

INSTITUTE OF PLASMA PHYSICS

NAGOYA UNIVERSITY

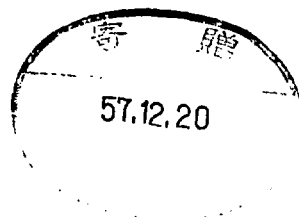
Study of Discharge Cleaning Process  
in JIPP T-II Torus by Residual Gas Analyzer

N. Noda, S. Hirokura, Y. Taniguchi, S. Tanahashi

(Received - Nov. 9, 1982)

IPPJ- 618

Dec. 1982



# RESEARCH REPORT

NAGOYA, JAPAN

Study of Discharge Cleaning Process  
in JIPP T-II Torus by Residual Gas Analyzer

N. Noda, S. Hirokura, Y. Taniguchi, S. Tanahashi

(Received - Nov. 9, 1982)

IPPJ- 618

Dec. 1982

Further communication about this report is to be sent to the  
Research Information Center, Institute of Plasma Physics,  
Nagoya University, Nagoya 464, Japan

## Abstract

During discharge cleaning, it has been observed that decay time of water-vapor pressure changes when the pressure reaches a certain level. A long decay time observed in the later phase can be interpreted as a result of a slow deoxidization rate of chromium oxide, which may dominate the cleaning process in this phase. Optimization of plasma density for the cleaning is discussed comparing the experimental results on density dependence of water-vapor pressure with a result based on a zero-dimensional calculation for particle balance. One of the essential points for effective cleaning is found to be raising electron density of the plasma high enough that the dissociation loss rate of  $H_2O$  is as large as the sticking loss rate. A density as high as  $10^{11} \text{ cm}^{-3}$  is required for a clean surface condition where sticking probability is presumed to be around 0.5.

## 1. Introduction

Effective discharge cleaning is important because, in future tokamaks, in-situ titanium gettering may be unfavorable. In D-T burning experiments, the titanium gettering technique cannot be used because of the necessity to manage a finite tritium inventory. For experiments on plasma-wall interactions using a tokamak machine, titanium gettering often confuses the results relating to origin of impurities. A titanium flake exfoliated from the wall makes another trouble in tokamak experiments. After flashing a large amount of titanium, it is sometimes observed that the exfoliated flake induces a severe disruption of the tokamak discharge. In spite of those defects, it has been widely used in present-day tokamaks because of its strong effectiveness for reducing low-Z impurities coming into a plasma<sup>1)</sup>. Discharge cleaning has been also used successfully in tokamaks for the same purpose<sup>1-8)</sup>. In some cases, however, it has been reported that the gettering is superior to the discharge cleaning for low-Z impurity reduction<sup>7,8)</sup>. In Doublet III, maximum density of tokamak plasma obtainable after gettering is higher than that after discharge cleaning<sup>8)</sup>. Thus, it is necessary to improve discharge cleaning technique and find an optimized condition of this technique for low-Z impurity reduction.

In JIPP T-II, we have made efforts to obtain high density, high temperature plasmas without titanium gettering by optimizing discharge cleaning technique. We have used 2.45 GHz

ECR and 60 Hz AC discharge for the cleaning. Effects of the discharge cleaning on JIPP T-II tokamak plasmas are given in Ref. [9] and [10]. A residual gas analyzer (RGA) has been used as a principal technique for analyzing the cleaning process. In this paper, we present some remarkable features found in behavior of water-vapor pressure during the cleaning and discuss the results in relation to the elementary processes on the wall surface. Modeling of the cleaning process and understanding the elementary processes are important for optimizing discharge cleaning. We modify a zero-dimensional calculation for particle balance in glow discharge cleaning proposed by Waelbroeck et al.<sup>11,12)</sup> and apply it to the cleaning process with ECR and AC plasma, in which chemically active hydrogen atoms play a dominant role for the cleaning<sup>10)</sup>. Experimental results obtained by RGA are compared and discussed with the calculated results.

## 2. Experimental arrangement

The vacuum vessel of the JIPP T-II device is made of stainless steel (SUS 304) and its major radius is 91 cm, minor radius is 20 cm. A main pumping system consists of two turbo-molecular pumps with a liquid-nitrogen trap. The effective pumping speed is 760 l/s for H<sub>2</sub>. An RGA (MSQ - 150 : ULVAC Co.) is attached to a manifold of the pumping system and is not differentially pumped. The time constant of the RGA measurement is determined from volume of the manifold and conductance between the main

chamber and the manifold. It is shorter than 0.1 s. We have paid attention mainly to partial pressure of  $H_2O$  ( $P_{18}$ ),  $CH_4$  ( $P_{16}$ ), during discharge cleaning.

We have investigated  $P_{18}$  as a function of wall temperature and plasma parameters. Plasma density and temperature were measured with a Langmuir probe for ECR plasma<sup>10)</sup>. For the AC discharge, the density was measured with 4 mm microwave interferometer and the temperature was determined by ratio of electric field strength to hydrogen pressure<sup>9)</sup>.

All the data shown in this paper were obtained after two days' pre-baking at 350 C which followed a few days' venting.

