 shows the temperature dependences of  $\xi$  for various cathodes [8]. Two classes of cathode material may be discerned: with  $\xi \gg 1$  and  $\xi \ll 1$ . A small value of  $\xi$  is typical for refractory and rare earth metals.

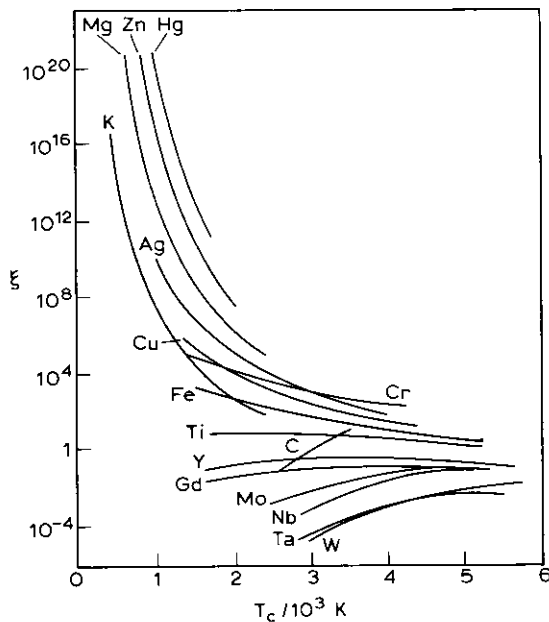


Fig. 2. Atom–electron ratio  $\xi$  for cathode metals depending on their temperature.

### 3. Results and discussion

#### 3.1. Arc on an HEC

The following parameters of VA with an HEC were measured: voltage–current characteristics, cathode temperature, evaporation rate with arc  $G_a$  and without arc  $G_0$ , temperature  $T_e$  and saturation ion current density  $I_{is}$ . The heat flow to the cathode  $Q_c$ , determined by the heat calibration of the cathode, was measured [7]. The parameter  $\xi$  changed from  $10^{-2}$ – $10^{-1}$  (for cathodes from La and Gd) to  $10^4$ – $10^5$  (for Cu and Cr). Results obtained [7–9, 13] revealed a set of special features of VA on an HEC. It has been established that the main mechanism of the charge transfer on the cathodes with  $\xi < 1$  is thermionic emission. It has been shown that arcing causes the return of a portion of the evaporated atoms back to the cathode. This part may reach 50%–70% of all evaporated atoms. A hydrodynamic model of a plasma jet in VA, including non-equilibrium processes, has been developed [9]. It has been shown that, in comparison with CCA, the investigated discharge gives some new possibilities for controlling plasma parameters. It has been found that external heating may be used to influence the plasma of VA with an HEC.

This effect may be seen from the data given in Fig. 3. A high rate of deposition (proportional to  $G_a$ ) may be achieved when the heating power  $N$  is large. However, a higher energy contribution in the arc and, consequently, desirable plasma parameters can be obtained when  $N$  is small. When  $N$  decreases to zero, the arc changes to a self-sustaining mode. Figure 4 shows several characteristics of this discharge. The discharge studied is distinct from CCA and has dropping voltage–current characteristics. In this mode, the voltage equivalent of the heat flow to the cathode  $V_c = Q_c/I_a$  is proportional to the arc voltage  $V_a$  and  $V_c = 0.25 V_a$ . Therefore the losses of the cathode heating and its evaporation are approximately 25% of the discharge power. The remaining power is spent on plasma generation and acceleration. In PAI-2 a non-self-sustained arc with an HEC with carbon and titanium was studied. In this apparatus, the diameter of the cathode spot (5 mm) was determined from the diameter of the electron beam. Similarly to results obtained in PAI-1, it was found that, when the cathode temperature was higher than some critical value, VA with an SDS may be obtained. The cathode temperature for carbon of 2900–3200 K was estimated from its evaporation rate. This temperature was sufficient to provide the necessary values of thermionic current. During arcing a cathode crater was formed which impaired arc operation. To remove this effect and to provide an even rate of cathode erosion, the heating electron beam was scanned continuously or stepwise. Figure 5 shows voltage–current characteristics for cathodes from Ti and C, which are similar. The values of

