Book of Abstracts

Effects of electrode surface morphology on microgap breakdown and microplasma properties

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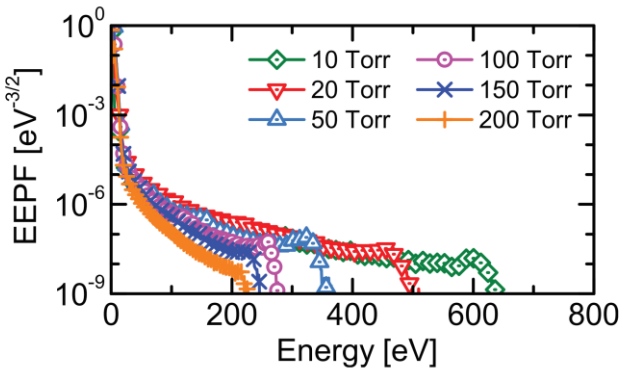
Microplasmas have received growing attention during the past decades due to their potential for a wide range of applications, such as photonic crystals, excimer sources, thin film synthesis [1,2]. This report summarized previous studies on the effects of electrode surface morphology, including surface protrusion [3], electrode roughness [4], and array microstructures [5], on the breakdown characteristics in microgap discharges. The local field enhancement and the coupled electron emission mechanisms are discussed focusing on their impacts on breakdown voltage, such as the flattened Paschen's curve. Also, by using two-dimensional particle-in-cell simulations, the microplasma properties scaling with gas pressure are studies in microgap with cathode array microstructures and the corresponding electron kinetics characteristics are evaluated. The formation conditions of the microhollow cathode discharges and the high-energy tail in the electron energy probability functions are confirmed, which is due to ballistic secondary electrons, as shown in Fig. 1 [6]. The results provide deeper insights on the fundamental microplasma physics and are helpful for developing more comprehensive optimization strategies for microplasma devices towards practical applications.



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[3] Y. Fu, P. Zhang, and J. P. Verboncoeur, Appl. Phys. Lett. **113**, 054102 (2018).
[4] Y. Fu, J. Krek, P. Zhang, and J. P. Verboncoeur, IEEE Trans. Plasma Sci. **47**, 2011 (2019).
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Computational studies of microplasma devices as variable capacitor

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Plasma based capacitors have presented themselves as possible alternatives to traditional electronic components owing to their attractive features like high capacitance ratio, good tuning rate, and high power and temperature tolerance, to name a few. The unique ability of plasma to change the reactance from capacitive to inductive nature could be applicable in the development of reconfigurable electronic components. Reconfigurability is an essential feature for the future communication systems as they must deal with congested electromagnetic spectrum. In this talk, we present the computational results of impedance characteristics of a type of microplasma capacitor in which same sets of electrodes are employed to deliver two different types of power: (i) plasma excitation power and (ii) probing signal power. Comparison between computational results and the results obtained using zero-dimensional analytical modelling show that the assumptions made on the 0D analytical modelling makes it less accurate especially for the plasma excited at lower frequencies. Impedance characteristics of two different types of plasma reactors are studied: (i) 1 cm diameter electrode operated at 1-3 Torr pressure and (ii) gas discharge tube (GDT) with 4.43 mm diameter electrode operated at 57 Torr pressure. Simulations were run in both 1D and 2D and the result comparison showed that the 1D simulations are more suited for reactors with larger diameter whereas, for the reactors with smaller diameter, the spatial variation of plasma properties made 1D results less accurate.

Spatio-temporal density of Ar($1s_5$) in a co-axial atmospheric-pressure micro-plasma jet

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In this work, we use a co-axial reactor (see Figure a) to produce an atmospheric-pressure micro-plasma jet (APMPJ). Argon is injected in the inner tube as the plasma gas (1 slm), while different N_2-O_2 gas mixtures are injected in the outer tube as shielding gas (0.1 to 3 slm and 0 to 100% of O_2). A squared voltage pulse is applied (see Figure b), allowing for a decoupling between the effects of the rising and the falling edges of the high-frequency (20 kHz) high-voltage pulses (4 kV). In the inner tube of the reactor, surface DBDs are produced that propagate as guided streamers along the argon jet outside the reactor. Spatio-temporal profiles of the Ar($1s_5$) metastable are measured using tunable diode laser absorption spectroscopy (TDLAS). APMPJs are complex systems in which flow and plasma dynamics are coupled [1]. One important interaction is the admixture of the argon jet into air. Ar($1s_5$) spatio-temporal profiles might help us understand more about this process and its effect on the plasma dynamics and characteristics. By using micro-second voltage pulses, we were able to inspect the Ar($1s_5$) spatial profiles related to the rising edge, showing characteristic features of the passage of a positive streamer, as well as the axially shorter spatial distributions and lower peak densities of Ar($1s_5$) related to the falling edge. Different shielding gas mixtures change the filamentary and diffuse regions of the plasma. Even small O_2 fractions in the shielding gas (<3%) allow for more reproducible discharges, affecting also the peak densities of Ar($1s_5$). Spatial profiles of the effective lifetime of Ar($1s_5$) are compared between different shielding gas mixtures.

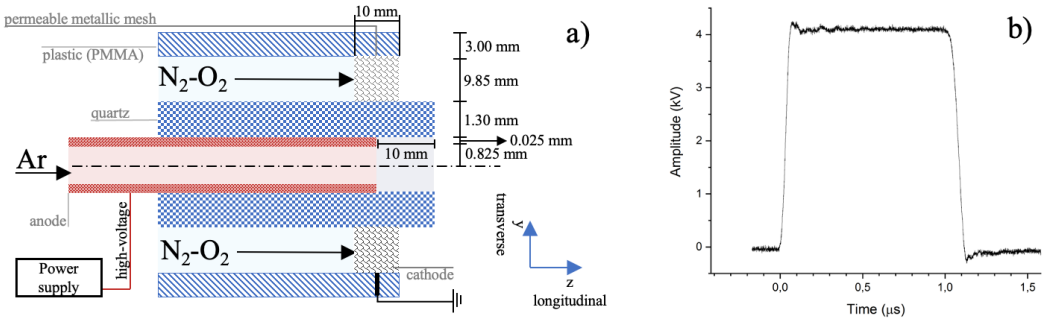

Figure: a) Cross-sectional scheme of the co-axial APMPJ reactor, b) Applied high-voltage pulse

[1] Thibault Darny *et al.* 2021 Plasma Sources Sci. Technol. **30** 105021

This work was partially supported by the Portuguese FCT UIDB/50010/2020 and UIDP/50010/2020, and grant PD/BD/142972/2018 (PD-F APPLAuSE).

