Development of a planar type of modulated inductively coupled thermal plasma for uniform oxidation processing

Dissertation Abstract

平面型変調誘導熱プラズマの開発とその表面酸化処理への応用

DEVELOPMENT OF A PLANAR TYPE OF MODULATED INDUCTIVELY COUPLED THERMAL PLASMA FOR UNIFORM OXIDATION PROCESSING

Doctor of Philosophy

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Submission Date : 06/01/2017

Abstract

Thermal plasmas have their unique advantages such as high gas temperature and high densities of reactive chemical species. From these advantages, they are anticipated to be utilized for new innovations in industrial applications. Among the various types of thermal plasmas, the inductively coupled thermal plasma (ICTP) has been widely used as an effective source of heat and chemical species in various material processings. Plasma processing has also found increasingly widespread industrial applications because it can produce unique effects of commercial value that can be obtained in no other way. For a large-area materials processing, a large volume of thermal plasma, cylindrical ICTP torches of 100–300-mm-diameter have been developed to generate a plasma. However, the conventional cylindrical type of ICTP is only slightly suitable to 'large-area' materials processing because the cylindrical ICTP requires an extremely large volume of the ICTP for large-area processing when the flame of the ICTP is used. For the same purpose, a planar type ICTP torch which is rectangular vesel has been developed with ferrite core core coil or air core coil from our group. For a further large-area materials processing such as oxidation process, we have used rectangular coil which is sandwiched to the torch to generate a long-laterally thermal plasma. In this work, a novel planar type of induction thermal plasma system with current modulation has been developed using a rectangular quartz vessel instead of a conventional cylindrical tube for thermal plasma processing for large-area material surface. To expand the generated thermal plasma laterally, the rectangular coil is used around the planar torch. A planar type of modulated inductively coupled thermal plasma (ICTP) with a molecular gas feeding on the substrate has been developed for the oxidation process. In this work, the stable operation of the modulated planar ICTP with lower Ar and molecular oxygen gas was studied for high speed oxidation processing. The changes in electrical properties of Ar–O_2 thermal plasma were investigated to confirm the modulation effect in planar ICTP system at a pressure of 10 torr at an input power about 10 kW.

Chapter 1

Development of a planar type of modulated inductively coupled thermal plasma for uniform oxidation process

1.1 Experimental setup and conditions

1.1.1 A planar type of inductively coupled thermal plasma torch with a substrate holder

The actual configuration of the planar-ICTP torch in the present work is shown in figure 1.1. The planar-ICTP torch is made of a rectangular quartz vessel with 5 mm thickness and it has a rectangular inner cross-section with dimensions of 120 mm $\times$ 20 mm $\times$ 100 mm. Under the rectangular quartz vessel, the vacuum chamber made of stainless steel was installed. This vacuum chamber was connected with a vacuum pump.

In the rectangular quartz vessel, a substrate holder with a size of 100 mm $\times$ 10 mm $\times$ 2 mm is located. This substrate holder is made of Si_3N_4. We used Si_3N_4 material as a holder because of its high melting point of 2173 K. The substrate holder is placed with a distance of 10 mm from the downer base edge of the vessel as indicated in figure 1.1. On this substrate holder, we can put a substrate with a size of 80 $\times$ 8 $\times$ 0.6 mm^3 from the vacuum chamber.

A rectangular air-core coil of 5 turns per side is located to the rectangular quartz torch as shown in figure 1.1. This air-core coil is made up of rectangular copper plates. The main performance of this rectangular coil is to generate an induced magnetic field