### 3. Variation of $P_{18}$ during long-run discharges

Figure 1 shows a typical behavior of the RGA signal. During AC discharge, it can be seen that the signals for  $m = 16$ , 18 and 28 rise greatly compared with the background levels. Figure 2 shows a result for time variations of  $P_{18}$  and  $P_{16}$  during discharge cleaning. Partial pressure is expressed by direct reading of output signal  $I_{RGA}$  in these figures. One can roughly obtain absolute values of the pressure in mbar by multiplying  $I_{RGA}$  by 7. The result in Fig. 1 was obtained for a wall temperature of 200 C after two days pre-baking at 350° C. The level and variation of  $P_{28}$  ( $CO + C_2H_4$ ) are almost the same as those of  $P_{16}$  in this case. In the later phase,  $P_{18}$  still decreases with a decay time of 40 h while  $P_{16}$  stays almost constant. This suggests an existence of some diffusion processes for carbon inside the wall.

Figure 3 shows time behavior of  $P_{18}$  for various wall temperatures  $T_w$ . One of the important feature of the result is that, for all variations of the wall temperature, the decay time of  $P_{18}$  changes at a time when  $P_{18}$  reaches a certain level. This suggests that a mechanism which dominates in the later phase is different from the one in the earlier phase. We have considered some possible causes which give rise to this critical change of the decay time.

The first is that the dominant mechanism in the later phase is indirect cleaning of the wall section where hydrogen atoms from the plasma cannot irradiate directly, such as cleaning of port sections. If this is true, the decay time in the later phase has relation to a spontaneous desorption rate of  $H_2O$ , not stimulated by hydrogen impact. The JIPP T-II device has only small number of ports and, inside the chamber, there is no object of complicated structure, such as microwave antennas, magnetic probes and so on in this series of the experiment. This means most of the inner surface of the chamber is irradiated by hydrogen atoms. In spite of such an environment, we cannot decisively conclude that the indirect cleaning process is not significant in JIPP T-II and it has no relation to the result shown in Fig. 3.

The second interpretation is that the difference in the decay time reflects the difference between oxygen-reduction rates for oxygen surface density of larger than one monolayer and that for density of smaller than one monolayer. It is

possible that  $H_2O$  production rate and/or sticking probability of  $H_2O$  change at around the oxygen surface density of one monolayer. It has been, however, found that  $Z_{eff}$  of tokamak is close to 1.0 when  $P_{18}$  falls as low as that in the later phase<sup>9)</sup>, and in such little contaminated cases, surface density of oxygen is presumed to be as low as 0.01 monolayer<sup>3)</sup>. This is inconsistent with the second interpretation, in which the critical change in decay time must occur at a density of around one monolayer.

The third interpretation is that the critical change in decay time is due to the difference between deoxidization rate of iron oxide and that of chromium oxide. It has been found in ISX<sup>5)</sup> from surface probe analysis that chromium oxide can also be deoxidized by low energy hydrogen atoms. In PDX, chemical behavior of chromium was also investigated during glow discharge cleaning<sup>13)</sup>, where a partial reduction was observed on iron oxide but not on chromium oxide. In the PDX case, however, proton flux incident on the wall is in the order of  $10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ , which is two orders smaller than the atomic hydrogen flux in the case of AC discharge on JIPP T-II. This difference in the flux intensity is presumed to be a reason why deoxidization of the chromium oxide cannot be observed in glow discharge cleaning.

In this experiment, we have obtained by RGA another kind of experimental data which supports the third interpretation. As is shown in Fig. 4, temperature dependence of  $P_{18}$  obtained in the later phase is weaker than that obtained by Juelich group



for glow discharge<sup>11)</sup>, which is a result probably for a less cleaned surface condition than the present condition because both conditioning time and particle flux are smaller in the Juelich's case than those in our case. An apparent activation energy is 16 kcal/mol in this experiment. Taking account of an activation energy of H<sub>2</sub> release as 16 kcal/mol<sup>11)</sup>, an activation energy of water formation is 32 kcal/mol, which is 1.5 times larger than 21.8 kcal/mol obtained by Juelich group<sup>11)</sup>. Since a larger value of the activation energy corresponds to larger binding energy, deoxidization of chromium oxide may possibly be dominant process in the later phase. It should be, however, noted that, if the indirect cleaning is dominant, the difference in H<sub>2</sub>O production mechanism may also affect the apparent activation energy.

Although there remains a possibility of indirect cleaning process, we have tentatively concluded that the long decay time of P<sub>18</sub> in the later phase is due to slow deoxidization rate of chromium oxide.