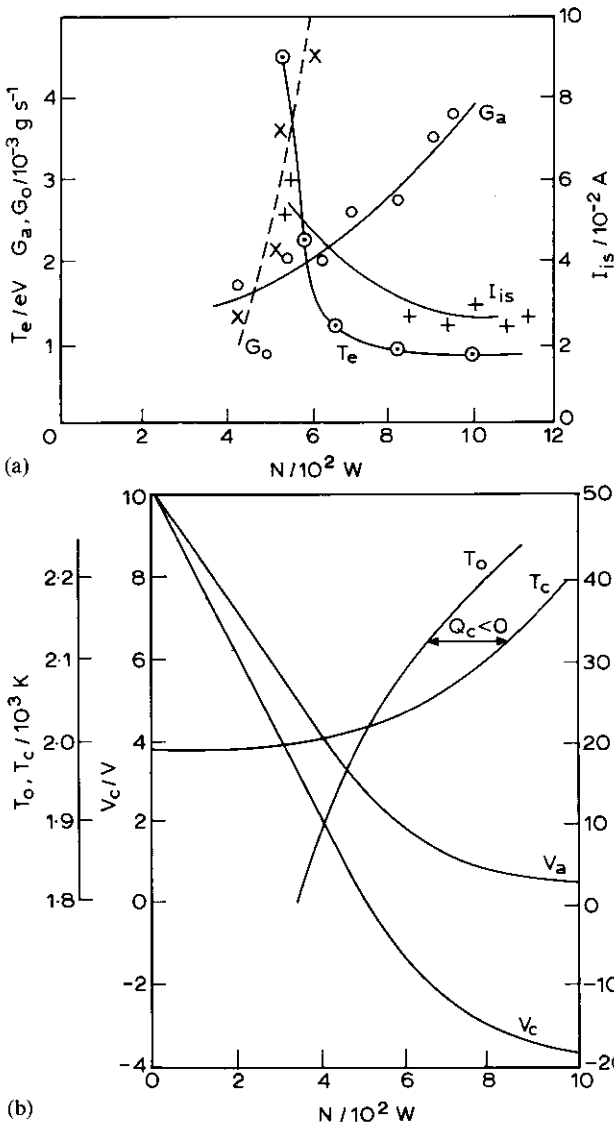


Fig. 3. The influence of heating power on the averaged arc characteristics for VA with a Gd HEC (arc current  $I_a=50$  A): (a) values of electron temperature  $T_e$ , ion saturation current  $I_{is}$ , rate of cathode evaporation with arc  $G_a$  and without arc  $G_0$ ; (b) cathode temperature with arc  $T_c$  and without arc  $T_0$ , heat flow to the cathode  $Q_c$ , arc voltage  $V_a$  and voltage equivalent  $V_e$ .

the ratio  $\xi$  for these materials at the operating temperatures used are also similar. The arc voltage itself is distinguished from the voltage in CCA, which has a slight dependence on arc current and is approximately equal to 20 V.

The results obtained allow the conclusion that for materials with  $\xi < 1$  (Gd, La, C and others) VA with an SDS may exist when the cathode temperature is sufficient to satisfy the following conditions: the thermionic current from the cathode must be approximately equal to the arc current and the vapour concentration near the cathode is greater than  $10^{19} - 10^{20} \text{ m}^{-3}$ . It may be noted that, when the ratio  $\xi$  increases, the minimum

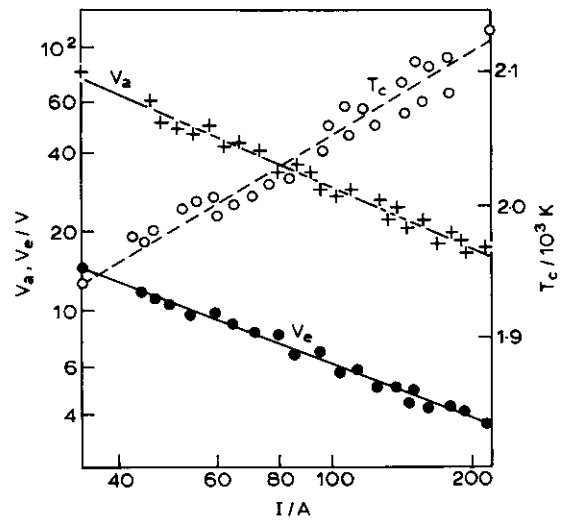


Fig. 4. Parameters of a self-sustaining arc on the hot cathode from Gd as functions of arc current: +, arc voltage  $V_a$ ; ●, voltage equivalent  $V_e$ ; ○, cathode temperature  $T_c$ .