On the efficiency of plasma aided nitrogen fixation

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Nitrogen is common in earths atmosphere but relatively rare in the biosphere. This is because molecular nitrogen is strongly bound and as a result almost chemically inert. In nature, unusual and energetic events such as lightning strikes are the means of transforming atmospheric nitrogen into chemically active forms such as oxides. Consequently, nitrogen is rather scarce in the biosphere, and the availability of nitrogen is often a limiting factor in processes such as plant growth. For this reason, agricultural productivity was transformed in the twentieth century by the development of artificial activation of nitrogen through a process called fixation, which makes nitrogen abundantly available. The principal method in modern use is the Haber-Bosch process, which consumes large amounts of fossil fuels in producing nitrogen fertilisers, typically in the form of nitrates. The carbon dioxide emissions associated with this process are increasingly unacceptable on account of their influence on anthropogenically induced climate change, but the implications for global food production of abandoning nitrogen fixation are equally unpalatable. Therefore, alternative methods of nitrogen fixation using sustainable energy sources are a matter of intense interest. A plausible possibility is a plasma based method using electricity from wind or solar or some other carbon free energy source. The efficiency of such a process is very important. This talk will discuss in more detail the background, and present work on modelling the relevant plasma chemistry, with particular reference to the uncertainty associated with imprecise knowledge of the relevant rate constants. On the basis of these results, a plasma process could plausibly approach the energy efficiency of the Haber-Bosch process.

Characterization and Design of Flexible Surface Dielectric Barrier Discharge Electrodes

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Two common ways of generating cold atmospheric pressure plasma include volume dielectric barrier discharges and surface dielectric barrier discharges. Surface dielectric barrier discharges (SDBDs) have been gaining interest for various applications including: microbe deactivation on the surface of produce, plasma medicine, and flow control. Surface DBD's offer some advantages to volume DBD's. SDBD's can be operated at lower voltages and they do not require the treatment target to be a part of the electrical circuit which can be beneficial in some applications. Electrical characterization of SDBD's has proved more difficult than volume DBD's due to their variable capacitance as the plasma expands onto the surface of the dielectric. In this work SDBDs generated by novel low cost electrodes [1] have been electrically characterized, for different geometries, using a simple electric circuit model. Due to the ease of manufacturing, multiple different geometries can be tested. Plasma power, total power, burning voltage, and discharge current have been derived from this model for a variety of electrode configurations. Plasma power was determined to vary linearly with respect to the perimeter of the electrode. In addition, averaged reduced electric field measurements were performed through the use of optical emission spectroscopy and a nitrogen radiative collisional model [2]. The reduced electric field was found to be constant among the electrodes tested and in regards to applied voltage. Finally, ICCD imaging has also been employed to determine how far the plasma expands onto the surface of the dielectric within one voltage period for both the positive and negative half wave. The work aims to adapt the simplest circuit model of volume DBD's to give circuit parameters necessary for understanding surface dielectric barrier discharges.

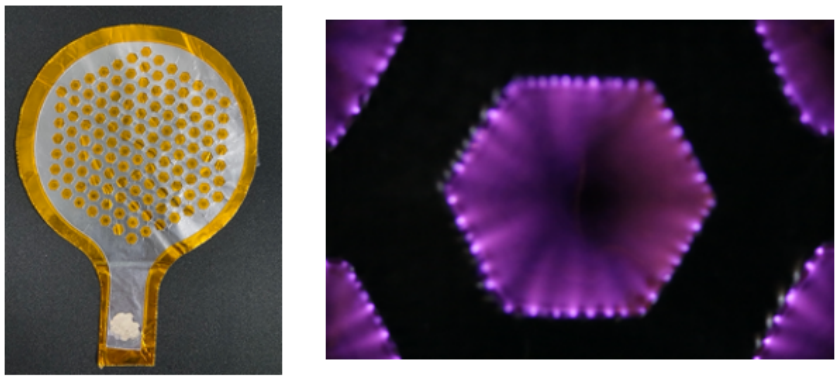

Fig. 1 Left: Prototype of surface dielectric barrier discharge electrode. Right: Close up of plasma generated inside of hexagonal lattice.

[1] A. D. Mazzeo *et al*., *Atoms 7*(1), 14 (2019)
[2] Kogelheide *et al. Plasma Process Polym*. (2020)

Microplasma-Engineered Nanoassembly of Core-Shell Plasmonic Nanoparticles for Ultrasensitive Flexible Surface-Enhanced Raman Scattering Substrates

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Surface enhance Raman scattering (SERS) is emerging as a powerful technology for the label-free detection of various biological and chemical species. [1]One of the important but often overlooked considerations in the design of surface-enhanced Raman scattering (SERS) substrates for trace detection is the efficiency of sample collection. Conventional designs based on rigid substrates such as silicon, alumina, and glass resist conformal contact with the surface under investigation, making the sample collection inefficient.[2]

The aim of our work is to develop an effective paper-based test strip to achieve high SERS performance for biosensing. Here we present one-step green-fabrication of bimetallic nanoparticles Au/Ag (3D porous stricture) loaded on filter paper using microplasma system. This approach requires a minimal fabrication effort and cost, also contains nanoisland and nanogaps (<20nm) on paper substrate to generate a high density of hot spot(~2000/um2). The 3D biomimetric substrate shows a low detection limit for Rhodamine 6G, high averaged enhancement factor, excellent signal uniformity, good stability and suitability in biosensing. In addition, the novel substrate was applied in label-free folic acid, dopamine and other Raman probe detection with small amount and low concentration of analyte, and it exhibited effective SERS platform potential for bio-detection and bio-sensing.

[1] YEH, Yi-Jui; CHIANG, Wei-Hung. Ag Microplasma-Engineered Nanoassemblies on Cellulose Papers for Surface-Enhanced Raman Scattering and Catalytic Nitrophenol Reduction. ACS Applied Nano Materials, 2021, 4.6: 6364-6375.

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Electrical Discharges in Water with Gas Bubbles: Time Scales Approach

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Pulsed electrical discharges in water with and without external gas bubbles have been investigated for decades for water decontamination and other applications. Time scales play a crucial role in the initiation and the development of the discharge. For the gas phase, the important time scales include electron avalanche time, electric field screening time, and the streamer formation time determined by the ion drift. Maxwell relaxation time, electrostriction time, the time for water layers to move into the high field regions, and heating and bubble formation times are all essential for the development of the discharge in the liquid phase. Phenomenological comparison of discharges in gas bubbles in water positioned between metal electrodes illustrates the importance of these time scales in comparison to the ramp rate of the applied voltage. For slow ramp rates of ~0.01 kV/ns, heating at the electrode and the formation of thermally induced bubbles is important, for scales of ~0.1 kV/ns streamers develop in gas bubbles in water, and at ramp rates >1 kV/ns, cavitation effects become important. For example, when a bubble is positioned between electrodes submerged in water and the applied voltage ramp rate of 3 kV/ns the discharge initiates at the tips of the electrodes and then propagates to the bubble before being transferred into the gas bubble.

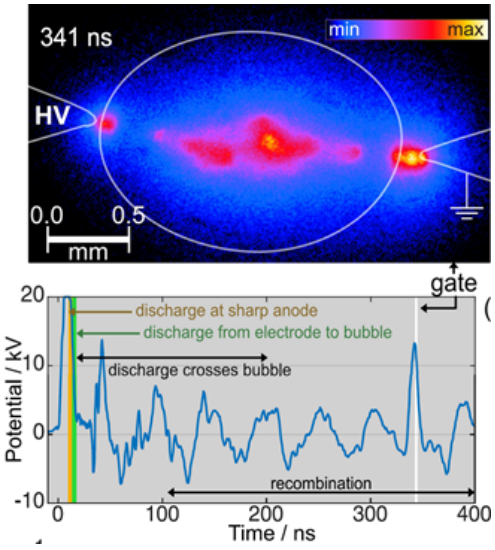

Fig. 1 Discharges in an Ar bubble in deionized water. Applied voltage ramp rate ~3 kV/ns, maximum 20 kV.

