Principle and application of a thermal probe to reactive plasmas

E. Stamate[a)] and H. Sugai[b)]
Department of Electrical Engineering, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

K. Ohe
Department of Systems Engineering, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan

(Received 22 January 2002; accepted for publication 26 February 2002)

A thermal probe for plasma diagnostics is introduced. The method is based upon measuring the equilibrium temperature of a conducting sphere as a function of its applied bias. The resulting temperature–voltage characteristic is processed using a theoretical model that accounts for charge and thermodynamic balance. The thermal probe is capable of detecting negative ions and shows sensitivity to certain chemical reactions. Measurements performed in Ar, Ar/SF_6, and O_2 show good agreement among the plasma parameters using thermal and Langmuir probes. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1473688]

Reactive plasmas produced in gases such as O_2, CF_4, Cl_2, and SF_6 are used extensively in microelectronic technologies.[1] In order to control the processing one should know not only the main plasma parameters (plasma density, n_0 and electron temperature, T_e) but also the density of negative ions, n_{ni}. There are several diagnostic methods available for these measurements including mass spectrometry,[2] optical emission[3] and photodetachment[4] that are reliable but it is rather expensive to equip each plasma reactor. On the other hand, electrostatic probes are simple to construct but require complicated analysis in order to obtain the plasma parameters[5] and the results can be seriously influenced by the probe's surface condition.[6]

In this letter, we introduce a diagnostic method based upon processing the temperature–voltage characteristic of a spherical probe.

A spherical probe of radius r_p biased at potential V is immersed into a plasma (it may also include negative ions of density n_{ni} and temperature T_{ni}) of positive ion density $n_i \cong n_0$. Depending on V, charges can reach the probe, resulting in heat transfer. Following the formalism introduced[7,8] the energy balance of the probe is given by

$$\frac{4}{3}\pi r_p^3 \rho C \frac{dT_p}{dt} = Q_H - 4\pi r_p^2 (q_R + q_K), \tag{1}$$

where ρ is the probe material's mass density, C the heat capacity, q_R the radiative cooling term, q_K the heat flux due to Knudsen conduction, and T_p is the probe temperature. The heating rate Q_H due to positive and negative charges impact and electron–ion recombination can be written as

$$Q_H = Q_{H,e} + Q_{H,i} + Q_{H,ni}, \tag{2}$$

where indices e, i and ni correspond to electrons, positive ions and negative ions, respectively. The heating rate $Q_{H,e}$ for $V \leqslant V_{\text{pl}}$ (V_{pl} is the plasma potential) is given by

$$\mathrm{d}Q_{H,e} = \frac{\mathrm{d}I_e}{e}\frac{m_e v_e^2}{2}, \tag{3}$$

where $\mathrm{d}I_e/e$ is the number of electrons per interval of time that reach the probe with velocity v_e. Also we have

$$\frac{\mathrm{d}I_e}{e} = S v_e \mathrm{d}n_e = S v_e n_e f(v_e)\mathrm{d}v_e. \tag{4}$$

Assuming Maxwellian distribution of the electrons, $Q_{H,e}$ becomes

$$Q_{h,e}(V) = \pi S n_e \frac{m_e}{2}\left(\frac{m_e}{2\pi k T_e}\right)^{3/2} \int_{v_{\min}}^{\infty} v_e^5 \left(1 - \frac{v_{\min}^2}{v_e^2}\right) \times \exp\left(-\frac{m_e v_e^2}{2kT_e}\right)\mathrm{d}v_e, \tag{5}$$