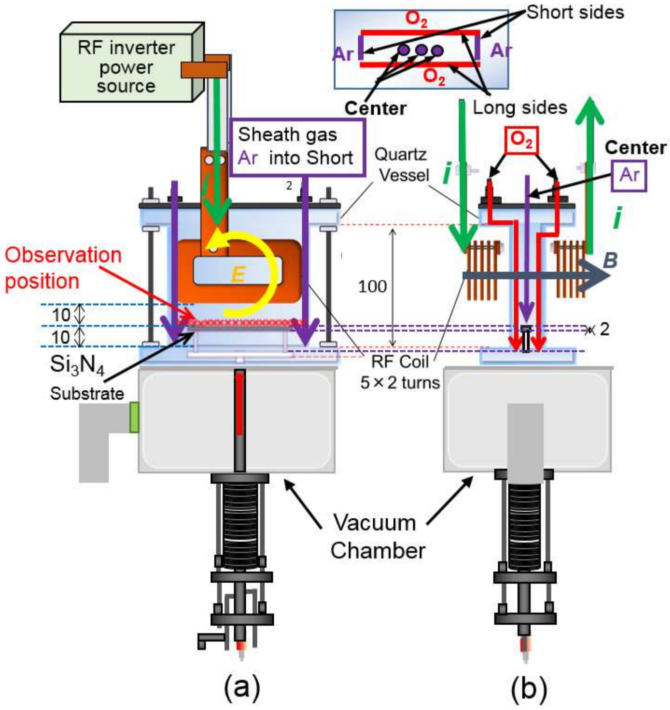


Fig. 1.1 : Experimental setup of a planar inductively coupled thermal plasma (ICTP) torch, (a) Front view and (b) Side view.

perpendicularly to the quartz vessel when the RF current is flowing through it. Since the coil is rectangular, it generates the magnetic field in a wider range than the round coil. The electric RF current was supplied to the coil is from an RF inverter power source at a rated power of 30 kW through a matching transformer and an LC series matching circuit. The fundamental frequency of the current was about 356 kHz in this work. The planar torch, including the coil, was immersed in cooling water in a water tank while the generation of thermal plasma. It is necessary to maintain the quartz vessel wall's temperature at around 300 K by circulating the cooling water outside of the vessel.

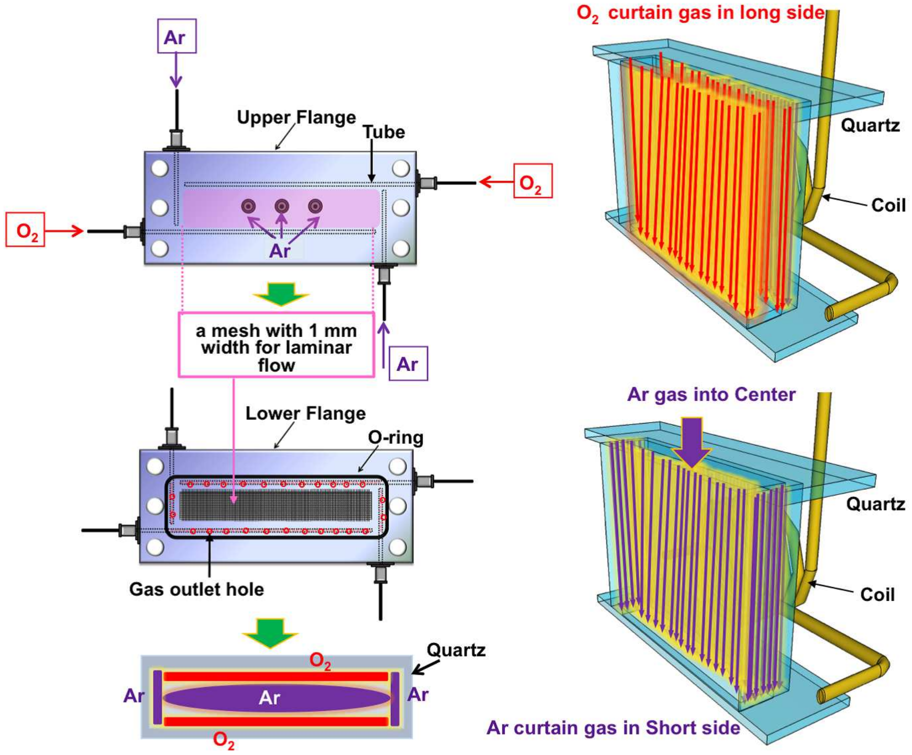


Fig. 1.2 : Gas allocation in a planar torch.

1.1.2 Gas flow pattern in the planar torch

In a planar ICTP torch, the thermal plasma should be thermally isolated from the inner wall of the torch to avoid high heat flux from the thermal plasma to the wall. Therefore, the arrangement and the shape of the upper flange for introducing the plasma gas and sheath gas are important factors to sustain the ICTP stably. The upper flange connected to the upper part of the torch is shown in figure 1.2. The working gases, Ar and O_2, are introduced from the head of a planar ICTP torch through the terminals. In this work, the gas flow patterns of Ar into the short sides and center, and O_2 into the long sides of a planar ICTP torch, can sustain the thermal plasma on the substrate with a lower Ar gas flow rates of 2 slm.