Ignition and propagation of nanosecond pulsed plasmas in water with different polarities

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Discharges in liquid enable a variety of applications ranging from wastewater treatment to nanoparticle formation. Pulsed plasmas in liquids ignited by voltage pulses with fast rise times and nanosecond pulse lengths yield a high degree of dissociation of the liquid, a high mass transport and efficient reaction rates with surfaces adjacent to the plasma. However, understanding of the ignition and propagation of these plasmas inside the liquid on these short timescales is still a matter of debate. An indirect method to analyze these processes is the comparison of different voltage polarities applied to the electrode. Different possible processes such as gas bubble or nanopore formation, field effects and the presence of a super critical fluid are evaluated and compared with optical emission spectra and ICCD imaging. A 10 ns long voltage pulse with amplitudes of ±20 kV is applied to a 50 µm thin tungsten wire inside distilled water. The emission intensity, electron density and number of H emitters are similar for both polarities except in the first few ns. This indicates different electron generation mechanisms for different polarities at discharge breakdown. Based on these data, we postulate the ignition and propagation processes to result from electron generation from field effects inside a super critical fluid surrounding the electrode tip.

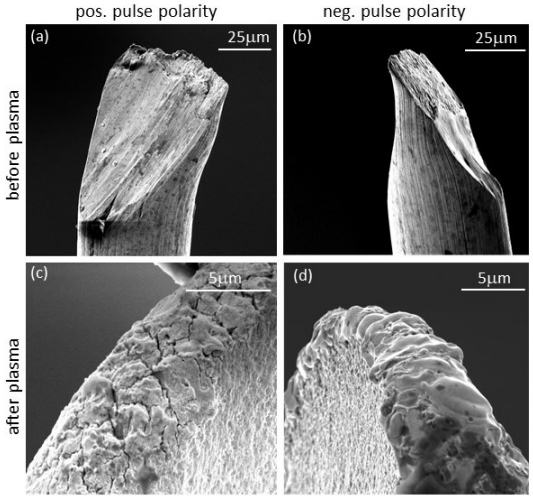

Fig. 1 SEM images before (top row) and after (bottom row) ns plasma ignition in water with positive polarity (left column) and negative polarity (right column) [1].

[1] K. Grosse, M. Falke, and A. von Keudell, Journal of Applied Physics **129**, 213302 (2021)

Recent results on nanosecond discharge in liquid water - signatures of direct and bubble-assisted mechanisms

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Electric discharges in contact with or directly in liquid water produce a number of reactive species that are of interest for many biological and medical applications. The nature of the discharge in liquid water, as in other dielectric liquids, depends on the specific conditions of the discharge (amplitude, rise time, energy of the HV pulse or the shape and polarity of the HV electrode) [1]. The simplest mechanism of discharge initiation in liquids is associated with local evaporation of liquid due to Joule heating and subsequent gas breakdown. However, in the case of (sub)nanosecond HV pulses, heating and liquid evaporation facilitating the onset of the discharge is impossible. Therefore, basic signatures of the direct and bubble-assisted mechanism are still the subject of the active research. The discharge mechanism is connected with very remarkable phenomena such as the emission of ultraviolet radiation, electrostriction, fast phase changes, and shock-wave production. A recent experimental study [2] pointed out that the nanosecond discharge is composed of dark and luminous phases. The dark phase creates bush-like structures made of thin hair-like filaments and occurs with a delay of a few nanoseconds after the HV pulse onset. The propagation of every filament is accompanied by GPa shock-waves [3]. In contrast, the luminous phase has a simple tree-like morphology and is characterised by broadband continua extending from ultraviolet to near-infrared wavelengths. The luminous-phase onset follows about 600 ps after the onset of the dark phase. In the post-discharge phase, the dark-phase filaments radially expand, and form macro-bubble settled at the electrode. The macro-bubble then collapses on the microsecond timescale leaving some long-living micro-bubbles behind. Recent experiments [4] pointed out that the number and the volume of these micro-bubbles remaining in the discharge region grows with the repetition frequency of HV pulses. Consequently, these micro-bubbles, containing non-condensable gases, significantly influence the initiation mechanism of the discharge as well as observable radiative signatures. In this contribution, we will thoroughly discuss the latest experimental and theoretical findings concerning the initiation mechanism of (sub)nanosecond discharges in deionized water.

[1] B. R. Locke et al., Plasma Chemistry and Plasma Processing, 32(5), 875-917 (2021).
[2] M. Šimek et al., Plasma Sources Science and Technology, 29, 9, 095001 (2020).
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[4] P. Hoffer et al., PSST, 31, 1, 015005 (2022).

The Formation of Solvated Electrons in Anodic Plasma-Liquid Systems

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Plasma electrolysis is a form of plasma-driven electrochemistry in which a gas discharge and a liquid electrolyte each form part of the same circuit. In a plasma anode configuration, a positively charged metal electrode ignites a discharge above the surface of the liquid, propelling ions toward the liquid surface. While used in a variety of applications, there are still open questions about the exact nature of these discharges and the solution chemistry they drive. Our group recently proposed a model, wherein incident ions interact with water either through ionization, which produces electrons and hydronium ions, dissociation, which produces hydroxide radicals, or charge exchange, which produces hydronium alone [1]. Electrons then are either ejected in a mechanism analogous to secondary emission, sustaining the gas discharge, or are localized and solvate. Estimates of secondary emission coefficients are ~10^{-5}, indicating that emitted electrons are produced very inefficiently, but it is unknown how efficiently solvated electrons are produced. In this work, we perform total internal reflection absorption spectroscopy (TIRAS) to directly observe solvated electrons at the plasma liquid interface. Their absorption signal is observed to be quenched with the addition of electron scavengers, and enhanced by the addition of hydroxide scavengers, and the absorption spectrum aligns with the well-known spectrum. The dissociative electron attachment of chloroacetate, which yields chloride, is used as a chemical probe to determine the yield of solvated electrons and hydroxide radicals per incident ion. The yield of solvated electrons is found to be ~1.04 electrons per incident ion. The yield of hydroxyl radicals is more difficult to discern, as the exact nature of their reaction with chloroacetate is unknown, but based on their relative faradaic efficiency is thought to be at least 2 per incident ion.

[1] H. E. Delgado, D. T. Elg, D. M. Bartels, P. Rumbach and D. B. Go, "Chemical Analysis of Secondary Electron Emission from a Water Cathode at the Interface with a Nonthermal Plasma," *Langmuir,* vol. 36, no. 5, pp. 1156-1164, 2020.