Another important feature of the result in Fig. 3 is that the decay of P<sub>18</sub> becomes faster as the wall temperature is raised except the later phase of 250° C. For temperatures lower than 200° C, the result is qualitatively consistent with the relation between P<sub>18</sub> and T<sub>w</sub> shown in Fig. 4. A long decay time has also been observed, however, for the wall temperature of 300° C. The results for 250° and 300° C cases show that the decay time of oxygen surface density becomes longer as the wall

temperature is raised above  $250^{\circ}$  C although the reduction rate increases with temperature, as is shown in Fig. 4. This suggests an existence of some processes which supply oxygen to the surface region. As was mentioned before in this section, spontaneous desorption process from the wall section not irradiated by hydrogen atoms possibly plays a role in the cleaning process. Diffusion of oxygen inside the wall materials is also a possible process with which we can explain the longer decay time for wall temperatures higher than  $250^{\circ}$  C. Before discharge cleaning, we usually bake out the vessel at  $350^{\circ}$  C for two days. During the pre-baking phase, the oxidized layer may possibly be expanded by inward diffusion of oxygen. Since the discharge cleaning reduces oxygen just on the top layer of the surface, outward diffusion of oxygen can be brought about during the cleaning phase. For temperatures higher than  $250^{\circ}$  C, oxygen supplied by this outward diffusion may play a role to elongate the apparent reduction time of the surface oxygen density.

#### 4. Density dependence of $P_{18}/P_2$

In order to study the relation between oxygen reduction rate and parameters of a cleaning plasma, we have investigated density dependence of  $P_{18}$ . Plasma density  $n_e$  was varied by changing a primary voltage  $U_0$  of AC discharge. Input power of AC discharge was 9.2, 2.2, 0.7 and 0 kW for  $U_0 = 1600, 1000, 600$  and 0 volt, respectively. That of ECR discharge was around 0.5 kW for each case of  $U_0$ . Hydrogen-gas pressure was arranged to

give the same electron temperature for each case and the value of the electron temperature is around  $8 \text{ eV}^9$ ). The results are shown in Fig. 5 by closed circles.

We have proposed a simple model to explain the result in Fig. 5 and to find an optimum plasma parameters for cleaning<sup>9</sup>). The reduction of oxygen consists of the following three processes: i) generation of an atomic hydrogen flux in the plasma, ii) production of water vapor by chemical reactions at the wall surface, iii) pump-out of the water vapor by the pumping system. In this model, we have taken account of  $\text{H}_2\text{O}$  molecules which are lost due to sticking to the wall and  $\text{H}_2\text{O}$  dissociation by electron impact. Both of these processes are competitive to the pump-out process (iii) of  $\text{H}_2\text{O}$ . This model is similar to the one proposed by Waelbroeck et al.<sup>11)</sup> except that atomic hydrogen mainly contributes to  $\text{H}_2\text{O}$  production in our case. Since the production rate of hydrogen atom is written as  $n_e n_2 k_2$  for a unit volume, the total number of hydrogen atoms incident on whole of the wall surface during a unit time is  $F_2 = n_e n_2 k_2 V_p$ , where  $n_2$  is molecular hydrogen density,  $k_2$  rate coefficient for production of one H atom by electron-impact dissociation of  $\text{H}_2$ ,  $V_p$  plasma volume. Denoting effective cross section of water production on the wall surface as  $s_{18}$ , we obtain the total number of  $\text{H}_2\text{O}$  released from the wall during a unit time as

$$F_{18} = F_2 n_{\text{MO}} s_{18} = n_{\text{MO}} s_{18} n_e n_2 k_2 V_p \quad (1),$$

where  $n_{MO}$  is surface density of metal oxide on the wall. When the density of  $H_2O$  is  $n_{18}$ , the pump-out rate  $F_p$  of  $H_2O$  by the pumping system which has an effective pumping speed  $S_p$  is written as

$$F_p = S_p n_{18} \quad (2).$$

Moreover, we take account of dissociation loss  $F_d$  as

$$F_d = n_e n_{18} k_{18} v_p \quad (3),$$

and sticking loss  $F_{st}$  as

$$F_{st} = (1/4) n_{18} v_{18} A_w p_{st} \quad (4),$$

where  $k_{18}$  is rate coefficient of electron impact dissociation of  $H_2O$ ,  $v_{18}$  thermal velocity of  $H_2O$  molecules,  $p_{st}$  sticking probability and  $A_w$  area of the wall surface. In a steady state, a relation  $F_{18} - F_p - F_d - F_{st} = 0$  holds. Substituting (1) - (4) into this relation, we obtain the following relation,

$$P_{18}/P_2 = n_{18}/n_2 = A / (1 + B n_e^{-1}) \quad (5),$$

where  $A = (k_2/k_{18}) s_{18} n_{MO}$ ,  $B = [(1/4) p_{st} v_{18} A_w + S_p] / k_{18} v_p$ .

This relation indicates that, for sufficiently high electron densities,  $n_{18}/n_2$  approaches  $(k_2/k_{18}) s_{18} n_{MO}$  asymptotically and is saturated. This saturation should be seen in the densities higher than  $B$ . This implies that one can improve the cleaning time by raising  $n_e$  up to around  $B$ .

In order to estimate the value  $B$ , it is necessary to know  $k_2$ ,  $k_{18}$  and  $p_{st}$ . Dissociation cross sections of  $H_2O$  have been measured by several authors and summarized by Olivero et al.<sup>14)</sup> We have calculated  $k_{18}$  using the data in this literature and plotted against electron temperature in Fig. 6. In this figure,  $k_2$  estimated from the data given in Ref. [15] are also plotted. As is shown in this figure, the ratio  $k_2/k_{18}$  decreases as the electron temperature rises. It has been pointed out qualitatively by Oren and Taylor<sup>2)</sup> that low electron temperature of the cleaning plasma is essentially important for oxygen removal. The result in Fig. 6 shows quantitatively that their remark is true.

