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ACTIVE THERMOELECTRIC PROBING

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A novel type of electrostatic probe was used for studying the plane plasma discharge of relatively low pressure. This electrostatic probe, based on the usual K-type thermocouple, also known as "Active Langmuir Probe", generates a thermoelectric voltage. This thermoelectric voltage is used for plasma investigation in studies of metal contact properties. A qualitative and quantitative analyses of the Active Langmuir Probe are presented in this work. Experimental studies have shown that the measured thermoelectric voltage corresponds with the temperatures, exceeding the melting point of the measuring probe itself.

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1. Introduction

Electrostatic probes, also known as Langmuir probes, are widely used to investigate various forms of glow discharges. Such a low-pressure glow discharge is actually characterized as weakly ionized plasma. General basic plasma parameters may be estimated from measured Volt-Ampere (I-V) characteristics, using electrostatic Langmuir probes, however, measurement interpretation is most difficult due to the absence of comprehensive theory of the probe construction. Existing theories of electrostatic probes are mostly developed for symmetrical and one-dimensional geometries, such as spheres in infinite planes, or infinitely long cylinders [1,2]. Therefore, evaluations, which are based on analysis of I-V Langmuir, probe characteristics, are only rough estimations [3], not applicable for accurate calculations such as plasma-chemical processes or similar.

Heat flux during plasma deposition may be measured using the so-called “thermal” probes [4]. Thermal resistor (thermistor) and a water-cooled probe are commonly used for this measurement. The thermal resistor is a tungsten rod, having its temperature measured at both ends (hot end and cold end) by a high-temperature thermocouple system. The second probe is used to measure the absorbed energy, transferred from the plasma to the cooling water. These “thermal” probes are passive devices, i.e. not generating electrical signal, aiming for heat transfer only.

In this paper, we present a novel method for computing basic plasma parameters, using an “active” thermal probe method. A conventional K-type thermocouple (Chromel-Alumel alloy), which is most useful for temperature measurement in the range of 100-1200 °C [5,6], was used as a basic element for the proposed active probe. The thermocouple was inserted into the plasma region without any external electric supply, thus behaving as a floating electrode negatively charged to the plasma. As in common thermocouples, it shows the heat influence of the plasma irradiation. While electrifying the thermocouple system, the measured I-V characteristics are used for extracting basic data on the plasma parameters. This thermoelectric data gives a direct indication on the heat transfer of the plasma and its kinetics. Thus, this novel thermocouple system enables combining the Langmuir probe method with a heat transfer measuring technique.

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2. Experimental details

The experiments were done using a laboratory triode sputtering setup equipped with a standard vacuum system of a residual vacuum of 2×10^{-5} Torr. Detailed description of the sputtering system was given in our previous paper [7]. Fig. 1 presents our sputtering system and the measurement set-up. The sputtering system consisted of a thermo-emissive cathode, an anode, placed opposite to the cathode, and a water-cooled target holder. All of the three electrical sources of the system, namely, the cathode heating supply, the anode supply (applied to anode and cathode), and the sputtering high-voltage supply (applied between target and a special ring electrode within the plasma), were isolated from ground. Two externally mounted electromagnet coils were positioned co-axially on the anode-cathode axis to create a homogeneous magnetic field. This homogeneous field created a unique type of plasma in a thin sheet form, confined between the target and substrate.

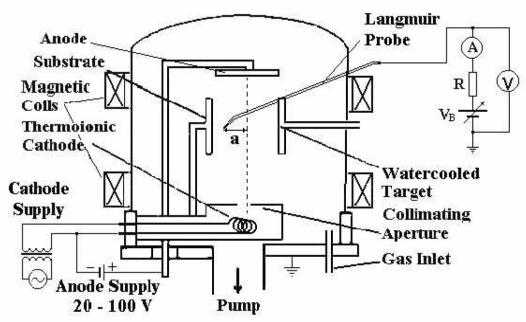

Fig. 1. A sputtering system equipped with Langmuir probe.

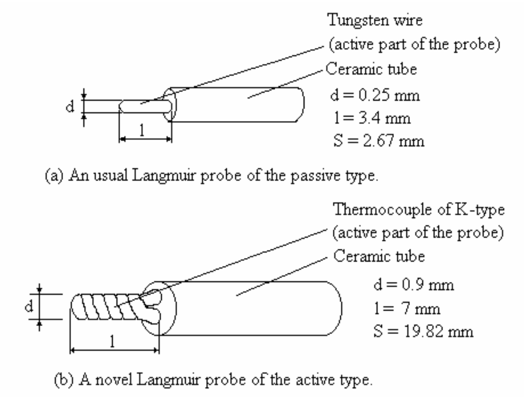

Fig. 2. Two types of Langmuir probes.

