	かわ じり こう た ろう
氏名	河 尻 耕 太 郎
授与学位	博士（工学）
学位授与年月日	平成17年3月25日
学位授与の根拠法規	学位規則第4条第1項
研究科，専攻の名称	東北大学大学院工学研究科（博士課程）機械知能工学専攻
学位論文題目	Optimization of a DC-RF Hybrid Plasma Flow System for Particle Processing （微粒子プロセス用DC-RFハイブリッドプラズマ流動システムの最適化）
指導教官	東北大学教授　西山 秀哉
論文審査委員	主査　東北大学教授　西山 秀哉　東北大学教授　圓山 重直 東北大学教授　南部 健一　東北大学教授　寒川 誠二

論文内容要旨

Introduction

A DC-RF hybrid plasma flow, which is produced by combining a radio frequency inductively coupled plasma (RF-ICP) flow with a direct current (DC) plasma jet, is expected as a next generation plasma flow to overcome some typical disadvantages of an RF-ICP flow, such as difficult ignition, difficult injection of reactants to the centerline region from the torch head owing to its strong recirculation and instability against its thermofluid characteristics. Then, it has been applied intensively to material processing by some researchers for decades because of its prominent advantages. On the other hand, the fundamental characteristics are not clarified enough even for a conventional RF-ICP flow because a direct measurement is restricted severely because of its extreme conditions such as high temperature more than 10000 K and strong radiation. Then, the further experimental investigation of the fundamental characteristics inside a DC-RF hybrid plasma flow is required to utilize its functionalities effectively. In addition, a novel method to optimize its complex operating conditions is also required because there are many controllable input parameters and some incompatible output parameters in a DC-RF hybrid plasma flow system. In the present study, detailed experimental investigations of thermofluid characteristics and interaction between a DC-RF hybrid plasma flow and in-flight particle behavior are conducted. After that, based on the previously obtained results, a material processing is conducted actually. In addition, a novel original method of multi-input and multi-objective optimization, in which statistical analysis (Taguchi method) are combined with genetic algorism, is applied to optimize the whole DC-RF hybrid plasma flow system to improve the process efficiency. The main objective of this Ph. D thesis is not only the comprehensive experimental investigation of a DC-RF hybrid plasma flow system from the fundamental phenomena to the real application, but also its optimization for in-flight particle processing from a view point of electromagnetic thermofluid dynamics.

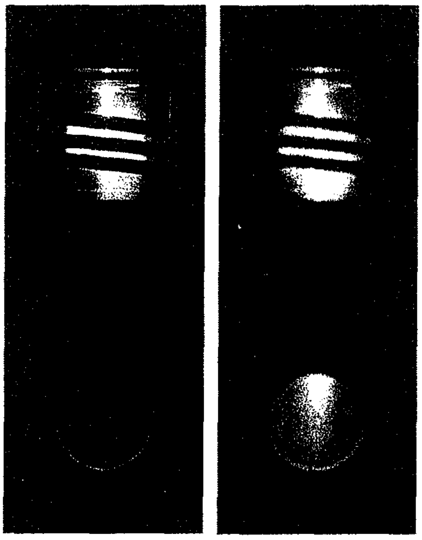
Advanced Control of Thermofluid Characteristics in a DC-RF Hybrid Plasma Flow System

Experimental investigation of thermofluid characteristics in a DC-RF hybrid plasma flow system and its advanced control are conducted to enhance its functionalities.

1. As shown in Figure 1, a DC-RF hybrid plasma flow is elongated further downstream, its temperature at the centerline region increases, the plasma flow becomes stable and a large amount of reactive gas can be injected without disturbing it under the optimized operating condition.

2. Two-factorial analysis is carried out effectively to analyze the effects of the input parameters on a DC-RF hybrid plasma flow characteristics precisely. Compared with a conventional RF-ICP flow, a DC-RF hybrid plasma flow has a prominent advantage in controllability.