Within the present knowledge, sticking probability of stainless steel for  $H_2O$  is not known at 150 C. It is, however, reasonable to consider that  $p_{st}$  is larger than 0.1<sup>16)</sup> because the data in Fig. 5 have been obtained after long time cleaning and surface density of oxygen is probably as small as 0.01 monolayer. By this assumption, we can neglect  $S_p$  in calculating the value of  $B$  and we have obtained  $B = 2.2 \times 10^{11} \text{ (cm}^{-3}\text{)}$  for  $p_{st} = 0.5$  and  $T_e = 8 \text{ eV}$ . We have calculated  $P_{18}/P_2$  for two values of  $p_{st}$ , 0.5 and 0.1, and plotted in Fig. 5. According to Eq. (5), a low value of  $p_{st}$  corresponds to a low value of  $B$  and this implies a saturation in  $P_{18}/P_2$  occurs at a low  $n_e$ . We can see in Fig. 5 that the calculated result for  $p_{st} = 0.5$  is closer to the experimental data than that for  $p_{st} = 0.1$ . In other words, the experimental results in Fig. 5 suggest that the

sticking probability is as large as 0.5 even though the wall temperature is raised up to 150° C.

From the above considerations, we can conclude that it is essential to raise the electron density so high that the value of  $n_e n_{18} k_{18} V_p$  (dissociation-loss rate) is as large as  $P_{st} (n_{18}/4) v_{18} A_w$  (sticking-loss rate) in order to optimize discharge cleaning. We also presume that  $P_{st}$  is considerably large after a long time cleaning, which requires plasma density as high as  $10^{11} \text{ cm}^{-3}$  for optimum cleaning.

#### Summary

From an observation of water-vapor pressure  $P_{18}$  during discharge cleaning, it has been found that the decay time of  $P_{18}$  changes at a certain level. A possible interpretation is that deoxidization of chromium oxide is a dominant process in the later phase of the cleaning while that of iron oxide is in the earlier phase.

An activation energy of  $\text{H}_2\text{O}$  production is estimated from the temperature dependence of  $P_{18}$ . The value obtained in the later phase is 32 kcal/mol, which is 1.5 times larger than that obtained for glow discharge cleaning by Juelich group. This large value also suggests deoxidization of chromium oxide in the later phase of the cleaning.

For wall temperatures below 200° C, decay time of  $P_{18}$  becomes shorter as the wall temperature is raised. On the other hand, the decay time above 250° C is longer than that below 200° C

although the oxygen reduction rate still increases with the temperature. This suggests that oxygen diffusion inside the wall plays a role in discharge cleaning with a high wall temperature.

The ratio  $P_{18}/P_2$  increases as the plasma density  $n_e$  and is saturated for  $n_e > 10^{11} \text{ cm}^{-3}$ . The saturation occurs above the density where dissociation loss of  $\text{H}_2\text{O}$  exceeds sticking loss. In order to interpret the saturation for large  $n_e$  value of  $10^{11} \text{ cm}^{-3}$ , sticking probability of  $\text{H}_2\text{O}$  must be as large as 0.5 at  $150^\circ \text{C}$  of the wall temperature.

#### Acknowledgement

The authors are indebted to Dr. Y. Sakamoto and his group (Inst. Chem. Phys. Res., Wakoshi) for their support to utilize  $\mu\text{CR}$  discharge cleaning. The author is grateful to Dr. K. Akaishi, Prof. A. Miyahara and Prof. J. Fujita for their stimulating discussions.

## References

- [1] H. F. Dylla, J. Nucl. Mater. 93 & 94 (1980) 61
- [2] L. Oren, R. J. Taylor, Nucl. Fusion 17 (1977) 1143
- [3] TFR Group, J. Nucl. Mater. 76&77 (1978) 587
- [4] H. F. Dylla, J. Vacuum Sci. Technol. 15 (1978) 734
- [5] Y. Gomay, R. E. Clausing, R. J. Colchin, L. C. Emerson, L. Heatherly, W. Namkung and J. E. Simpkins, J. Vac. Sci. Technol. 16 (1979) 918
- [6] H. F. Dylla, S. A. Cohen, S. M. Rossnagel, G. M. McCracken and Ph. Staib, J. Vacuum Sci. Technol. 17 (1980) 286
- [7] K. Bol, et al., in: Proc. 7th Intern. Conf. on Plasma Physics and Nuclear Fusion Research, Innsbruck, 1978, Vol. 1 (IAEA, Vienna, 1979) p. 11
- [8] N. Fujisawa, M. Nagami et al., JAERI-M 9181 (Japan Atomic Energy Research Institute, Tokai, 1980)
- [9] N. Noda, S. Tanahashi, K. Kawahata, Y. Kawasumi, Y. Ishibe, S. Ishii, K. Okazaki and H. Oyama, Y. Sakamoto and K. Yano, IPPJ-595 (1982, IPP Nagoya), to be published in J. Nucl. Mater. 111 & 112 (1982)
- [10] Y. Sakamoto, Y. Ishibe, S. Ishii, K. Okazaki, H. Oyama, K. Yano, N. Noda, K. Kawasumi, K. Kawahata and S. Tanahashi, IPPJ-539 (1981, IPP Nagoya), to be published in J. Nucl. Mater. 111 & 112 (1982)