Interdisciplinary Multiphysics: Using Interface-Resolved Direct Numerical Simulation to Inform Plasma Streamer Modeling

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The ignition of plasmas in liquids has applications ranging from medical instrumentation to manipulation of liquid chemistry. Formation of plasmas directly in a liquid often requires prohibitively large voltages to initiate breakdown. Producing plasma streamers in bubbles submerged in a liquid having a higher permittivity can significantly lower the voltage needed to initiate a discharge by reducing the electric field required to produce breakdown. The proximity of the bubble to electrodes and the shape of the bubbles play critical roles in the way the plasma is produced and propagates through the bubble. Thus, a Multiphysics framework (Figure 1) was devised to transport bubble shape data from Direct Numerical Simulation (DNS) into plasma hydrodynamics simulations wherein the plasma behavior within the bubbles could be studied.

To achieve this goal, an air/liquid plasma reactor that could reliably produce ellipsoidal bubbles was designed, in which air is injected through a novel orifice geometry not unlike that of a flute submerged underwater. After validating the bubble formation behavior seen via DNS with airflow through one orifice, a much larger case was designed to simulate the evolution of bubbles produced out of many orifices in the flute, which was also validated using experiments. In the latter stages of the larger case, the physics of bubbles crashing into electrodes was resolved. The bubble shape data from this case was then transported through the novel Multiphysics framework to a plasma modeling code (nonPDPSIM) that could simulate the plasma hydrodynamics in the bubbles near the electrodes. The behavior of the plasma in those bubbles matched that seen in prior experiments, with the bulk of the plasma typically traveling on bubble surfaces.

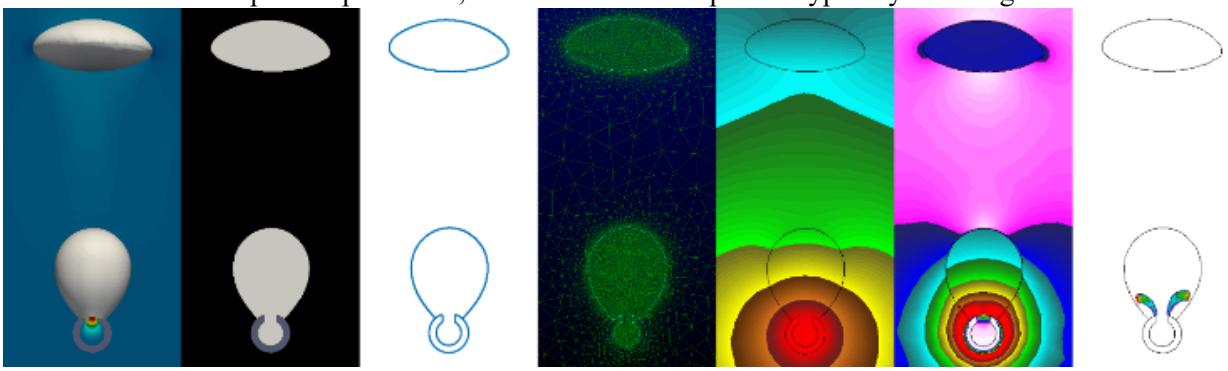

Figure 1: The Multiphysics Framework. In the leftmost pane is a snapshot from a fluid dynamics simulation, and the rightmost pane shows streamer formation in one of the same bubbles through a plasma simulation. The panes in between showcase various steps of the information transfer process from one code to the next.

Plasma-ozone combined advanced oxidation process for decomposition of persistent organic compounds

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Water treatment processes that use OH radicals to decompose persistent organic compounds are called advanced oxidation processes (AOPs). Plasma in contact with liquid is a kind of AOPs, which has been studied for several decades. Many types of plasma treatment have been proposed, and a wide range of processing speeds and energy efficiencies have been observed for the reduction of total organic carbon (TOC) as summarized in [1]. It is well known that hydrogen peroxide (H_2O_2) generated by a self-quenching reaction of OH radicals acts as a scavenger of OH radical, which resulted in a tradeoff between the decomposition rate and energy efficiency in most plasma-based water treatments. Therefore, we proposed a plasma–ozone combined system that uses the reaction between H_2O_2 and ozone (O_3) in a solution to re-generate OH radicals from H_2O_2 by adding O_3 to the plasma-treated solution, as shown as "Remote plasma" in Fig. 1 [1]. The combined system solved the issue of the tradeoff, resulting in a relatively high efficiency comparable to that of conventional AOPs with a higher decomposition rate.

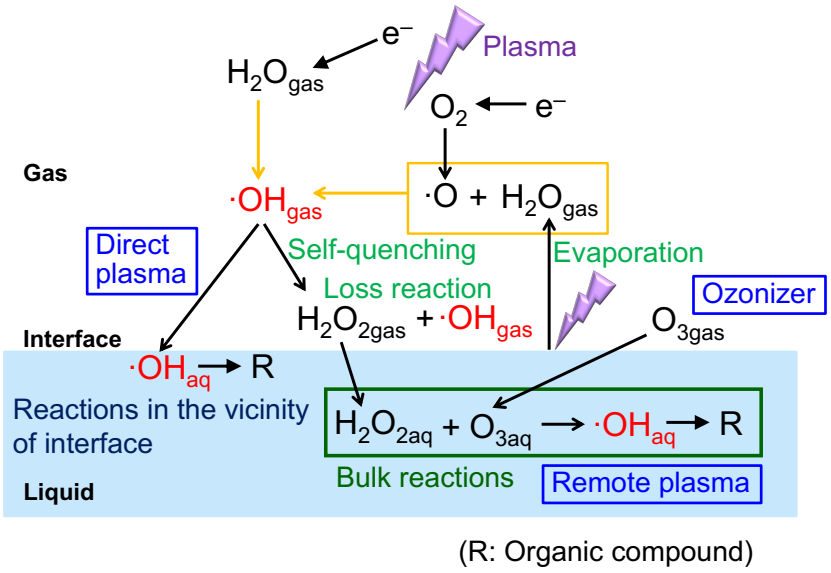

Fig. 1 Schematic of important reactions in the decomposition of organic compounds using O_2 plasma in contact with liquid [1].

[1] N. Takeuchi and K. Yasuoka, Jpn. J. Appl. Phys. **60**, SA0801 (2021).

Experimental investigation of a ns-pulsed Ar plasma jet for the fast desorption of weakly volatile organic compounds deposited on glass substrates at variable electric potential

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A ns-pulsed argon atmospheric pressure plasma jet (APPJ) was studied for the fast desorption of weakly volatile organic molecules (bibenzyl) deposited on glass substrates at variable electric potential (floating-potential or grounded) [1]. The experiments focused particularly on thin resistant bibenzyl films (a large thick bibenzyl deposit was also studied), which are more difficult to be desorbed when using a substrate at a floating-potential. The APPJ was probed by means of high-resolution laser absorption spectroscopy to map the Ar($1s_5$) metastable absolute density (spatially and temporally resolved) at the close vicinity of the glass plate where bibenzyl was deposited. Furthermore, the electrical, optical and thermal features of the APPJ were investigated systematically. In this way, the plasma desorption efficacy on thin resistant bibenzyl deposits was evaluated for the envisaged application, by varying the exposure time of the molecules to the APPJ (t_{exp}, from 10 s up to 180 s). The obtained results confirm the relatively low desorption efficacy in the case of a floating-potential substrate, which improves to some extent with increasing t_{exp}. However, when the substrate is grounded, the effect of the plasma becomes much more significant (i.e. much higher desorption efficacy). Besides, contrary to the case of a floating-potential substrate, an almost complete desorption of bibenzyl is achieved for t_{exp} = 180 s. Similar effects of the APPJ were recorded on a thick bibenzyl deposit, validating the previous results. For both operating conditions (floating-potential and grounded substrate), the plasma action should be due to the production close to the glass surface of relatively high densities of Ar($1s_5$) (up to 2×10^{13} cm^{-3}) and of oxidative species, such as atomic oxygen, hydroxyl radical and ozone. Thermal effects might play a synergistic role only when the substrate is grounded, since relatively high gas and glass-surface temperatures (>60 °C) are reached only in this case. The present results are of interest for public-security applications related to the fast detection of resistant prohibited substances, such as narcotics and explosives.