3. Multi-factorial analysis using Taguchi method is applied to clarify the effects of the many input parameters on a DC-RF hybrid plasma flow and to show the correlation between the input parameters and the output parameters by the concrete formula as shown in Figure 2. The controllability of a DC-RF hybrid plasma flow is also confirmed by the multi-factorial analysis.



(a) (b)

Figure 1 Photographs of a DC-RF hybrid plasma flow for (a) conventional condition and (b) optimized condition

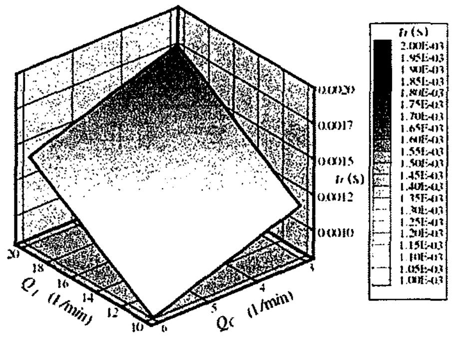


Figure 2 Particle residence time

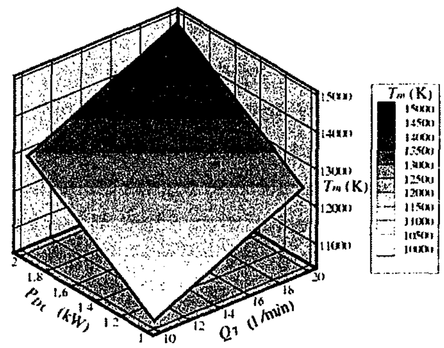


Figure 3 Plasma temperature

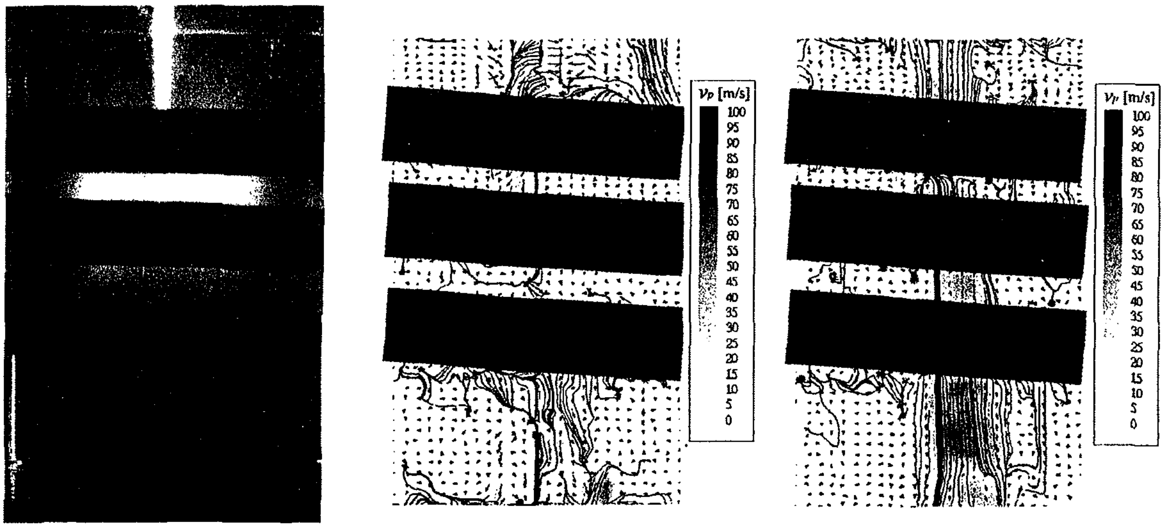
Optimization of In-flight Particle Characteristics in a DC-RF Hybrid Plasma Flow System

Experimental investigation to clarify the correlation between the operating condition and particles behavior in a DC-RF hybrid plasma flow. In addition to the fundamental investigation, the optimization of in-flight particle characteristics to increase particle temperature and particle velocity is conducted in a DC-RF hybrid plasma flow system by using multi-input and multi-objective optimization.