- [11] F. Waelbroeck, J. Winter, P. Wienhold, in: Proc. 5th Symp. on Plasma Chemistry, Edinburgh, 1981, Eds. B. Waldie et al., p. 351
- [12] J. Winter, F. Waelbroeck, B. Brandt, K. J. Diets, I. Ali-Khan and P. Wienhold, J. Nucl. Mater. 93 & 94 (1980) 812
- [13] P. Staib, H. F. Dylla and S. M. Rossnagel, J. Nucl. Mater. 93 & 94 (1980) 315
- [14] J. J. Olivero, R. W. Stagat and A. E. S. Green, J. Geophys. Res. 77 (1972) 4797
- [15] E. M. Jones, CLM-R175, Culham lab. (1977)
- [16] H. Kumagai, G. Tominaga, Y. Tsuji and G. Horikoshi, "Vacuum Science and Engineering", Syokabo, Tokyo, 1970, p.130 (in Japanese)

## Figure Captions

Fig. 1 Signals of RGA during discharge cleaning

Fig. 2 Long time behavior of RGA signals for  $m = 16$  ( $\text{CH}_4$ ) and  $m = 18$  ( $\text{H}_2\text{O}$ )

Fig. 3 Time behavior of  $\text{H}_2\text{O}$  pressure for various wall temperature  $T_w$ .

Fig. 4 Partial pressure of  $\text{H}_2\text{O}$  as a function of wall temperature

closed circle : for AC discharge (present)

open circle : for glow discharge

(by Waelbroeck et al.<sup>3</sup>)

Fig. 5 Density dependence of  $P_2/P_{18}$

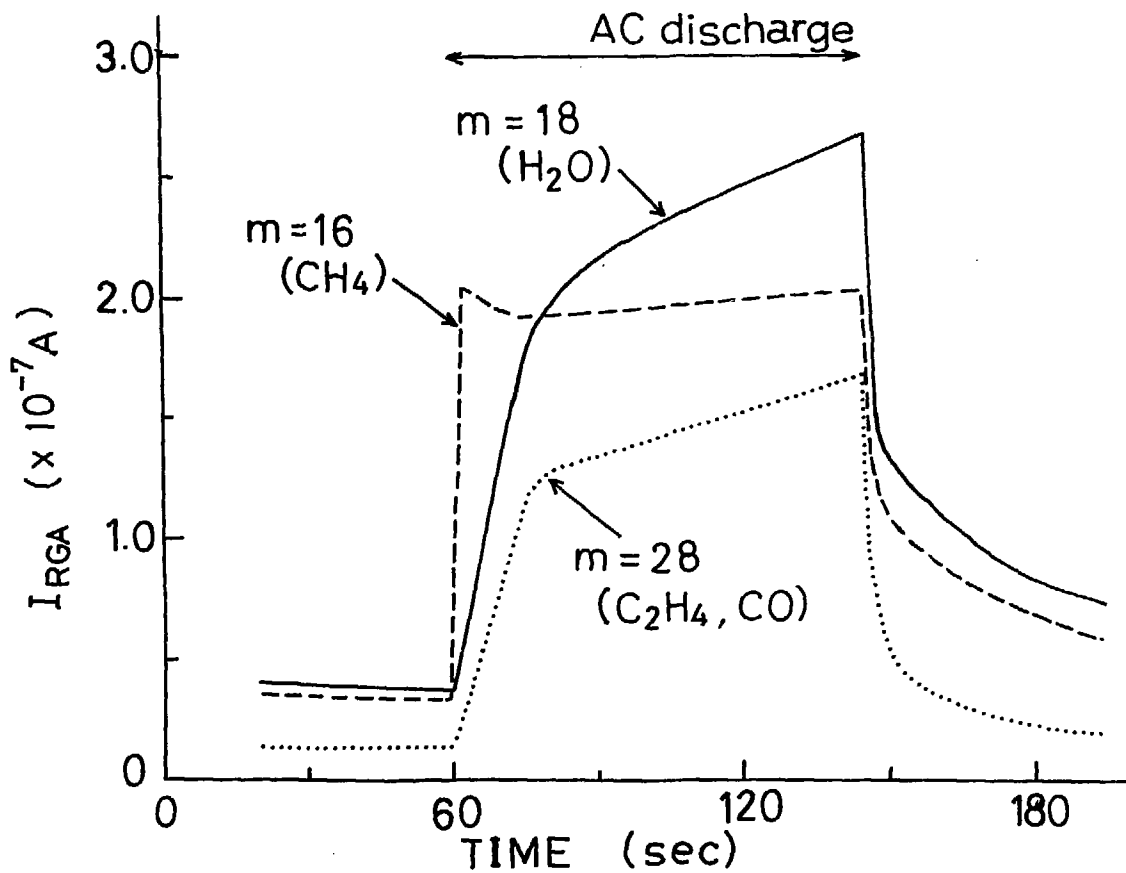
closed circle : experimental

solid line : calculated as  $P_{st} = 0.5$

dashed line : calculated as  $P_{st} = 0.1$

Fig. 6 Dissociation rate coefficients  $k_2$  for  $\text{H}_2$  and  $k_{18}$  for  $\text{H}_2\text{O}$  as a function of electron temperature