which finally gives

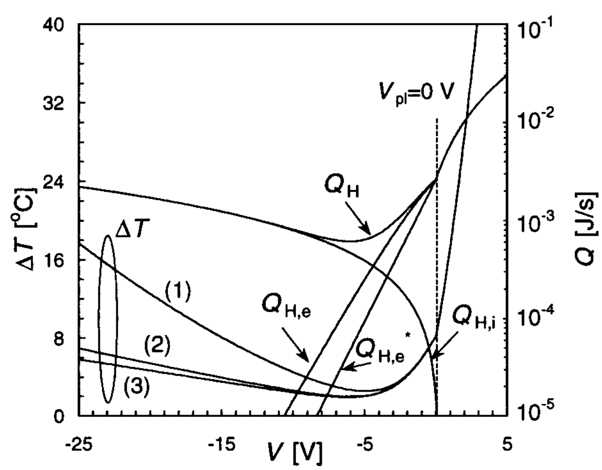

FIG. 1. Q_H, $Q_{H,e}$, $Q_{H,i}$, $Q_{H,e}^{*}$ and $\Delta T = T_p - T_n$ in Ar, numerically obtained by Eq. (1) at thermal equilibrium ($\mathrm{d}T_p/dt$) = 0, where $T_e = 1.5$ eV, $T_i = 0.15$ eV, $V_{\text{p1}} = 0$ V, $T_n = 300$ K, $p = 1$ mTorr, $r_p = 2$ mm, $\phi = 5.4$ eV, $n_0 = 5\times 10^{14}$ m^{-3}, $\epsilon_R = a_R = 1$, $\alpha = 1$, $\xi = 0$, $\eta = 1$ [(1) $\zeta = 1$, (2) $\zeta = 0.1$ and (3) $\zeta = 0.01$].

[a)]Also at Faculty of Physics, Al. I. Cuza University, B-dl Carol No. 11, Iasi-6600, Romania; electronic mail: stamate@nuee.nagoya-u.ac.jp
[b)]Electronic mail: sugai@nuee.nagoya-u.ac.jp

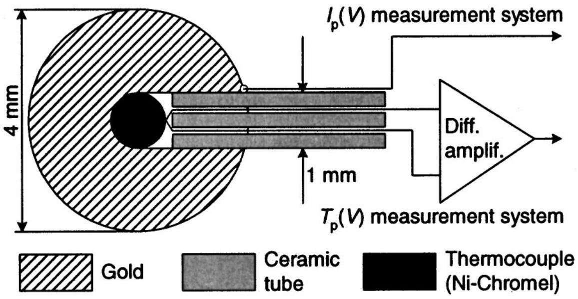

FIG. 2. Details of the construction of the thermal probe.

$$Q_{H,e}(V)=n_e S\frac{kT_e}{\sqrt{\pi}}\sqrt{\frac{2kT_e}{m_e}}\left(\frac{eV}{2kT_e}+1\right)\exp\left(\frac{eV}{kT_e}\right), \quad (6)$$

where S is the probe surface. A similar expression can also be deduced for $Q_{H,ni}$. The heat rate $Q_{H,i}$ is given by

$$Q_{H,i}(V)=\frac{I_i(V)}{e}[\zeta eV+\eta e(\varepsilon_i-\phi)], \quad (7)$$

where $I_i(V)$ is the positive ion current collected for $V \leqslant V_{\mathrm{pl}}$, ε_i the ionization energy, ϕ the work function of the probe material, ζ a coefficient that expresses the transfer efficiency of the kinetic energy between the positive ion and the probe and η is a coefficient that describes the inelastic energy transfer. The q_R is given by Stefan–Boltzmann law,

$$q_R=\sigma(\varepsilon_R T_p^4-a_R T_n^4), \quad (8)$$

where σ is the Stefan–Boltzmann constant, ε_R the probe emissivity, a_R the absorptivity and T_n the gas temperature.

The heat flux q_K is given by

$$q_K=\frac{1}{4}\,\frac{\gamma+1}{4(\gamma-1)}\,\frac{p}{\sqrt{T_n}}\left(\frac{8k}{\pi m_i}\right)^{1/2}\alpha(T_p-T_n), \quad (9)$$

where p is the pressure, γ the gas heat capacity ratio and α the accommodation coefficient.

Q_H, $Q_{H,e}$, $Q_{H,i}$, $Q^*_{H,e}$ and $\Delta T=T_p-T_n$ in Ar, numerically obtained by Eq. (1) at thermal equilibrium $(\mathrm{d}T_p/\mathrm{d}t)=0$ are presented in Fig. 1, where as example, $T_e=1.5$ eV, $T_i=0.15$ eV, $V_{\mathrm{pl}}=0$ V, $T_n=300$ K, $p=1$ mTorr, $r_p=2$ mm, $\phi=5.4$ eV, $n_0=5\times10^{14}\ \mathrm{m}^{-3}$ $\varepsilon_R=a_R=1$, $\alpha=1$, $\eta=1$, $\xi=0$, and $\zeta=1$, 0.1, and 0.01, respectively. $Q^*_{H,e}$ is computed by assuming that each electron impacts the probe's surface with an average electron energy of $E_m=2kT_e$. For $p<100$ mTorr q_K was found to be negligible. $I_i(V\leqslant V_{\mathrm{pl}})$ and $I_e(V\geqslant V_{\mathrm{pl}})$, were expressed in the orbital motion limited model,[9]

$$I_{i,e}(V)=en_{i,e}S\sqrt{\frac{kT_B}{2\pi m_{i,e}}}\frac{2}{\sqrt{\pi}}\left(1\pm\frac{e(V_{\mathrm{pl}}-V)}{kT_B}\right). \quad (10)$$