1.1.3 Experimental conditions

Table 1.1 summarizes the experimental conditions adopted for a planar Ar–O_2 ICTP for modulation and non-modulation. The pressure in the chamber was fixed at 10 Torr. The sheath gas for the long sides was O_2, and its total gas flow was set at 0.1 slm including 0.05 slm for each long side. For Ar gas of 2 slm, 1 slm Ar is injected into the short sides and 1 slm Ar into the head center of the torch. The driving frequency of the RF coil current was set to 356 kHz.

In the present work, we set mainly the conditions: with coil modulation and without modulation to study modulation effect

1.2 Results and discussion

1.2.1 Dynamic behaviour of planar Ar–O_2 ICTP with and without coil current modulation

Figure 1.3 depicts a photograph of a planar Ar–O_2 ICTP on the Si_3N_4 substrate holder at a pressure of 10 Torr at an input power of 8.5 kW without modulation.

Next, we modulated the coil current sustaining the Ar-O_2 planar ICTP at a modulation frequency of 40 Hz and 51%DF, and at a modulation frequency of 20 Hz and 51%DF. In both cases, an average input power was 8.5 kW.

The radiation intensity is shown here with colour fringes, where higher intensity is indicated in red, and lower intensity is in blue. As indicated in figure 1.4, the radiation intensity from the planar ICTP is increased from t=0 ms to 12 ms in the on-time. On the other hand, at t= 16 and 24 ms, i.e. during the off-time, the radiation intensity from the planar ICTP decays because the input power to the planar modulated ICTP is decreasing. In such a way, the coil current modulation controls the input power to the ICTP, and then may control the temperature of the ICTP. In the present work, Ar-O_2 planar ICTP may have such a significant temperature change by the modulation of the coil current.

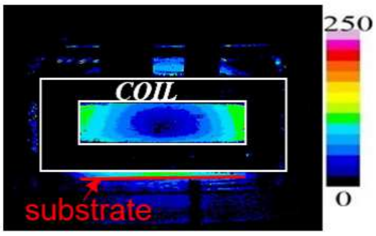


Fig. 1.3 : Visible light emission from a planar Ar-O_2 ICTP at 8.5 kW and 10 Torr without modulation.

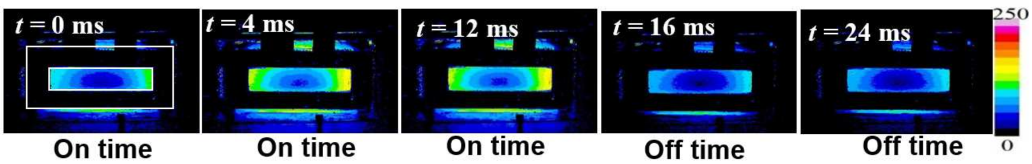


Fig. 1.4 : Visible light emission from a planar Ar-O_2 ICTP at 5.26-11.90 kW and 10 Torr with modulation (40 Hz and 51%).

1.2.2 Electrical properties of planar induction thermal plasmas in non-modulation and modulation condition

Figure 1.5 portrays (i) the modulation control signal for coil current modulation, (ii) the measured inverter current $i_{\mathrm{inv}}(t)$ and voltage $v_{\mathrm{inv}}(t)$, (iii) the calculated output power p_{inv} and $P_{\mathrm{avg}}(t)$ from the inverter power supply, and (iv) the calculated effective impedance $Z(t)$. Figures 1.5(a) and 1.5(b) respectively correspond to results for non-modulation and modulation.

1.2.3 Surface oxidation of a Si substrate irradiated by modulated and non-modulated ICTP

A planar Ar–O_2 ICTP was adopted to a Si substrate for the surface oxidation to confirm the improvement of uniform oxide layers with the modulation and non-modulation. For this measurement, the oxide layer was assumed to be SiO_2 with