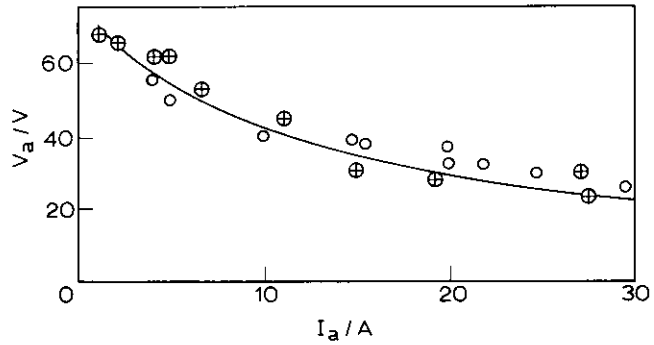


Fig. 5. Voltage-current characteristics of an arc on an HEC from carbon (○) and titanium (⊕) for an electron beam current of 0.3 A (accelerating voltage, 15 kV).

vapour pressure at which this discharge may be observed also increases from 0.11 Pa (Gd and La) to  $10^2$  Pa (Cr).

### 3.2. Arc on an HEA

In discharges with an HEA, zones of vapour and charged particle (electrons) generation are located near the anode and thermionic cathode respectively. Modification of the conditions in these zones affects the arc characteristics. Changing the EBH power applied to the anode causes a change of evaporation rate, which in turn controls the plasma parameters. The influence of EBH power on arc operation is similar to the effect shown in Fig. 3. The modification of the cathode conditions also affects arc properties. Increasing the thermionic emission current of the cathode reduces the discharge power contribution. As the power  $N$  tends to zero the arc becomes self-sustaining. Figure 6 shows some characteristics of this mode. It is interesting that changing of the cathode filament current  $I_f$  allows us to

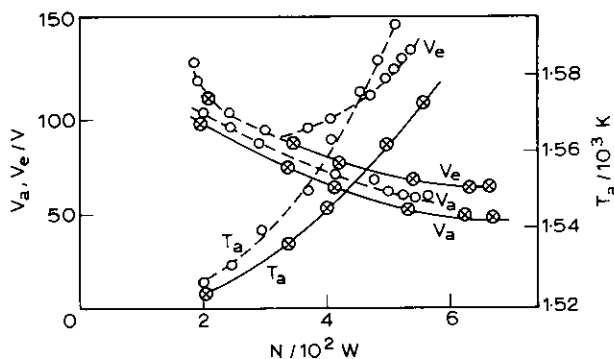


Fig. 6. Parameters of a self-sustaining arc on the hot Cu anode for  $I_t = 20$  A (○) and 23 A (⊗); arc voltage  $V_a$ , voltage equivalent of heat flow to the anode  $V_e = Q_a/I_a$  and anode temperature  $T_a$ .

modify the heat flow to the anode  $Q_a = I_a V_e$  and its temperature. The experiments in PAI-1 showed that the self-sustaining form of discharge on both electrodes may be obtained in the limit values of arc current:  $I_1 < I_a < I_2$ . When  $I_a$  tends to  $I_1$ , the heat flow  $Q_a$  cannot provide the necessary vapour generation. The current  $I_2$  is defined by the thermionic current of the cathode. The conditions shown in Fig. 6 are  $I_t = 20$  A,  $I_1 = 2$  A and  $I_2 = 6$  A.

3.3. A.c. vacuum arc on an HEE

A.c. vacuum arcs allow a plasma flow with periodically changing characteristics. These arcs generate a plasma environment with a periodic change of parameters. In this work an a.c. VA with a frequency of 50 Hz was realized. The conditions for the existence of VA with an SDS for both electrodes were established. Oscillograms of arc current and voltage and the influence of external heating on these parameters were studied. It was found that arcing modes without appreciable effects of electrical breakdown and zero-current intervals may be obtained. For these modes the current oscillograms differ slightly from the sinusoidal. Figure 7 shows the dependence of the maximum values of voltage in the a.c. VA with a Cr

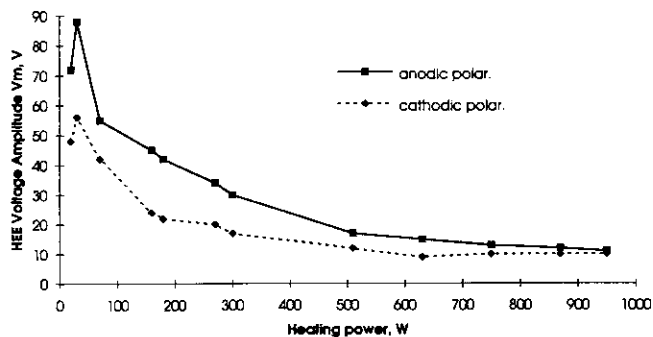


Fig. 7. Maximum voltage amplitude of a.c. VA for cathode (◆) and anode (■) polarities of HEEs as a function of heating power.