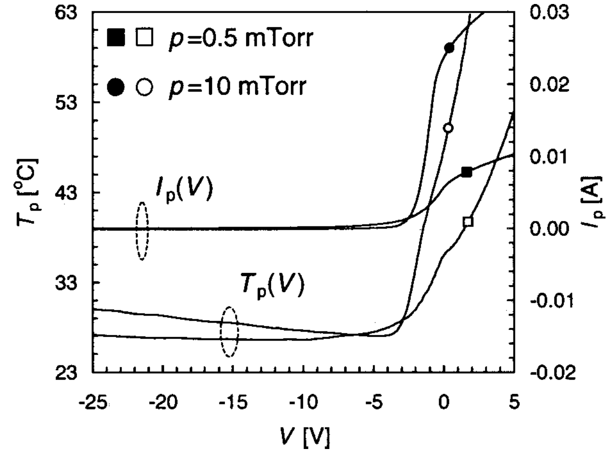

FIG. 3. $I_p(V)$ (closed symbols) and $T_p(V)$ (open symbols) in Ar for $I_{\mathrm{d}}=100$ mA and $U_{\mathrm{d}}=100$ V.

T_B is the "effective" temperature given by $T_B=T_eT_i(\xi+1)/(\xi T_e+T_i)$ where T_i is the positive ion temperature and ξ the negative ion to electron density ratio, $\xi=n_{ni}/n_e$. For $V\leqslant V_{\mathrm{pl}}$, $Q^*_{H,e}$ shows linear variation. ΔT is parabolic for $\zeta=1$ $(V\ll V_{\mathrm{pl}})$ and changes linearly for $\zeta\leqslant 0.1$ (a low ζ is expected because $m_i \ll m_p$).

By using $T_p(V)$ we have several ways by which to detect the plasma parameters. For instance, if V_1 and V_2 are two distinct potentials with $V_1<V_2\ll V_{pl}$ and $\Delta V=V_2-V_1$ then using Eqs. (1), (7), (8) and (10) one can deduce the following relation:

$$n_0=\frac{\pi\sigma\varepsilon_R(T_{p1}^4-T_{p2}^4)}{\psi\left[\zeta\left(1-\frac{e\Delta V}{kT_B}+2\frac{e|V_1|}{kT_B}\right)+\eta\left(\frac{e(\varepsilon_i-\phi)}{kT_B}\right)\right]}, \quad (11)$$

where $\psi=e\Delta V(2kT_B/m_i)^{1/2}$ and $T_{p1}=T_p(V_1)$ and $T_{p2}=T_p(V_2)$. Equation (11) is independent of a_R and for low ζ it becomes linear with respect to ΔV. If Eq. (1) is expressed for $T_p(V_f)$ and $T_p(V_{pl})$ then one can obtain

$$\xi=\frac{\sigma\varepsilon_R[[T_p(V_{pl})]^4-[T_p(V_f)]^4]-n_0(\Omega_e+\Lambda_i)}{n_0(\Omega_{ni}+\Lambda_i)-\sigma\varepsilon_R[T_p(V_{pl})]^4-[T_p(V_f)]^4}, \quad (12)$$

where

$$\Omega_{e,ni}=kT_{e,ni}\sqrt{\frac{2kT_{e,ni}}{\pi m_{e,ni}}}\left[1-\left(\frac{eV_f}{kT_{e,ni}}\right)\exp\left(\frac{eV_f}{kT_{e,ni}}\right)\right], \quad (13a)$$

$$\Lambda_i=\frac{e|V_f|}{\pi}\sqrt{\frac{2kT_B}{m_i}}\left[\zeta\left(\frac{eV_f}{kT_B}-1\right)-\eta\frac{\varepsilon_i-\phi}{kT_B}\right]. \quad (13b)$$

In order to obtain ξ by Eq. (12) one should estimate T_e from V_f or by fitting it from the electron retarding region of the