[1] K. Gazeli *et al.*, J. Phys. D: Appl. Phys. **53**, 475202 (2020).

Far UV-C Microcavity Plasma Lights for Inactivating Airborne Pathogens in Occupied Spaces

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The natural pandemics of influenza and coronaviruses over the past decade have caused national security threats from civil unrest, economic destruction, and morbidity across the globe [1]. Natural pandemics are likely to recur based on zoonotic outbreaks as evidenced over the past decade (e.g., H1N1, SARS, MERS, Ebola) and the generation of new vulnerable birth cohorts every year [2]. COVID-19 illustrates the gravity of biological threats, especially to the United States and highly populated countries given its inadequate preparations and means of response [3].

In this paper, a microplasma flat lamp with its center emission wavelength at 222 nm from an atmospheric pressure of $KrCl^*$ excimer will be introduced for its performance to prevent airborne transmission in an occupied indoor space. Recently, the far UV-C wavelength (200-235 nm) has been known (reported) not to induce essentially any damage to skin or eyes even during the exposure against the treatment by the conventional germicidal lamp. At a controlled UV fluence, specified by UV TLVs guidelines from the both ACGIH (American Conference of Governmental Industrial Hygienists), the microplasma lamps demonstrated technical (and scientific) advantages over conventional techniques in inactivating various infectious pathogens, including various coronaviruses including SARS-CoV2 (COVID-19) virus [4]. Details concerning the lamp characteristics, application studies, and prospects of the preventive uses of these microplasma far UV-C tiles in both pandemic and post-pandemic eras will be discussed.

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Microplasma-enabled synthesis of graphene quantum dots from plastics

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Plastic products are common used in our daily life, however, most of the plastic wastes are not well-treated or recycled. Here we report a simple and environmental-friendly method to synthesis grapheme quantum dots from plastics using microplasmas. The synthesized GQDs indicate emission-tunable property and high physical and chemical stabilities, making them useful materials for applications. Moreover, the synthesized GQDs exhibited a 4.0 nm averaged diameter with narrow size distribution based on TEM analysis. Furthermore, the synthesized GQDs show potentials for various detection applications including temperature, metal ions, nitroarenes, and biomolecules, providing a new direction for sensing technology. Our work provides a step for simple fabrication of GQDs from plastics and emerging applications using GQDs

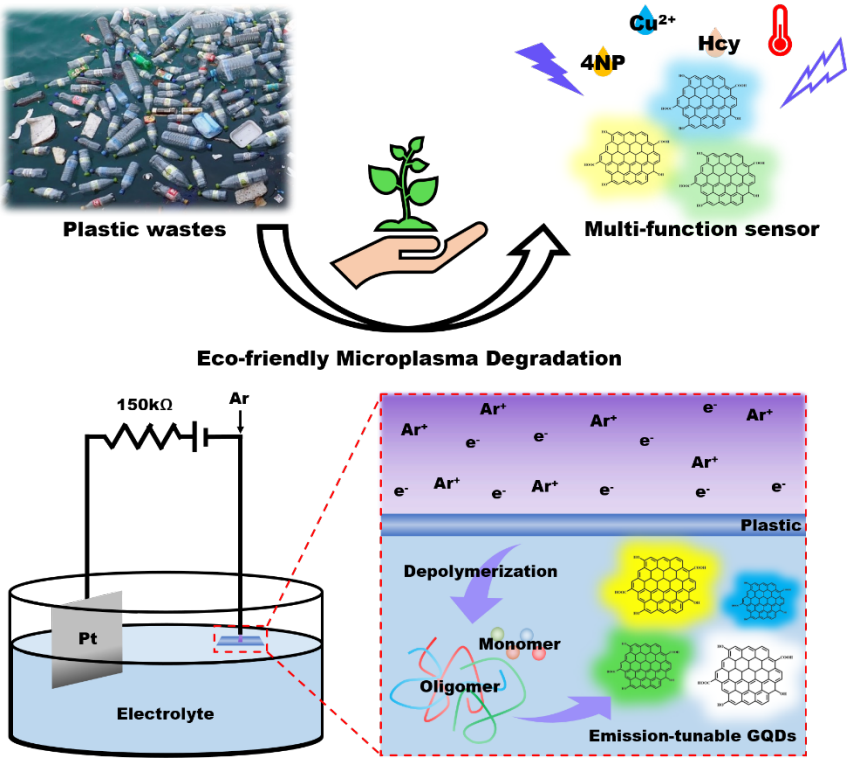

Fig. 1 Schematic preparation and application of WGQD

Microplasma assisted and One-step fabricated Silver/Nitrogen doped graphene quantum dot (NGQD) nanohybrids for enhance Raman detection

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Recently experimental and theoretical works have reported that graphene quantum dots (GQDs), a unique form of zero-dimensional nanostructure, and their exceptional properties make them promising in biosensing applications. Surface-enhanced Raman scattering (SERS) is an ultra-sensitive analytical technique for bio-molecules detection. While the potential of surface plasmon resonance (SPR) metals (e.g. Au and Ag) and graphene for SERS has been demonstrated but the work of GQDs applied as SERS substrates is still lacking.

Furthermore, modified GQD with metal nanostructures will lead to important advance for SERS-based detection. Here we demonstrate a facile synthesis of NGQD/AgNP nanohybrids by the atmospheric-pressure microplasma-assisted electrochemistry. Detailed nanomaterial characterizations including transmission electron microscopy, UV/Vis spectroscopy show that the microplasma-assisted electrochemical reaction can successfully grow Ag nanoparticles (AgNP) onto the NGQD surfaces to form the Ag@NGQD NP nanohybrids with heterodimeric nanostructures within minute scale. Besides, the photoluminescence (PL) optical study of NGQDs and AgNP@NGQD indicated that the non-radiative fluorescence resonance energy transfer (FRET) involved in the AgNP@NGQD nanohybrids. We further studied the NGQD-AgNP nanohybrids with different FRET efficiency. The results revealed that FRET of the as-produced AgNP@NGQD nanohybrids is the dominant factor to SERS properties in our study.