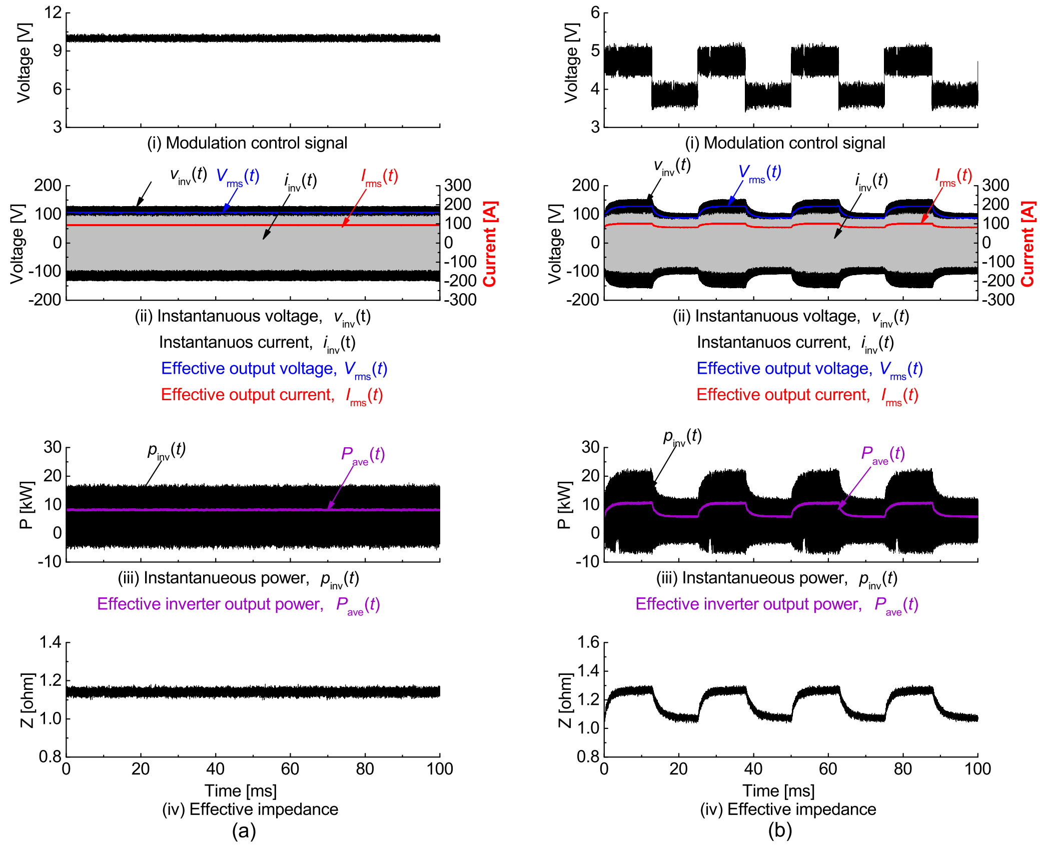
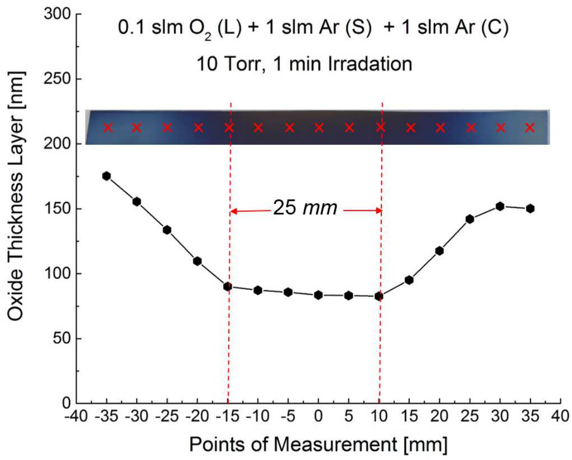
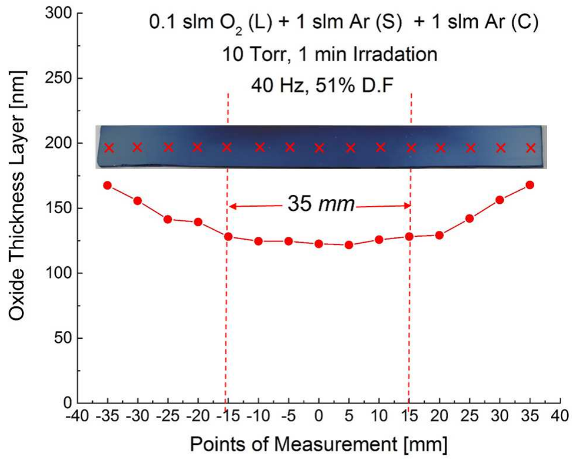