FIG. 1



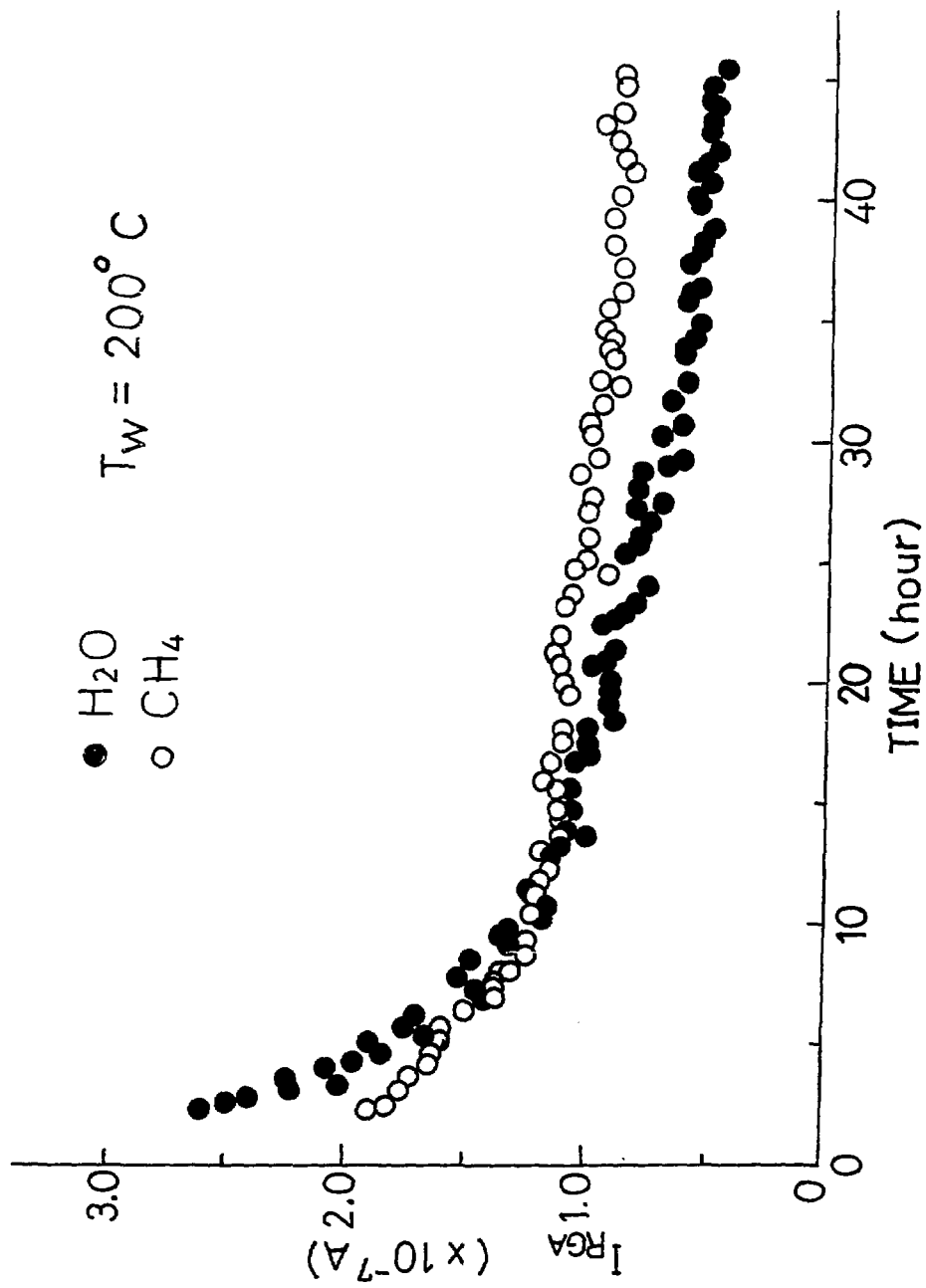


FIG. 2

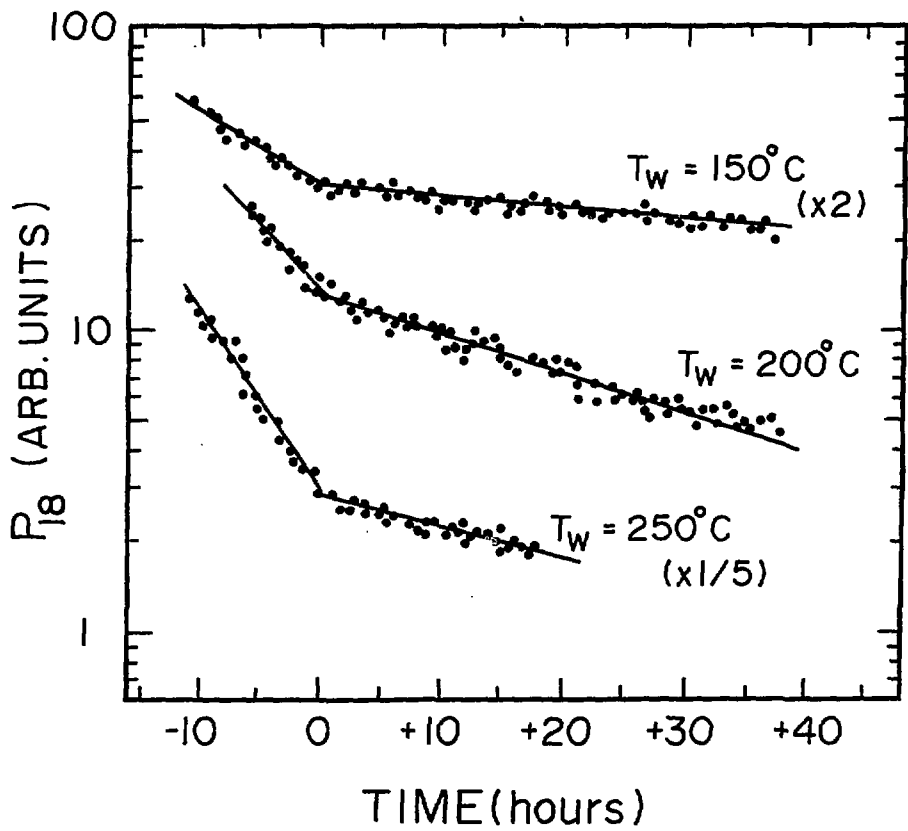