Microplasma-Reinforced Fabrication of Crosslinked Nitrogen-Doped Graphene Quantum Dot Sponges for Environmental Water Purification Applications

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Quantum confinement of graphene quantum dots (GQDs) yields a unique photoluminescence (PL) properties which has been widely used for many applications including imaging, sensing, LED, optoelectronic devices, and energy related applications. This performance can be further enhanced by introducing heteroatom, such as nitrogen (N) into the sp^2 carbon core. The high electronegativity of N atom has been proven to provide better electron transfer for improving both sensing and catalytic performances. Despite the promising facts, the applicability of NGQDs have been hampered by their synthesis methods, including uncontrolled NGQDs properties, long reaction time, laborious synthetic procedures, harsh reaction conditions, and scalability. Moreover, the incorporation of NGQDs in to an appropriate polymeric matrix can serve as both stimuli response probe in the matrix and nanoscale cross-linker which can further enhance the mechanical properties with the absence of any additional chemical cross-linkers.

Water contamination has undoubtedly been one of the most concerning problem faced by many people, including heavy metal ions, organic dyes, and organic pollutants which are highly toxic and dangerous to human being. Among other available methods, adsorption has been one of the most promising method to remove those contaminations from water owing to its effectiveness, efficiency, low cost, and simplicity. Microplasma as gaseous discharge with one dimension geometrically confined to <1 mm emerges as an alternative way to promote crosslinking and polymerization process with the absence of toxic chemical crosslinker at ambient conditions. Besides, microplasma is also capable of cleaving polymeric chain to generate porous structures in the resulting 3D materials, resulting in enhanced pore size, surface area, hydrophilicity, and wettability. Herein, we utilized a microplasma system to synthesize NGQDs from chitosan at ambient conditions. The post-plasma solution was converted into NGQDs gel by neutralizing the unreacted chitosan with sodium hydroxide (NaOH) and subjected to another microplasma treatment for crosslinking process to generate NGQDs hydrogel. The fabricated NGQDs hydrogel can be further converted to NGQDs sponge by subsequent freeze-thawing method, showing remarkable water uptake and dye adsorption ability for remazol brilliant blue R (RBBR). Our work provides an insight into the green and sustainable development of NGQDs sponge applicable for water environmental application.

Plasma-assisted materials processing with inkjet droplets

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With development of microplasma sciences and technologies, plasma-solution interactions have been extensively studied in various fields. Here, I am focusing on plasma-assisted materials processing with controlled microdroplets. Two systems will be presented in the workshop. The first one is (A) plasma-assisted inkjet printing (Fig. 1), where high spatial and temporal reproducibility of inkjet-droplets is combined with plasma processing. Relatively low-temperature, rapid, and simple line fabrications with various inks [1–3] will be presented. The other is (B) plasma-assisted monodisperse particles synthesis (Fig. 2), where the feature of high size reproducibility of inkjet-droplets is applied to plasma processing. Monodisperse particles synthesis [4] as well as those with porous structures will be presented with several examples. Particles-synthesis process will be discussed with direct observations of flying droplets and a model considering transports of materials and energy in/on the droplet [5].

This work was performed under collaborations with Prof. Kobayashi (Keio Univ.), Dr. Shimizu (AIST), Prof. Tershima, Dr. Muneoka, Mr. Nitta, Mr. Tsumaki, Mr. Hato, Mr. Sakai, Mr. Ishizumi, Mr. Kawano, and Ms. Jeon (UTokyo). This work was partially supported by JSPS KAKENHI.

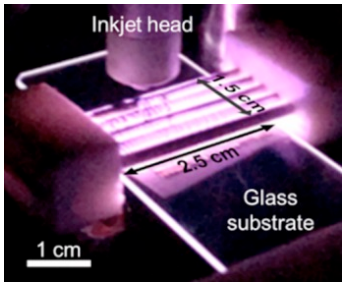

Fig. 1 Plasma-assisted inkjet printing [3].

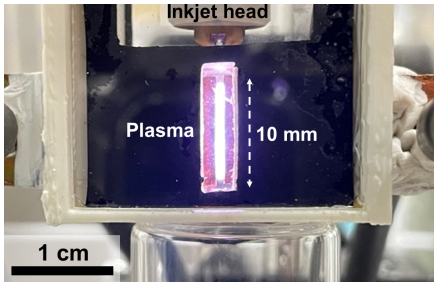

Fig. 2 Plasma-assisted particles synthesis with inkjet-droplets [4].

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SurfAP3® – Direct Writing Micro Plasma Printing for Localized Surface Modification of Biosensors and Microfluidic Devices

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Plasma-assisted surface modification processes can generate materials with novel properties by creating new surface functionalities or depositing thin layers, which can be useful for biosensing and microfluidic applications, in order to control the physical, chemical and biological interface properties that guarantee the optimal performance regarding the interaction with the biological environment. This contribution presents a robust and flexible approach for the fabrication of chemical micropatterns of controllable dimension, based on the deposition of plasma-polymerized (pp) polymer films using a proprietary atmospheric-pressure plasma printing technology, known as SurfAP^3®. This technology, in which a microplasma is generated via a sinusoidal high voltage driven spotlight transient spark discharge, allows the targeted, maskless modification of small and large surfaces regardless of their geometry and cavities, using a fast and eco-friendly procedure, without additional washing steps. Moreover, it offers easy scalability, long-term stability of the modified surface, applicability to nearly all materials/surfaces independent of their conductivity and the production of a multitude of functional groups. SurfAP^3® is able to produce structures with dimensions starting from 50 µm and coating thickness between 20 and 500 nm, on multiple materials (Fig. 1). Additionally, the amount and type of functional groups and the surface morphology, can be tailored to adjust a certain chemical platform, suitable for multiple biosensing and microfluidic applications. [1].

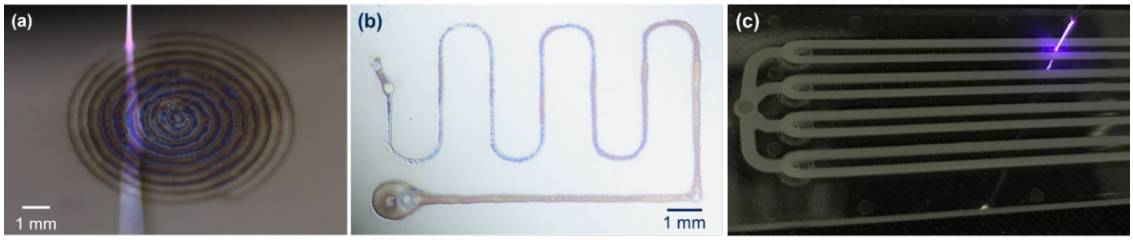

Fig. 1 Deposition of plasma-polymerized micropatterns on silicon wafer (a-b) and a polystyrene microfluidic chip (c), using SurfAP^3® technology. The microplasma discharge is seen in (a) and (c).

[1] L. Barillas, E. Makhneva, K. D. Weltmann, H. Seitz and K. Fricke, Microelectron. Eng. **233**, 111431 (2020).

Plasma polymerization with single-filament DBDs at atmospheric pressure: The role of ions

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Thin film deposition activated by dielectric barrier discharges (DBDs) has been widely investigated regarding plasma-chemical processes and thin film properties. To this day it is not understood to what extent radicals or ions take part in thin film formation. Often only radicals are assumed while the production of ions via direct ionization and especially via Penning ionization is proven. The use of single-filament DBDs (SF-DBDs) with short gas residence times in mixtures of hexamethyldisiloxane (HMDSO) with argon has shown that the pentamethyldisiloxanyl cation ($PMDSO^+$, $(CH_3)_3Si\text{-}O\text{-}Si(CH_3)_2^+$) is the main contributor for film formation. For monomer fractions x_M exceeding 1000 ppm an ionic oligomerization process is observed.

