称号及び氏名	博士（工学）　Tran Trung Nguyen
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論　　文　　名	「Optimization of an Atmospheric-Pressure Plasma Source for Biomedical and Environment Applications（バイオ及び環境応用のための大気圧プラズマ源の最適化）」
論文審査委員	主査　　松浦　寛人
	副査　　川又　修一
	副査　　梅澤　憲司

論文要旨

Non-thermal plasmas (NTPs) have exhibited great potential for various fields, such as agriculture, environmental, and biomedical due to their operational flexibility. NTPs sources can produce a chemically rich environment at close to room temperature both at reduced and ambient pressures. This unique condition enables the delivery of highly reactive plasma species in a non-destructive and beneficial way to even extremely heat-sensitive surfaces. Electric field, as well as reactive oxygen and nitrogen species produced by NTPs, may inactivate bacteria,

and stimulate skin regeneration (dermatology), tumor reduction (oncology), seed germination (agriculture), and wastewater purification (environment). These new fields of research, based on plasma-liquid chemistry are very promising and developing quickly. However, due to the very high level of complexity, understanding is still poor concerning mechanisms involved in biological processes, in particular, the respective role of reactive species and electric fields. To help elucidate these effects, advanced approaches have been brought to bear on many issues. In addition, with the aim of enhancing efficient treatment and reducing operation costs under similar operating conditions, it is important to develop control methods that allow for the minimization of the effects of environmental conditions while at the same time optimizing the treatment by adjusting the electrode configuration or using a variety of gas mixtures, etc. The main objective of this thesis is based on both fundamental and applied research in non-thermal atmospheric pressure plasma interacting with water for biological and environment applications and aims at a better understanding of the physical and chemical processes of plasma-liquid interaction.

The thesis has been divided into six chapters.

Chapter 1 describes namely basic knowledge about physical plasmas and introduces the different types of NTPs. A special focus addresses the atmospheric-pressure plasma jets (APPJs), as the main core of our work, are discussed. Finally, the emerging fields of APPJs applications for environmental and biomedical.

Chapter 2 presents the fundamentals of plasma-liquid interaction as well since the interaction between plasmas and the biological targets usually involves liquids as an interface. A special focus addresses the transport process of reactive species from gas to liquid phase and several measurement methods to evaluate density and distribution of reactive species.

In Chapter 3, the main effect of insulating oil was investigated on ring power electrode and needle power electrode configurations. To reduce operation costs, our group proposed using a cheaper working gas—argon (Ar), as a replacement for helium (He). When using available low-cost Ar at atmospheric pressure, the Ar plasma discharge also needs to higher breakdown voltage, leading to a higher gas temperature. Therefore, the Ar plasma jet is known to be difficult to sustain, and unstable. The combination of a needle power electrode configuration and Ar gas was applied as an alternative solution. However, the difficulty in sustaining a glow discharge under a high supplied voltage during a long irradiation time leads to a new set of challenges. A high temperature at the ground electrode leads to thermal damage to the quartz tube. Recently, studies have illustrated that two-ring electrodes immersed in an electrically insulating oil bath could prevent arc formation. Based on these ideas, our group confirmed that the ring power electrodes plasma source could operate steadily for a long time due to the electrical field modifying the function of the insulating oil. This chapter shows positive results as insulating oil prevents arc formation, while it improved the supplied power and plasma jet length and increased radical production. Furthermore, the heat balance property of the insulating oil was observed with the needle power electrode configuration. Using the needle power electrode configuration, the heat damage could be prevented by covering the ground electrode with insulating oil. In this study, the heat balance property of the insulating oil was observed, and the operating parameters of the needle power electrode plasma source are investigated using Ar gas.

Chapter 4 proposes Polyvinyl Alcohol-Potassium Iodine (PVA–KI) as a new chemical probe in the form of gel, to visualize the distribution of reactive species. Our group has reported the development and biomedical application of PVA-KI liquid-type in previous research as a novel chemical probe to study free radicals. In particular, the liquid-type PVA–KI was used to observe the liquid flow and obtain the relative reactive species concentration distribution. Recently, reactive species' transport and distribution phenomena into biological tissue following non-thermal plasma treatment have received much attention. Therefore, the gel-type PVA-KI is developed to give information regarding the distribution of reactive species by coloration on the gel surface. The effects of the gas-phase surrounding plasma jet on the distribution and diffusion of reactive species have been investigated using plastic shielding put on the PVA-KI gel surface. The concentration of reactive species produced by Ar plasma sources distributed on the surface of PVA-KI gel in the different nozzle to target distances was measured from visualized color distributions by a color meter device.