HEE on heating power. These results show a critical value of the heating power for a.c. arc initiation. When the heating power increases, the voltage for both polarities of HEE converge.

3.4. Arc on a cold cathode

In PAI-3 the plasma flow generated by cathode spots of CCA were studied. Figure 8 shows the angular distribution of mass flows from the cathode, its deviation from the usual cosine law and variations of this distribution occurring at a large distance from the cathode. The variation of the angular distribution for a large distance from the cathode corresponds to curved plasma arc flow lines in an internal and external magnetic field. Because of this effect, the calculation of angular distribution at a great distance from the cathode is incorrect. When using thin cathodes, a current increase causes appreciable cathode heating. It has been established that cathode heating results in a decrease of the portion of multi-charged ions in the plasma jet. In this case the angular distribution of mass flow tends to the cosine law. When the arc current was high enough, the cathode overheated and melted and a discharge with a mean current density on the cathode of 200–300 A cm<sup>-2</sup> occurred. These values of current density are similar to data for arcs on an HEC.

Various materials used in CCA influence the characteristics of VA. It has been found that the atom–electron ratio  $\xi$  may be used as a parameter which defines the properties of VA on the different cathodes. Figure 9 shows the dependence of specific erosion rates in CCA on  $\xi$ . The parameter  $\xi$  was calculated at a temperature close to boiling for each cathode material. In accordance with ref. 15 this parameter also determines other important characteristics of CCA, namely arc voltage, voltage equivalent of heat flow to the cathode, ion energies and

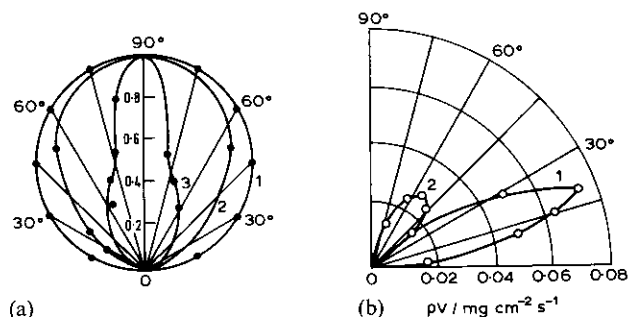


Fig. 8. Angular distribution of mass flow in an arc with a Cu cathode ( $I = 56$  A) at different distances  $L$  to the cathode: (a) distribution of total mass flow for a cosine law (curve 1),  $L = 1.5$  cm (curve 2) and  $L = 9$  cm (curve 3); (b) distribution of the neutral particles mass flow for  $L = 1.5$  cm (curve 1) and  $L = 9$  cm (curve 2).

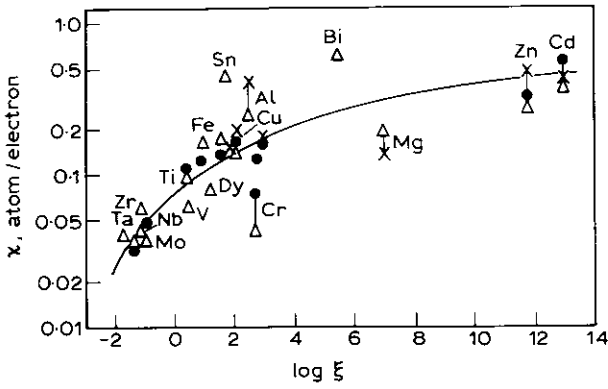


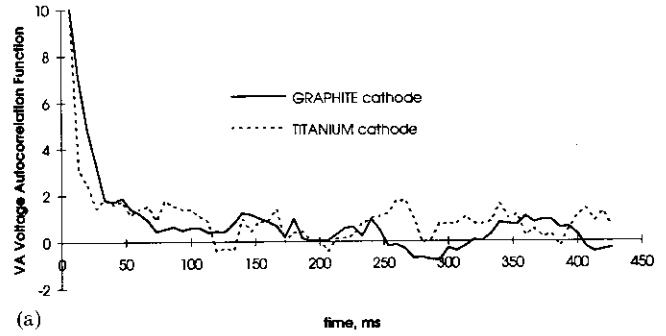
Fig. 9. Specific cathode erosion  $\kappa$  in the VA with a cold cathode as a function of the atom–electron ratio  $\xi$ .

mean ion charge in the plasma jet and the value of mean and minimum current on a cathode spot.