Fig. 1.5 : Time evolutions in fundamental electrical properties: (a) non-modulation and (b) modulation (40 Hz, 51% duty factor).

a refractive index of 1.46. The thickness of oxide layers produced by a planar Ar–O_2 ICTP with the non-modulation and with modulation are shown in figures 1.5(a) and (b). These figures contain the images of the irradiated Si substrates with non-modulation and modulation respectively. Almost the uniform oxide layer was found to be formed in 25 mm around the center of the substrate and the thickness was below 100 nm under non-modulation condition. On the other hand, the uniformity of oxide layer is increased in a range more than 35 mm around the center position $X=$ 0 mm. The thickness of the oxide layer is also increased to 125 nm there just only by one-minute irradiation of a modulated Ar–O_2 planar ICTP. This may be because the relatively higher density of O atoms produced during the off-time can increase the thickness of the oxide layer while a lower density is produced during the on-time.



(a)



(b)

Fig. 1.6 : The thickness of oxide layer of Si substrate under (a) non-modulation condition and (b) modulation condition with one-minute irradiation.

Moreover, the modulated ICTP has high radiation intensity at the center compared to both sides during the off-time or at the low current level. This is one of the facts that could improve the oxide layer thickness at the center of the substrate. This can be deduced from the radiation intensity distribution discussed in the previous section. Consequently, the coil current modulation with the on-time and off-time or HCL to LCL could control the time-average atomic O densities, and also the time-average temperature distribution on the substrate to improve the uniformity of surface oxidation.

1.2.4 Effect of modulation condition on oxide layer thickness of a Si substrate

Figure 1.7 shows the lateral distribution of the oxide layer thickness on the surfaces of the Si substrate for surface oxidation. The thickness of the oxide layers irradiated by the non-modulated and modulated ICTP were compared to study its effect on the uniformity of the oxidation processing. It is significantly evident that the oxide layer with non-modulation is thinner and its uniformity is lower. The uniformity of the oxide layer thickness can be improved by adopting the coil current modulation. Among the modulation conditions, the oxide layer is moderately more uniform along the substrate with a higher thickness around $x = 0$ mm under a modulation condition with 40 Hz and 51%. Decreasing the modulation frequency to 20 Hz at the same duty factor of 51% causes the higher thickness layer around the edges of the substrate and the lower thickness around the center compared to 40 Hz modulation. This means decreasing modulation frequency makes the uniformity of oxide layer thickness lower. On the other hand, increasing duty factor to 60% at the same modulation frequency of 40 Hz lessens the uniformity of the oxide layer thickness compared to 51% duty factor. This means increasing the duty factor with same frequency elevates the thickness of oxide layer around the edges and lower the thickness layer around the center. Therefore, we confirmed that the higher modulation frequency with a lower duty factor could improve the uniformity of oxide layer thickness.

1.3 Conclusions

In summary, a planar type of Ar induction thermal plasma system with O_2 gas with the coil current modulation was adopted for the rapid large-area surface oxidation processing. The required amounts of Ar and O_2 was studied for stable operation of a modulated planar ICTP were investigated. Electrical properties, such as time variations in active output power and effective electrical impedance change, were calculated to check the change in thermal plasma impedance. In addition, spectroscopic observations were carried out to investigate the controllability of the atomic O density distribution in thermal plasmas during the on-time and off-time of the modulation. This planar Ar–O_2 ICTP was used to irradiate to Si substrates for a period of one minute to study adaptability of planar ICTP for large-area rapid oxidation processing with the modulation and non-modulation. Results indicate that a planar Ar–O_2 ICTP can be adopted for the rapid large-area oxidation of Si substrates and coil current modulation could improve the uniformity in thickness of the oxide layer fabricated.