FIG. 3

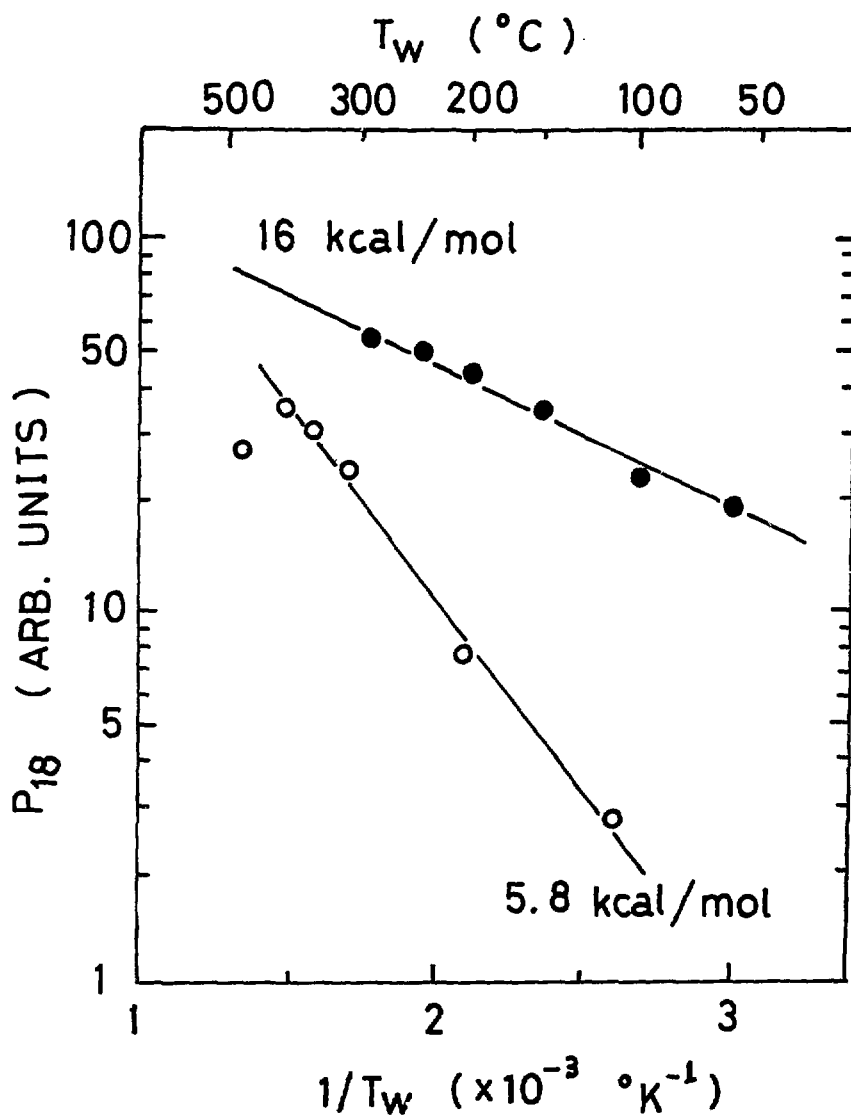


FIG. 4