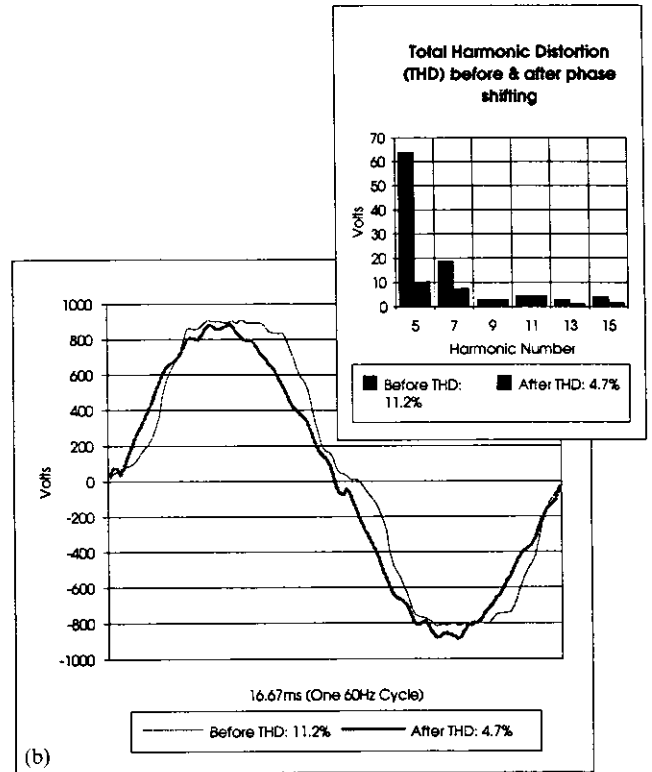
3.5. VA power supply effects

The ideal power supply for VA with a cold cathode is a constant current supply within a limited range of voltages. The variation in the low frequency characteristic periods of the VA voltage is determined by the creation process of cathodic spots. The characteristic period varies depending on the type of cathode material used. Figure 10(a) demonstrates that VA is a source of low frequency white noise. The voltage of VA with carbon cold cathode has a low characteristic frequency  $\nu_{\min} < 10$  Hz that is determined by a low velocity of cathodic spot movement on the carbon cathode. For  $I_{\text{arc}} = 100$  A the average values of the arc voltage  $\langle V_{\text{arc}} \rangle$  are equal to 29.3 V for carbon and 25.5 V for titanium, and the differential resistance  $(dV/dI)_{i_a, v_a}$  is equal to 30  $\Omega$  for carbon and 10  $\Omega$  for titanium near the working point. A larger value of differential resistance for carbon VA may be explained by the larger specific resistance of carbon.

The vacuum arc is a highly non-linear load that presents challenges to the design of practical power supplies. To realize a practical constant-current supply in d.c. VA installations, three-phase three-wire controlled bridge rectifiers are often utilized. D.c. VA sources with rectified power supplies inherently create power system harmonics which are harmful to the operation of rectifier control systems and other equipment in the system. These harmonics are integer multiples of the fundamental frequency (in North America 60 Hz). One very effective method to treat this type of harmonic distortion is phase shifting between various harmonic sources (Fig. 10(b)). The phase-shifting approach is a practical solution to power system harmonic problems when a PAI has more than one VA source [16].



(a)



(b)

Fig. 10. Electrical parameters of the VA power circuit: (a) voltage autocorrelation functions for VA with graphite (—) and titanium (---) cold cathodes; (b) mitigating harmonic distortion for the power supply of PAIs.

4. Conclusion

The investigations of VA were completed for various methods of plasma generation. A dimensionless parameter to analyse and predict VA properties on the various cathodes has been suggested. This parameter provides a common method of explanation for the processes of plasma arc generation for both the HEC and cold cathode. It was found that VAs on an HEE have several properties not observed in the VA with a cold cathode. This discharge has some practical advantages over CCA, namely the plasma jets are free of macroparticles and plasma parameters are readily controlled by changing

either the heating power supplied to the evaporated electrode or the arc current. VA of industrial frequency a.c. has been studied. A.c. VA sources may provide new advantages for technological applications.

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