1. The measurement of particle velocity and particle trajectory in a DC-RF hybrid plasma flow is succeeded by using advanced particle image velocimetry (PIV) system. The strong back flow due to Rorenz force is suppressed and the particle velocity is increased by decreasing operating pressure as shown in Figure 4.

2. Particle heating is enhanced by increasing operating pressure because particle residence time decreases and Knudsen effect becomes weak as shown in Figure 5. Particle temperature increases and particle velocity decreases with decreasing the swirl gas ratio because the recirculation is suppressed by decreasing the swirl gas ratio. While particle heating is also enhanced by using the advanced particle injection system, the plasma flow becomes instable.

3. The operating condition is optimized by multi-input and multi-objective optimization, in which Taguchi method is combined with genetic algorism, to increase particle velocity and particle temperature in Figure 6. The negative correlation between particle velocity and particle temperature is shown on the Pareto resolutions obtained by the novel optimization method.



(a) (b) (c)

Figure 4 Visualizations in the RF coil region for a DC-RF hybrid plasma flow; Q_{Sh} =17 l/min, Q_C = 3 l/min and R_{Sw} = 0. (a) PIV photograph at P=93 kPa, (b) particle velocity and particle trajectory at P=93 kPa, (c) particle velocity and particle trajectory at P=13 kPa.

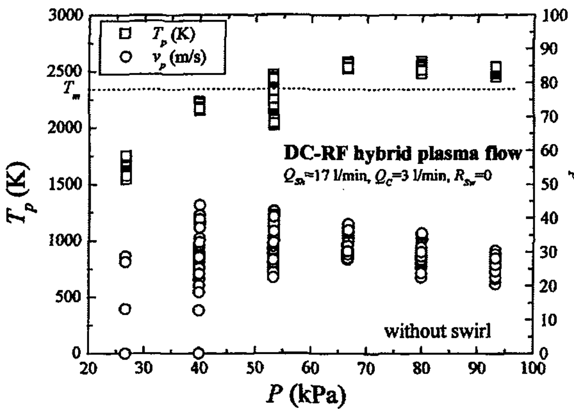


Figure 5 Particle temperature and particle velocity for various operating pressures

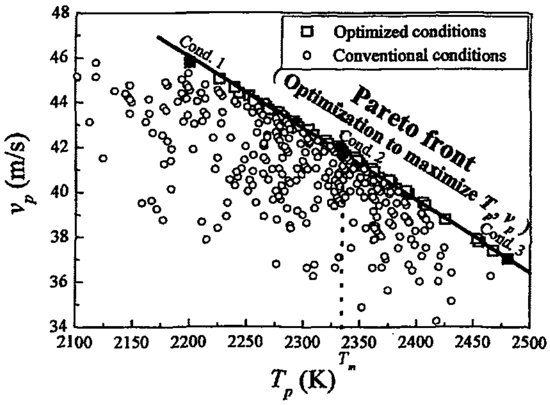


Figure 6 Pareto-optimal resolutions to maximize particle temperature and particle velocity

Optimization of Particle Processing in a DC-RF Hybrid Plasma Flow System

The main objective of this chapter is to clarify the fundamental processes such as evaporation, nucleation, agglomeration and sintering during in-flight particle treatment using a DC-RF hybrid plasma flow by focusing on the particle size. Here, the initial precursor of rutile TiO_2 particle (2 micrometer) is injected from the torch head and much finer rutile TiO_2 paritcle is obtained from the downstream collector after the treatment. Based on the previously obtained results, the following discussion is conducted in a view point of thermofluid dynamics. The operating condition is optimized by multi-input and multi-objective optimization to decrease the mean size of ultra fine particles, increase the conversion ratio to ultra fine particles and stabilize the plasma flow during the processing, additionally.

1. Heterogeneous nucleation on the surface of the seeds, which pass through the plasma flow without any treatment, is dominant at reduced pressure because of a small amount of evaporation of the initial precursor as shown in Figure 7. The ultra fine particles less than 0.3 μm due to homogeneous nucleation, of which illustration is shown in Figure 8, are obtained in agglomeration for more than 33 kPa in Figure 9.