Plasma parameters were studied using the Langmuir probe stationary method [8,9]. Two different probes were applied: a passive probe and an active probe. Fig. 2 describes the structure of these probes. The passive probe was made of a tungsten wire, 0.25 mm in diameter, screened by a ceramic tube. The active part of this probe was 3.4 mm long. The surface probe area exceeded 2.67 mm^2. A K type (Chromel-Alumel) thermocouple pair was introduced as an active probe in a cylindrical shape, 0.9 mm in diameter and 7.0 mm long. The active surface area of this probe was 19.82 mm^2. Probes of both types were positioned within the plasma at the same location, defined by a distance **a** from the cathode-anode axis, (Fig. 1). They were biased from –60 V to +60 V through a 5.7 Ω resistor. Measurements were taken in various pressures within the vacuum chamber. At first, measurements were taken using the conventional (passive) Langmuir probe, and then the passive probe was changed on the active one (thermocouple pair).

An artificial plasma discharge of low pressure was used along these experiments. Argon was applied as a working gas. The main source of electrons in this discharge was a white heated cathode. External magnetic field, directed in parallel to the applied voltage between cathode-anode, confined the electrons and created the required conditions for ionization of the argon gas in order to ignite the plasma. This type of the discharge enables the processes at Ar pressure of $(5\text{-}15)\cdot10^{-4}$ Torr with the plasma current of 1-3 A for a plasma voltage of 30-50 V.

3. Theoretical discussion

On the boundary of a metallic surface, electrons leave the plasma and charge the metal surface negatively compared to the plasma. This phenomenon happens due to a large difference in mass between the charged carriers and the electrons, which consequently leads to a large difference in their velocities. In steady state, plasma separates from a metal by a space called sheath that contains a volume charge. Electrons arriving to the metal surface from plasma overcome the potential barrier of the metal work function between vacuum and Fermi level of the metal. The flow of electrons to metallic surface results in heat release at the metal surface due to the energy decrease of electrons during their transfer from plasma to metal. This heat release effect is similar to the Peltier effect [6] in a thermo-electric semiconductor device and may be described by follows formulae:

$$Q_1 = KI \tag{1}$$

where K, similar to the Peltier coefficient, is given by:

$$K = \frac{1}{e}\left[(\Phi_P - E_F) + \frac{3}{2}kT\right] \tag{2}$$

where $\Phi_P = eV_P$, is the electrons energy on the sheath boundary (V_P represents the plasma potential); k is the Boltzmann's constant; e is the elementary charge, and E_F is the Fermi level of the metal for temperature T.

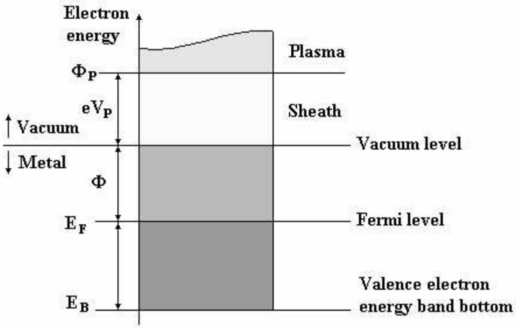

Fig. 3. Energy levels at plasma-metal contacts.

Fig. 3 demonstrates a schematic energetic diagram at the plasma-metal contact. The value of V_P may be approximately found from the I-V characteristics of the probe in the shifted part of the curve. Furthermore, this value of V_p may be estimated from the continuity equation with an assumption of collision-less thin volume charge, and using the Bohm velocity [10]:

$$eV_P = T_e \ln\left(\frac{M}{2\pi m}\right)^{\frac{1}{2}} \tag{3}$$

where T_e is the electron temperature in the plasma boundary, M is the ion mass, and m is the electron mass. The factor $\ln(M/2\pi m)^{1/2}$ equals approximately 4.7 for argon gas. Thus, the plasma potential equals to $eV_P \approx 4.7\ T_e$.