TABLE I. Plasma parameters obtained by thermal probe (index th) and by Langmuir probe, where V_q^{th} corresponds to the voltage that gives the minimum value of $T_p(V)$. T_e^{th} is the electron temperature obtained by fitting and $T_e^{\mathrm{th}*}$ is that by semilogarithmic plotting of $T_p(V)$.

p (mTorr)	T_e (eV)	T_e^{th} (eV)	$T_e^{\mathrm{th}*}$ (eV)	n_0 (m^{-3})	n_0^{th} (m^{-3})	V_{pl} (V)	$V_{\mathrm{pl}}^{\mathrm{th}}$ (V)	V_f (V)	V_q^{th} (V)
0.5	4.6	4.8	5.7	2.4×10^{15}	1.9×10^{15}	−0.39	−0.6	−32.6	−12.6
10	0.8	0.7	1.3	1.2×10^{16}	9.3×10^{15}	−1.1	−1.4	−6.17	−4.2

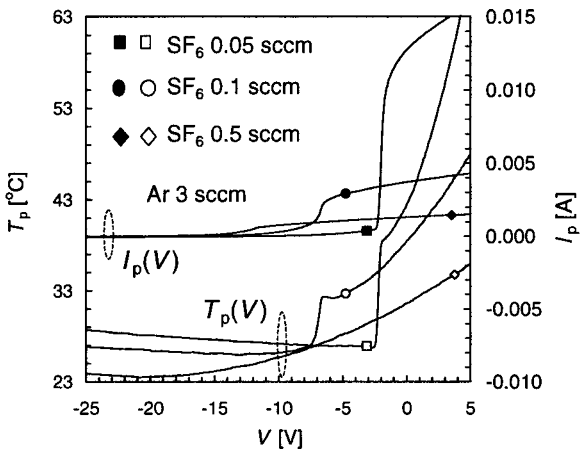

FIG. 4. $I_p(V)$ (closed symbols) and $T_p(V)$ (open symbols) in Ar/SF_6 for various SF_6 flows where $I_d = 100$ mA, $U_d = 100$ V and $p = 1$ mTorr.

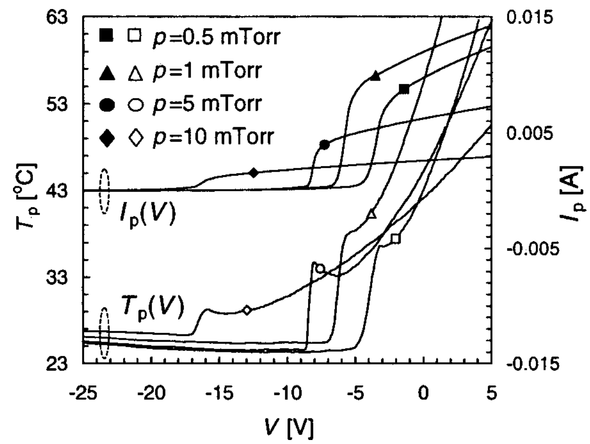

FIG. 5. $I_p(V)$ (closed symbols) and $T_p(V)$ (open symbols) in O_2 for various p where $I_d = 100$ mA and $U_d = 100$ V.

$T_p(V)$ and then detect n_0 by Eq. (11). Because Eqs. (11) and (12) are slightly dependent on T_{ni} it is a satisfactory approximation to suppose $T_i \sim T_{ni} \cong T_e/10$.

Experiments were performed in a cylindrical, multipolar, magnetically confined device, the details of which are presented elsewhere.[10] Two identical spherical probes, 4 mm in diameter and made of gold (see Fig. 2 for details of their construction), were available to measure $T_p(V)$ by thermocouples mounted at their centers. A 790 Fluoroptic® system was used to determine the calibration coefficient between the surface and inside temperature. The probe current, $I_p(V)$, was measured simultaneously with $T_p(V)$. In order to ensure thermal equilibrium the acquisition time of one single point of $T_p(V)$ was set to 10 ms.