2. The mean size of ultra fine particles increases with increasing operating pressure and decreasing swirl gas ration because particle residence time in the condensation region becomes small as shown in Figure 9. Quench gas effect of producing ultra fine particles becomes prominent more than 50 l/min.

3. Two kinds of reduction of rutile TiO_2 particles are confirmed. The former slight reduction comes from Ar^+ sputtering at the upstream region. The latter large reduction comes from the excess heating during long residence time.

4. The operating condition is optimized by multi-input and multi-objective optimization to decrease the mean size of ultra fine particles, increase the conversion ratio to ultra fine particles and stabilize the plasma flow during the processing as shown in Figure 10. Under the optimized operating condition, the mean size of ultra fine particles is less than 130 nm, the conversion ratio is nearly 90 % and the plasma flow is very stable during the processing.

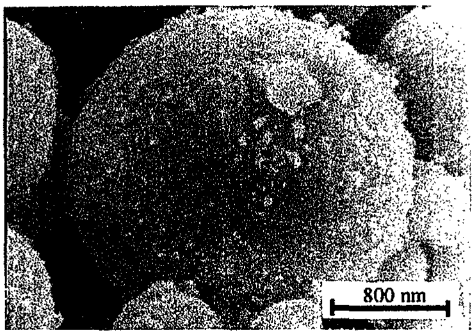


Figure 7 SEM image of heterogeneous nucleation at reduced pressure

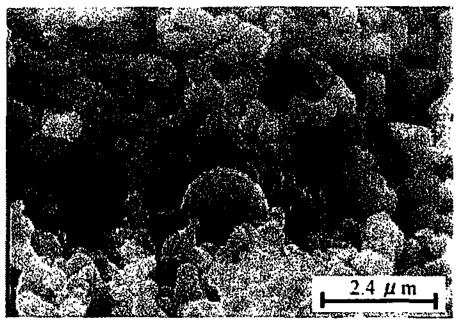


Figure 8 SEM image of homogeneous nucleation

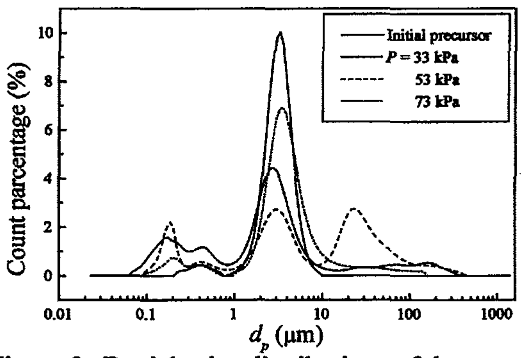


Figure 9 Particle size distributions of the initial precursor and of the products for various operating pressures

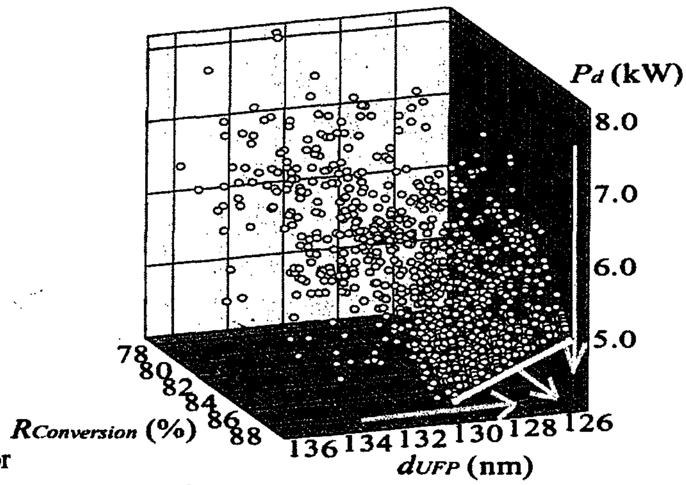


Figure 10 Pareto-optimal resolutions

Concluding Remarks

The characteristics of electromagnetic thermofluid dynamics, in-flight particle behavior and particle processing are clarified experimentally in a DC-RF hybrid plasma flow system. The efficiency of thermofluid control of a DC-RF hybrid plasma processing is confirmed in this study. Based on the detailed experimental investigation, the whole system is optimized by the novel optimization method. Finally, the further possibility of the flow control approach in a DC-RF hybrid plasma flow system for a real material processing is shown.