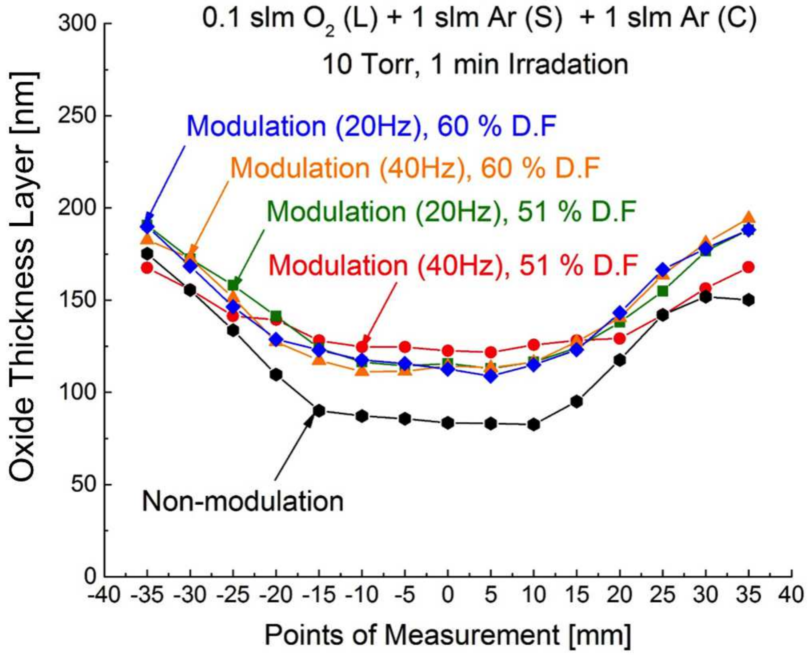


Fig. 1.7 : The thickness of oxide layers of Si substrates under non-modulation and modulation conditioins with different modulation frequencies and duty factors.

Table 1.1 : Experimental conditions

Experimental No.	001	002	003	004	005
Modulation	Yes	Yes	Yes	Yes	No
Inverter output voltage [V_{rms}]	114	117	108	111	112
Inverter output current [A_{rms}]	78	75	76	75	76
frequency [kHz]	356	356	356	356	356
Inverter effective power [kW]	5.62-11.9	4.71-11.35	4.56-11.19	4.66-11.31	8.51
Modulation control signal [V]	4.7-3.8	4.8-3.8	4.7-3.7	4.8-3.8	-
O_2 (long)[slm]	0.1	0.1	0.1	0.1	0.1
Ar (short)[slm]	1.0	1.0	1.0	1.0	1.0
Ar (Center)[slm]	1.0	1.0	1.0	1.0	1.0
Pressure [Torr]	10	10	10	10	10
Modulation frequency [Hz]	40	20	40	20	0
Duty factor(%)	51	51	60	60	100
On-time [ms]	12.75	25.5	15	30	-
Off-time [ms]	12.25	24.5	10	20	-

平成２９年　2月　3日

学位論文審査報告書（甲）

１．学位論文題目（外国語の場合は和訳を付けること。）

Development of a planar type of modulated inductively coupled thermal plasma for uniform oxidation processing

（平面型変調誘導熱プラズマの開発とその表面酸化処理への応用）

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３．審査結果の要旨（600～650 字）

平成 29 年 2 月 3 日に第 1 回学位論文審査委員会，同日に口頭発表，第 2 回審査委員会を開催し，慎重審議の結果，以下のとおり判定した。なお，口頭発表における質疑を最終試験に代えるものとした。

誘導熱プラズマは，無電極で，ガス温度が数万度の熱プラズマ空間を形成できるため，これまでもプラズマ溶射，ナノ粒子生成などの材料プロセスへの応用が活発である。しかし従来の円筒型の誘導熱プラズマ装置は，大面積材料表面処理に応用することは不向きであった。大面積表面処理に応用できる新たな誘導熱プラズマの開発が望まれていた。

本論文では，従来の円筒型でない平板型容器にコイルを適切に配置することで，角型容器内に誘導熱プラズマを安定に生成・維持できることを明らかにした。この平面型誘導熱プラズマの温度分布を分光観測から明らかにし，温度 6000 K 程度の熱プラズマが生成できていること，さらにこの温度を，熱プラズマを維持する電流を振幅変調させることで 5000-6500 K の間で制御できることも示した。この平面型誘導熱プラズマの応用例として，Si 基板の表面酸化処理を行い，分子性ガス O_2 を導入しても平面型誘導熱プラズマが安定維持できること，長さ 50mm にわたり酸化速度 100nm/min の超高速酸化処理を実現できること，コイル電流の変調により熱プラズマ・酸化膜の一様性を向上できることを明らかにした。

以上のように，本研究は新規の誘導熱プラズマの開発とその応用分野に対して大きく貢献するものであり，本論文は，博士（学術）に値すると判定した。

４．審査結果　(1) 判　　定（いずれかに○印）　○合　格　・　不合格

(2) 授与学位　博　士（学術）