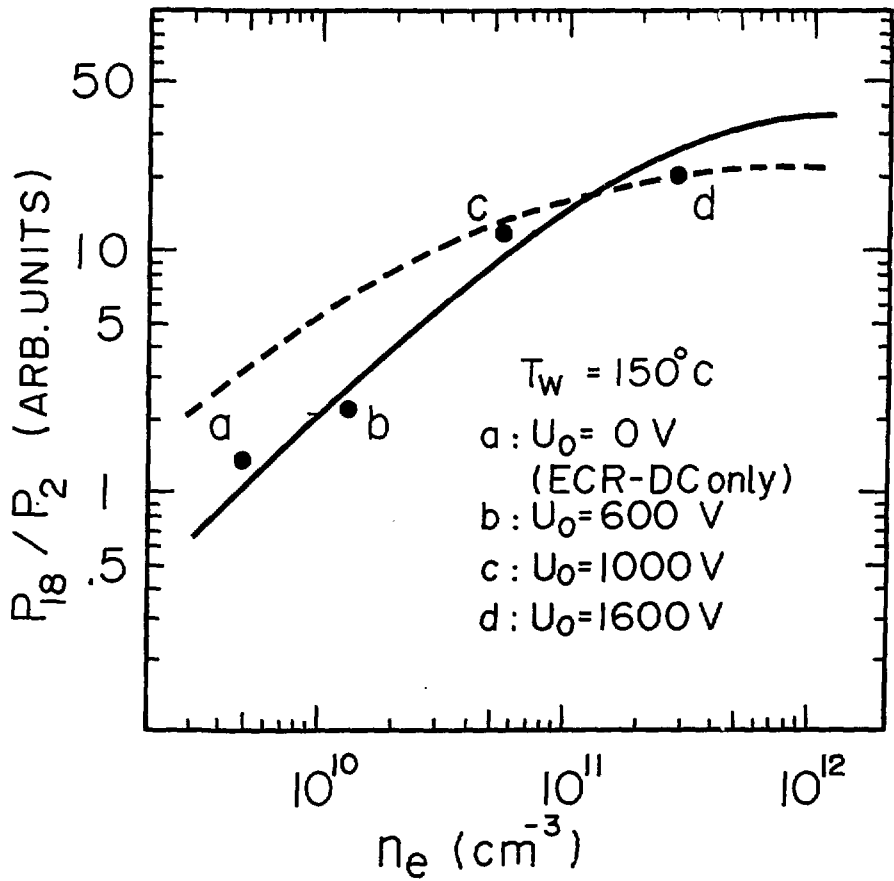


FIG. 5

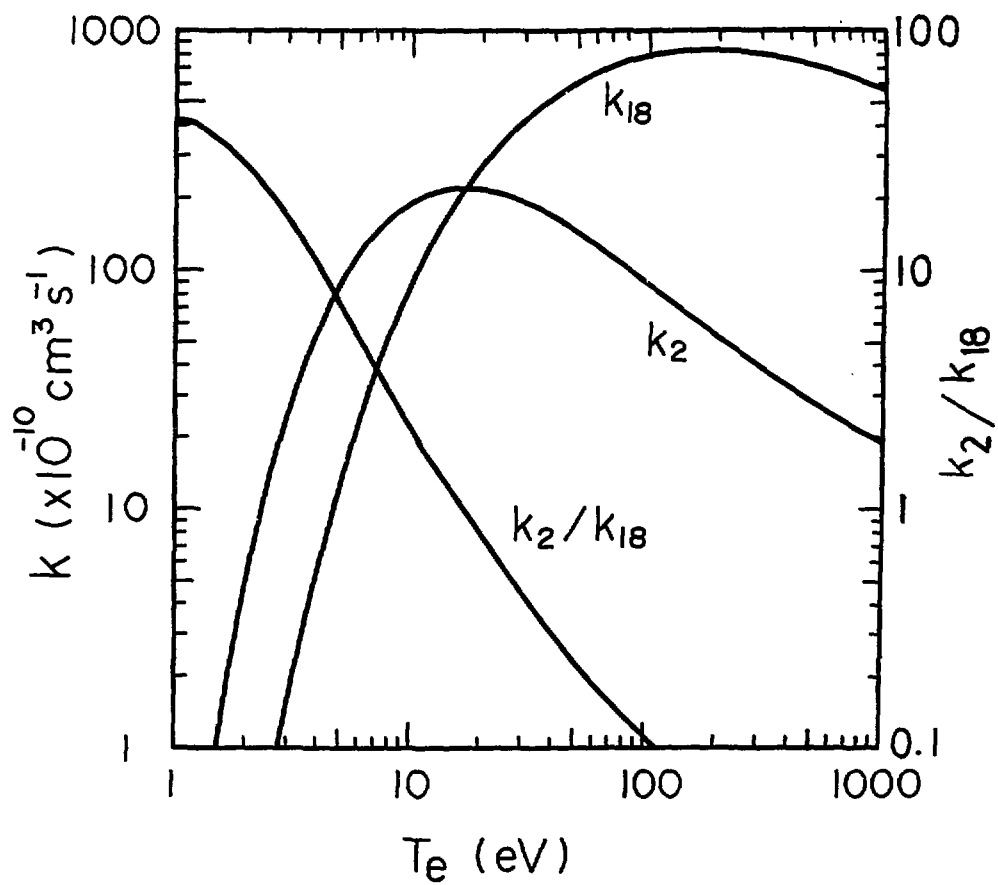


FIG. 6