Studies of thin film deposition under conditions of dominating ionic deposition can lead to a better understanding of the underlying mechanisms and possibly to new processes and applications. In the present contribution, we report results of SF-DBD investigations on mixtures of argon with silicon-containing monomers or hydrocarbons such as hexamethyldisilane (HMDS), allyltrimethylsilane (ATMS), tetramethylsilane (TMS), and ethane, respectively. Due to ionization energies of the monomers lower than the energies of long-lived excited argon species, a strong Penning effect for x_M growing up to a few 100 ppm is observed, resulting in an overall diffuse plasma appearance, a decline in the extinction voltage, earlier discharge ignition and a decrease in the dissipated energy. For some precursors, HMDS for example, it is not clear which ion is formed via Penning ionization at atmospheric pressure: From mass spectrometry data obtained with electron-collision ionization [1], the formation and deposition of trimethylsilyl cations ($TriMS^+$, $Si(CH_3)_3^+$) could be expected but a comparison of the deposited volume with the volume calculated from the transferred charge indicates a deposition of pentamethyldisilyl ions ($PMDS^+$) rather than $TriMS^+$.

For elemental and structural analysis of the plasma polymers FTIR-ATR, Raman and WDXS measurements were used. The oxygen content of the deposits was generally below 5 at%. Interestingly, spectra of deposits obtained by single-filament DBDs often resemble spectra reported for films obtained by low-pressure plasma processes (see [2] for HMDS).

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Advances in optical diagnostics of Microplasmas: imaging and spectroscopy

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Microplasmas are a sizable part of non-equilibrium non-thermal plasma sources. The latter have been intensively developed since the mid-nineties and revealed their unique properties to yield a rich reactive chemistry at elevated pressure up to atmospheric pressure [1]. By definition, microplasma reactors have – at least– one of their dimension in bellow the millimeter scale. Micro-hollow cathode discharges (MHCD) are well-known example of miniaturized reactor of hundreds micrometers which can operate at atmospheric pressure in DC or AC[2]. Widely studied to support many kind of applications related to environment purposes, microplasmas are still being challenging to study experimentally due to their characteristic dimensions [1].

Indeed, to better understand the elementary processes of the discharge as well as to control the reactive kinetic chemistry involved in the targeted plasma processes, experimental diagnostics are mandatory. Apart from a thorough analysis of the electrical circuit which can already bring substantial information, optical diagnostics techniques are commonly used to measure some plasma parameters. Due to the limited access, the investigations are often delicate and require more efforts to carry out proper measurements. In this contribution one will present several recent approaches used to determine key parameters of the discharge such as the gas temperature, the electron number density, metastables densities or the hydrodynamic forces[3,4]. The focus will be put on the capability to performed space resolved measurements and to reveal the gradients in two different microplasma reactor systems. Comparison with some modeling results published in the literature will be discussed. This presentation will be also an opportunity to discuss further outlook and opportunities to extend already existing diagnostic methods to micrometer scale plasma sources.

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Fast framing imaging of the kINPen science onto a surface – transient spark formation and subsequent guided streamer pathways

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The science on plasma jets interacting actively with a surface is of significant relevance for many applications. Open jets are known to generate guided streamers per voltage period. In case a surface is placed downstream, varieties of effects occur from charge storage to electrical field distortion.

The present contribution investigates the kINPen science interacting with a copper surface. The kINPen science is a plasma jet based on a 1 MHz driving frequency, voltages of up to 3 kV peak to peak and with a gas supply of 5 slm argon. The operating voltage and input current are measured as well as the current at the surface. The gap distance is set to 7 mm. A fast framing camera (Kirana, Specialised Imaging) is operated with up to 5 MHz repetition rate, acquiring 180 images in a row. The formation of a transient spark with an unsteady repetition frequency occurred. The subsequent guided streamers follow the channel created by the transient spark. Over longer exposures, movement of repetitive channel structures equals the gas flow velocity. In single acquisitions, structures similar to striations were observed as well.

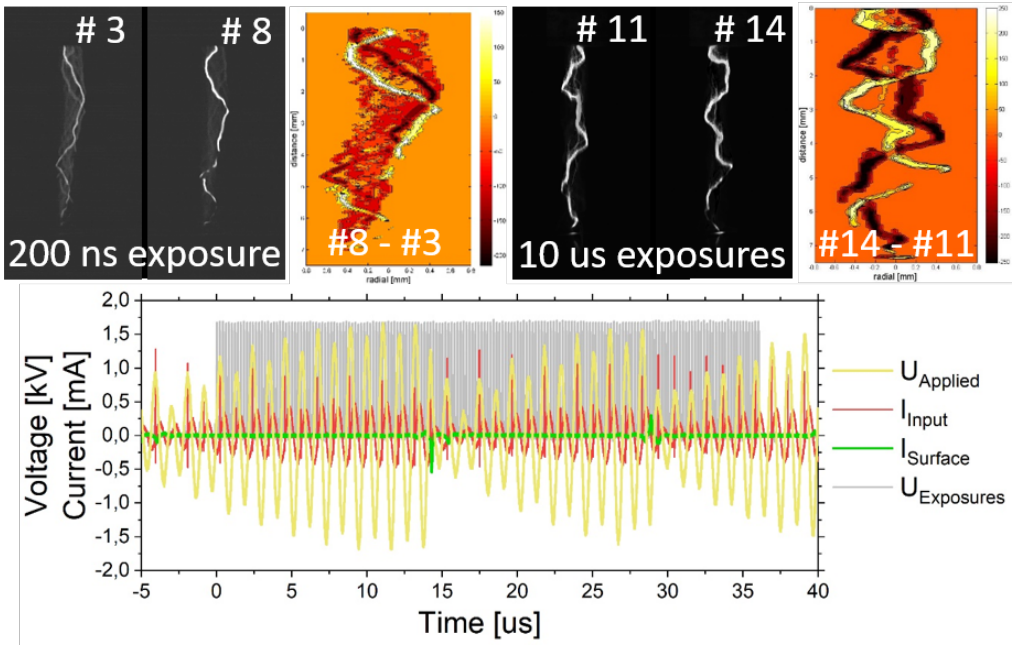

Fig 1: Images of guided streamer formation within two voltage cycles and an image subtraction (200 ns and 10 us exposures), electrical signal of correlated voltage, input and surface current

Atomic nitrogen spatial profile in three different MHCD configurations

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Atomic nitrogen (N) sources are highly requested for many material synthesis applications. A typical example is the production of hexagonal boron nitride (h-BN) which is a very attractive substrate for graphene-based technologies. However, the generation of N atoms is challenging due to the elevated bond energy of the N_2 molecule (~10 eV). Micro-Hollow Cathode Discharges (MHCD) are promising plasma sources that allow for the production of high densities of N atoms, while preserving a low injected power (1 W). A previous study of a one-hole MHCD [1], which was ignited in an Ar/N_2 gas mixture by means of a DC power supply, showed its capacity to generate high densities of electrons (up to 10^{14} cm^{-3}) as well as N atoms (up to 8×10^{14} cm^{-3}). These values could be even further improved when using a pulsed excitation, e.g., electron density can reach up to a few 10^{15} cm^{-3} [2].