The basic physical phenomenon behind thermocouples claims that when two dissimilar metals are adjoined, a predictable voltage will be generated. This voltage relates to the difference in temperature between the measuring junction (the hot end) and the reference junction (the cold end), connected at the measuring device. Both thermocouple metals obtain electrons from the plasma and behave similarly, although they have different values of Fermi levels (different work functions). That explains a difference in quantities of electrons transferred to the thermocouple conductors at its hot ends from the plasma. When a thermocouple is positioned within the plasma in a steady state, disconnected from an external voltage source, its cold end will generate voltage, which indicates the plasma heat radiation, similarly to the thermoelectric Peltier effect. The electrons drift direction through a metal wire heated in one end and cooled on the other end is from the hot end to the cold end. The potential difference ΔV across this wire due to a temperature difference ΔT, as in the Seebeck effect, equals to:

$$\Delta V = S \cdot \Delta T \tag{4}$$

where S is the Seebeck coefficient, that is also referred to as the thermoelectric power.
Because the voltage across the wire changes proportionally to the temperature and the temperature is the average energy measure, one can find the Seebeck coefficient as follows [6]:

$$S \approx \frac{\pi^2 k^2 T}{2eE_{F0}} \tag{5}$$

where E_{F0} is a Fermi energy at 0^0 K. A thermocouple generates a voltage called EMF (electro-motive force) that equals to the voltage difference between two branches of the thermocouple:

$$EMF = V_{AB} = \frac{\pi^2 k^2}{4e}\left[\frac{1}{E_{FA0}} - \frac{1}{E_{FB0}}\right](T^2 - T_0^2) \tag{6}$$

where T_0 and T are the reference temperature and the temperature of the hot end, correspondingly and the thermocouple Seebeck coefficient:

$$S_{AB} = \frac{\pi^2 k^2}{2e}\left(\frac{1}{E_{FA0}} - \frac{1}{E_{FA0}}\right) \tag{7}$$

The heat capacity of the thermocouple immersed in the plasma consists of two factors: Q_1 (see relation (1)) and Q_2, defined by the cathode heat radiation. Therefore, the absorbed heat by the thermocouple may be given as:

$$cm\Delta T = Q_1 + Q_2 \tag{8}$$

where c is the specific heat of the metal and m is the thermocouple's mass. The heat Q_2 is inversely proportional to the square of the distance cathode-probe. This factor is constant and is not influenced by the external voltage supplied to the thermocouple. Temperature of the thermocouple, defined by the heat Q_2,
Therefore, for the steady state, one can define the thermocouple voltage obtained by the heat of electrons leaving the plasma and absorbed in the thermocouple (without Q_2), using relations (1), (6), (7), and (8), as:

$$U_{AB} = \frac{1}{2} S_{AB}\left[\left(\frac{KI}{cm}\right)^2 + T_0 \frac{2KI}{cm}\right] \tag{9}$$

A positive voltage applied to the active probe of the thermocouple attracts electrons from the plasma and repels the positive ions. However, up to the floating potential point, the thermoelectric voltage defined by the direct heat and photon irradiation, continues to be constant. From the floating potential point and up to the plasma potential, there's a sharp increase in the current from plasma to the probe. Electrons behave according to the Boltzmann distribution in this voltage interval. Thus, the electron current may be given as:

$$I = I_0 e^{\frac{eV}{kT_e}} \tag{10}$$

where V is the bias voltage and I_0 is the steady state current, measured without bias.
The thermoelectric voltage, measured on the active probe, is obtained as:

$$V_{AB} = f(V) = \frac{1}{2} S_{AB} \left[\left(\frac{KI_0}{cm} \right)^2 e^{\frac{2eV}{kT_e}} + T_0 \frac{2KI_0}{cm} e^{\frac{eV}{kT_e}} \right] \tag{11}$$

The measured V_{AB} characteristics enable calculating the quantity of electrons repelled from the plasma and the heat flux transferred from the plasma into the probe.

4. Experimental results and discussion

Passive Langmuir probe characteristics measured for various gas pressures are presented in Fig. 4. The floating potential V_F and the plasma potential V_P are indicated by arrows on these characteristics. At a probe bias voltage $V = V_P$ (30-45 V for various pressures), the probe is at the same potential of the plasma and mainly draws current from the more mobile electrons. This current is designated as positive current, flowing from the probe into the plasma. For V exceeding this value, the probe current tends to saturate at the electron saturation current. The saturation current is defined by the probe geometry (see Fig. 2).

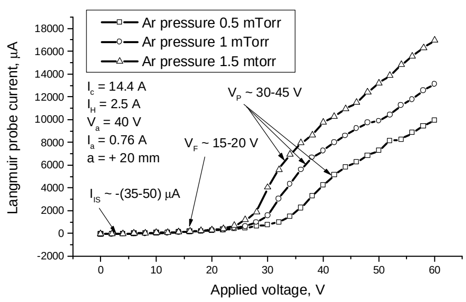

Fig. 4. Passive Langmuir probe graphs.