In Chapter 5, the aim of this chapter is to test the efficacy of the Ar-ethanol plasma jet in transporting the reactive species to the target surface. Our group investigated the effect of ethanol addition on the discharge characteristics of Ar plasma jet in previous research. However, Ar-ethanol plasma jets' reactive species distribution has not been studied enough. Based on previous research combined with a new chemical probe, Ar-ethanol plasma jets' reactive species distribution can be visualized. To estimate the total amount of reactive species generation, we employ a PVA-KI liquid type. The absorbance of the PVA-KI solution near 490 nm can be measured by a UV-Vis spectrometer. On the other hand, the gel-type of PVA-KI can easily detect reactive species distribution on the gel surface. Then, the RGB value is recorded by using a color meter device to determine color distribution on the surface of PVA-KI gel. The amount of reactive species distribution is strongly dependent on the amount of ethanol addition.

In Chapter 6, the general conclusion of this study summarizes the main outcomes and points out the future perspective.

審査結果の要旨

本論文は、最近になってプラズマ応用の対象となってきた、生体や環境物質の処理に今日する事の可能な、大気圧下で運用可能な非平衡プラズマ源の改良につながる研究をすすめたものである。プラズマ生成のコストは主に動作ガスと放電電力であり、特に環境応用については大規模化の可能なシステムが好ましい。他方、プラズマが誘起する反応は、プラズマ源の置かれた環境で、あるいはプラズマ照射ターゲットとの相互作用で生成される化学活性種の種類と量で支配される。本研究では、誘電体バリア放電で生成されるプラズマジェットをベースとして、プラズマ源の最適化につながる研究を行った。特に、注目すべき成果は以下のとおりである。

(1)プラズマ源に低周波高電圧を印加するパワー電極をリング電極から針電極に変更することにより、安価なアルゴンガスを使ったプラズマ生成が可能になっていた。しかしながら、誘電体としてアーク放電の発生を防止するガラスチューブへの熱ダメージの低減が新たな課題となっていた。本研究では、韓国の研究グループがリング電極の装置に適用を提案していた絶縁油での電極の被覆が、針電極の装置にも適用可能であり、その安定化のメカニズムが、電磁気学的な効果ではなく、伝熱的な冷却効果によるものであることを明らかにすることができた。

(2)ポリビニールアルコールーヨウ化カリウム（PVA-KI）混合物は新しい放射線化学線量計や紫外線の照度計として注目されている。同様に、プラズマ照射された液中に生成された活性酸素種の定量化に利用することも行われている。本研究では、ゲル状の PVA-KI サンプルを利用し、プラズマ源内部で生成され、ガス流で輸送される活性酸素種を可視化する可能性が示されている。予備的な試験の結果として、名古屋大学の狭ギャップガス流路を備えた大型プラズマ源では、照射ターゲットを近づけすぎるとプラズマ処理の効果が低減することが予想されている。

(3)放電ガスに微量のアルコールを添付すると、ペニング効果で放電維持電圧が低下することが報告されているが、化学活性種の生成へのアルコール添付の影響はまだ明確にされていない。本研究では、PVA-KI を化学プローブとして利用し、アルコールの効果を明確にすることを試みた。プラズマ電子分布の最適化とラジカル生成の最適化が異なる可能性を初めて示すことができた。

以上の諸成果は、大気圧放電プラズマ源の内部及び照射環境で起こっている物理化学過程についての重要な知見を与える。本研究はこのように特定のプラズマ装置の改良のみならず、プラズマ化学やプラズマ生体反応の上からも有益な情報を提供したものであり、関連分野の学術的・産業的な発展に貢献するところ大である。また、申請者が自立して研究活動を行うのに必要な能力と学識を有することを証したものである。

学位論文審査委員会は、本論文の審査および最終試験の結果から、博士（工学）の学位を授与することを適当と認める。