論文審査結果の要旨

プラズマ流体は高エネルギー密度，電磁場制御性，化学的高活性，導電性，変物性等を有する多機能性流体であり，現在材料プロセスや環境浄化プロセスに積極的に応用されている．更なる産業界での応用拡大をはかるため，熱流動特性の解明と，最適制御による高機能化が求められているが，プラズマ流は高温，高発光等の極限流体でもあり，実験計測において大きな制約があり，未だ十分に研究がなされていない．本論文では，低電力型で高機能次世代熱プラズマ流である DC-RF ハイブリッドプラズマ流を用いた微粒子プロセスに関して，内部の複雑な微粒子熱流動現象を実験的に解明し，その考察に基づいて，実際の微粒子プロセスにおけるプラズマ流動システム全体の最適化を行った．また，最新の統計解析手法である田口法と，最新の最適化手法である遺伝的アルゴリズムを組み合わせ，多入力多目的最適化手法を独自に構築し，微粒子プロセスの最適制御に適用した．

第１章は緒論である．

第２章では，ガスの注入条件により内部熱流動場を制御し，プラズマ流の発生，安定性，温度，輸送特性，反応性ガスへの発生限界等の様々な出力に関して，DC-RF ハイブリッドプラズマ流動システムの高機能化を行った．また，統計的手法である，２変数解析，多変数解析(田口法)を前述した出力向上に応用した．本章は，DC-RF ハイブリッドプラズマ流動内部の複雑現象の解明という観点から極めて有用な研究である．

第３章では，DC-RF ハイブリッドプラズマ流と注入した内部飛行微粒子との相関を考慮した，熱流動場制御による粒子特性の最適制御を行った．本章では，ガスの注入条件に加えて，新たに作動圧力を入力として採用し，プラズマ流内部の粒子軌跡，粒子速度，粒子加熱に対する作動条件の影響について詳細な考察を行った．また，田口法と遺伝的アルゴリズムを組み合わせた，多入力多目的最適化手法を完成させ，プロセス領域の粒子温度，粒子速度の２出力について最適化した．最適条件下では，粒子温度，粒子速度は負の相関関係にあることを明らかにした．本章において，後述される実際の粒子プロセスを考察する上で，非常に有用な基礎的成果を得た．

第４章では，DC-RF ハイブリッドプラズマ流動システムを用いたルチル TiO_2 粒子の微粒化プロセスの最適化を行った．また，得られた粒子の結晶構造，形態についても詳細な考察を行った．本章の考察は，２章，３章で得られた結果に基づいて展開され，DC-RF ハイブリッドプラズマ流中の熱流動場制御による微粒化プロセス制御の有用性が確認された．また，３章において完成された多入力多目的最適化手法により，生成された超粒子の平均粒径，超微粒子への変換率，プラズマ流安定性の３つの出力についてプロセス全体を最適化した．最終的に，２μm のルチル TiO_2 粒子から，安定したプラズマ状態下で，0.13 μm 以下の平均粒径を持つ微粒子を 86 %以上の変換率で生成することに成功した．

第５章は結論である．

以上を要するに，複雑干渉系混相流である DC-RF ハイブリッドプラズマ流を用いた粒子プロセスに関して，プラズマ流の内部熱流動場，プラズマ流中の微粒子の挙動，実際の粒子プロセスとテーマを一貫させて実験的基礎的研究を組み上げ，最終的に最新の最適化手法を用いてプラズマ流動システム全体を最適化した．

本論文は，機械工学およびプラズマ工学の発展に寄与するところが大きく，よって，本論文は博士(工学)の学位論文として合格と認める．