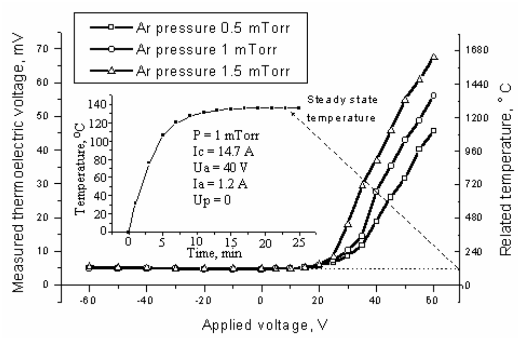

Fig. 5. Thermoelectric probe characteristics.

At $V < V_P$, electrons are repelled, in accordance with the Boltzmann relationship, until at V_F (~15-20 V in our case) the probe is sufficiently negative with respect to the plasma, so that the electron and ion currents become equal and the measured current I = 0. For $V < V_F$, the current is mainly an ion current (negative with respect to the plasma) and tends to saturate at an ion saturation level. This saturation level may also get varied with the applied voltage and due to variations in the probe effective collection area. The value of the ion saturation current is of -(35-50) μA in our measurements. Due to the collisionless thin 3-D charge of the applied model, the electron temperature and the ions concentration are calculated from the experimental graphs at the $V_F<V<V_P$ region [7]. The electron temperature was found of 2 eV on the frontiers of plasma and of 6 eV in the center of the plasma sheet at vacuum of 5×10^{-4} Torr. The ion concentration was found of $(0.8\text{-}1.5)\cdot10^{11}$ cm^{-3}.

The thermocouple, positioned at the same location of the Langmuir passive probe, behaves as a passive probe immersed in plasma, without bias voltage. A measured voltage at the thermocouple is then correlates to the heat obtained by the cathode irradiation and the electrons leaving the plasma due to the sheath formation. The temperature of the thermocouple obtained by the measured voltage at its ends, using the Seebeck's coefficient of the K type thermocouple (S_K = 0.04 mV/°C), is presented as the inset in Fig. 5. It is possible to notice that the thermocouple reaches its steady state position after about 7 minutes. Maximum obtained temperature is no more than 140 °C. Therefore, the heat irradiation becomes constant and may be considered as such in the heat transfer approximation.

It is also interesting to note that the measured thermoelectric voltage characteristics look similar to the Langmuir probe curves as shown in Fig. 5. The right axis here is graduated in the temperature units (°C). The obtained voltage of 54.875 mV is known to correspond to a temperature of 1370 °C for a K-type [11] thermocouple. This is the maximal value in the lookup table. However, in our measurements, the maximal measured thermocouple voltage was obtained as of 67.4 mV, which corresponds to a temperature of 1658 °C. This temperature exceeds the melting points of Ni and Al in the Cromel-Alumel alloy. If we reduce the obtained value to 4.46 mV (a constant factor due the cathode irradiation), the voltage readout reflects directly the active probe detection. This effect may be explained as follows. When a voltage source is connected with its plus to the thermocouple probe, an increase in the electron flux to the probe is resulted. Here, similar to the conventional Langmuir probe, the excess electrons that accumulate at the probe are in direct relation with the increase in the applied voltage. That happens up to a current saturation level shown in the I-V characteristics as a shift in the curve. This shift expresses directly the plasma potential voltage [8]. Thus, the voltage generated by the thermocouple increases relatively to the steady state point. Here again, voltage increase is caused due to two factors: A. Heat by the bombarding electrons, B. Dissimilarity in electron quantities, carried out in both thermocouple branches. This dissimilarity may be of higher values, and the measured voltage between the thermocouple branches may reflect temperatures that are higher than the melting point of the thermocouple metals.

5. Conclusions

A K-type thermocouple was engaged as an active probe for plane plasma investigation. Obtained thermoelectric voltages exceeded the maximum possible temperature values for such thermocouples. This excess in voltage was found to be a contribution of the electron flow from the plasma into the metal branches of the thermocouple. Thus, basic plasma parameters may be estimated using the above-mentioned theory in conjunction with the thermocouple measured voltage. Application of thermocouples as active Langmuir probes enables studying of metal contacts by using the I-V plasma characteristics. Conversely, plasma parameters may be studied by using thermoelectric voltage measurements.

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