$I_p(V)$ and $T_p(V)$ curves obtained in Ar for two different pressures, p, are presented in Fig. 3, where the discharge current $I_d = 100$ mA and the discharge voltage $U_d = 100$ V. From $T_p(V)$ one can identify by similitude with $I_p(V)$ the positive ion saturation region for $V \ll V_{pl}$; the retarding field region for electrons followed by an inflection point that corresponds to V_{pl} for $V \leqslant V_{pl}$, and the electronic saturation region for $V > V_{pl}$. At $p = 0.5$ mTorr we detected $T_e = 4.6$ eV that together with the contribution by the primary electrons led to $V_f = -32$ V and a nearly flat $T_p(V)$ for $-20 < V < -10$ V. However, for $p = 10$ mTorr the electrons approached a Maxwellian distribution function with $T_e = 0.8$ eV, so that we obtained a linear increase of T_p, with V decreasing for $V < -5$ V. From this, correlated with $T_p(V_{pl}) \gg T_p(V = -25\text{ V})$, we concluded that $\zeta \ll 1$. Using n_0 and T_e from $I_p(V)$ we detected $\varepsilon_R = 0.32$ and $\eta = 0.75$ using Eqs. (11) and (12) assuming that $\zeta = 0.05$. A value of $\eta < 1$ is the result of energy losses by secondary emission and internal energy transfer during positive ion neutralization.[11] The plasma parameters obtained by thermal probe (index th) and by Langmuir probe are presented in Table I, where V_q^{th} corresponds to the voltage that gives the minimum value of $T_p(V)$. T_e^{th} is the electron temperature obtained by fitting Eq. (1) for $(\mathrm{d}T_p/\mathrm{d}t) = 0$ and $T_e^{\text{th}*}$ by semilogarithmic plotting of $T_p(V)$. V_{pl}^{th} was decided to be the voltage that corresponds to the maximum value of the first derivative of $T_p(V)$ and n_0^{th} was obtained by Eq. (12) for $\xi = 0$ and $T_B = T_e^{\text{th}}$. Good agreement was found between T_e and n_e using thermal and Langmuir probes. Since in the retarding region $T_p(V)$ was not only an exponential dependent of V, $T_e^{\text{th}*}$ overestimated T_e by a factor of about 1.5.

Measurements were also performed in electronegative plasmas of Ar/SF_6 and O_2. In Ar/SF_6 $T_p(V_{pl})$ decreased with an increase in n_{ni} due to the reduction in electron flux by attachment as can be seen in Fig. 4. $T_p(V)$ was processed as follows. T_e was detected from V_f ($T_e \gg T_{ni}$). By initially forcing $T_B = T_e$ in Eq. (11) an intermediate value of n_0 ($= n_e + n_{ni}$) was obtained. Then, the resulting value of ξ using Eq. (12) was corrected by reconsidering $T_B = T_e(\xi + 1)/(10\xi + 1)$ in both Eqs. (11) and (12). Finally, the agreement for ξ was within 20% compared with that from $I_p(V)$.[10] In O_2 plasma we observed that $T_p(V)$ decreased for V slightly higher than V_{pl} and then increased exponentially with V as is seen in Fig. 5. This result may be related to some endothermic chemical reaction on the probe's surface, a phenomenon that could not be observed from $I_p(V)$. Using Eq. (12) again, we detected $\xi = 8$ for $p = 10$ mTorr while $\xi = 5$ from $I_p(V)$.

In conclusion, we introduced a thermal probe that is capable of detecting n_0, V_{pl}, T_e and ξ. Moreover, one can obtain other important coefficients that characterize the plasma–probe system, such as ζ, η, ε_R, and a_R.

This work was partially supported by the Japan Society for the Promotion of Science.

[1] M. A. Liberman and A. J. Lichtenberg, *Principles of Plasma Discharges and Materials Processing* (Wiley, New York, 1994), p. 472.
[2] V. M. Donnelly, J. Appl. Phys. **79**, 9353 (1996).
[3] K. L. Junck and W. D. Getty, J. Vac. Sci. Technol. A **12**, 2767 (1994).
[4] M. Bacal, Rev. Sci. Instrum. **71**, 3981 (2000).
[5] E. Stamate and K. Ohe, J. Appl. Phys. **89**, 2058 (2001).
[6] E. Stamate and K. Ohe, Appl. Phys. Lett. **78**, 153 (2001).
[7] K. G. Emeleus and A. C. Breslin, Int. J. Electron. **29**, 1 (1970).
[8] J. E. Daugherty and D. B. Graves, J. Vac. Sci. Technol. A **11**, 1126 (1993).
[9] J. G. Laframboise, Report No. 100, Institute of Aerospace Studies, University of Toronto (1966).
[10] E. Stamate and K. Ohe, J. Appl. Phys. **84**, 2450 (1998).
[11] V. Grill, J. Shen, C. Evans, and R. G. Cooks, Rev. Sci. Instrum. **72**, 3149 (2001).