The main drawback of the MHCD is their small plasma volume, which is confined in a hole of a few hundred of µm (figure 1.a), preventing the treatment of relatively larger surfaces. To expand the plasma volume, we studied different MHCD configurations by introducing a pressure differential between the two sides of a MHCD, creating a plasma jet in the low pressure chamber (figure 1.b), and adding a third electrode in front on the MHCD (figure 1.c). In each case, the spatial distribution of the density of N atoms was measured by means of nanosecond Two-photon Absorption Laser Induced Fluorescence (TALIF).

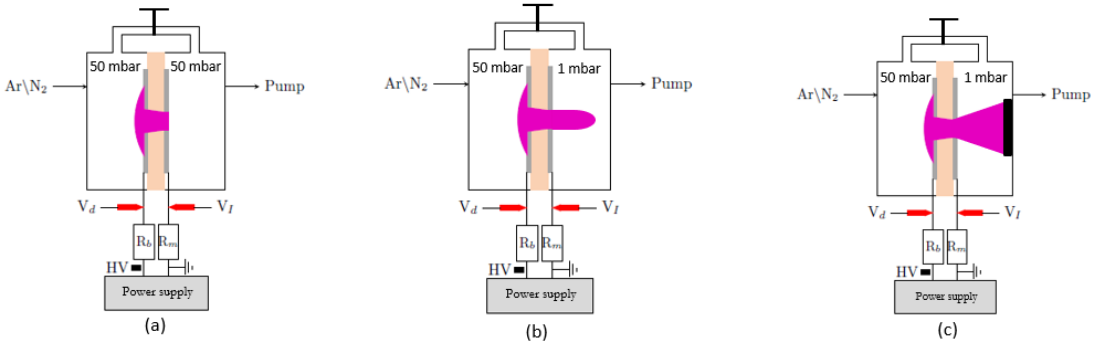

Fig. 1. Scheme of the MHCD configurations studied: (a) traditional MHCD, (b) MHCD jet, (c) MHCD with a third electrode.

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Using Non-Thermal Plasmas to Direct Chemistry at Solid and Liquid Interfaces

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One of the primary features of non-thermal plasmas is that they generate an abundance of reactive species that can be used to produce useful chemistry. While many of these reactions occurs in the gas phase, the abundant reactive species, including electrons, ions, radicals, and metastables, can also initiate chemistry at a surface, forming the basis of plasma technologies for applications ranging from microelectronics fabrication to medical therapeutics. Two areas where there have been significant advances over the past several decades are plasmas in contact with liquids to drive liquid chemistry and plasmas in contact with catalysts to drive gas chemistry. In both cases, multi-phase interfacial phenomena are critical to understanding the underlying plasma physics and the ensuing chemistry. This talk will overview developments in both of these areas, highlighting recent advances by our group and pointing to opportunities for future developments. One of the primary challenges in plasma-liquid systems is understanding the coupling between the plasma and liquid, including the electrostatics and charge transfer that are essential to maintaining the plasma in contact with the liquid. In our work, we have extensively studied how electrons are transferred from the plasma and dissolve into the liquid, becoming solvated and leading to a wide variety of reduction reactions. More recent efforts include understanding how solvated electrons are produced by ions bombarding the liquid. In plasma-catalysis, there are a wide variety of unknowns about how plasma species couple to a catalytic material. Typically, reactor design and optimization has been based on heuristic approaches, often starting from known strategies from thermal catalysis. Our recent efforts indicate that both catalyst and reactor design need to be rethought when coupling the non-equilibrium environment of a plasma to a catalyst.

Fabrication process and characterization of through silicon-based microplasma reactors

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To answer new challenges such as the development of original deposition methods, a new design of silicon-based microplasma reactors with a through silicon via (TSV) was developed. Based on a previous version of Micro Hallow Cathode Discharge (MHCD) [1], the novelty of this work lies in the backside processing of the microreactor to open the cavity and create a gas flow in the hole. This was performed using the patented STiGer etching process developed at the GREMI laboratory [2] to obtain an anisotropic 300 µm deep TSV (Fig.1 a)).

Electrical and optical diagnostics were conducted in helium microplasma for different pressures. So far, the MHCD with a TSV proved themselves more durable and capable of sustaining higher current than the previous version [1]. Although, a so-called "self-pulsing regime" [3] is observed below a certain current threshold Fig.1 b), a steady glow discharge following a normal regime is obtained at both high and atmospheric pressure. This is because the TSV enhances considerably the size of the cathode, which gives more surface to the plasma to expand. Electrical and spectroscopic characterizations have been carried out and compared to the results obtained in a closed cavity.

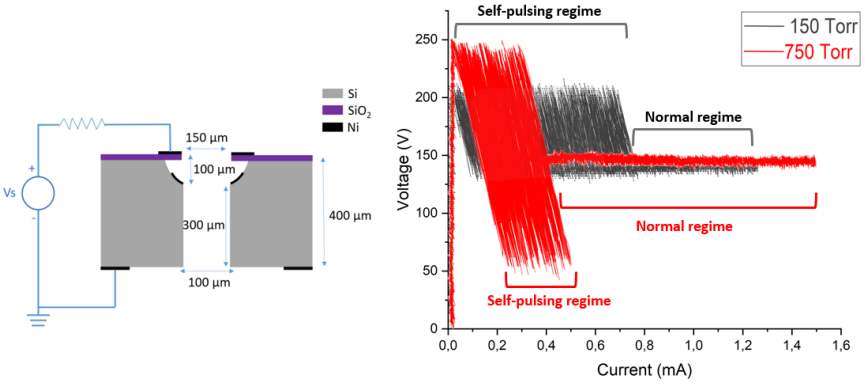

Figure 1: a) Design of the microreactor, b) V-I characteristics in He with a 1 MΩ ballast resistor.

Acknowledgement: This work is supported through the ANR project PlasBoNG. (ANR-20-CE09-0003). The CERTeM 2020 platform has provided most of the equipment necessary to prepare the microreactors.

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A High-*Q* Microwave Resonator-based High-Efficiency Atmospheric Microplasma Jet

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There is a need for high efficiency, low power, and low-cost plasmas to facilitate widespread use. Due to their ability to store and enhance electromagnetic energy, it is possible to employ microwave resonators to achieve high-efficiency plasmas. This study uses evanescent-mode (EVA) cavity resonator technology to introduce a highly efficient microwave microplasma jet at atmospheric pressure. The EVA cavity resonator is formed by loading a normal cavity by a post at the center. One important consequence of this loading is the electric field concentration in the gap between the resonator post and the top wall. This feature was previously used to realize novel plasma-based microwave limiters [1] and switches [2]. However, this work implements a gas flow mechanism to realize a high-efficiency resonant microwave plasma jet. The theory of operation, as well as simulation and measurement results of EVA cavity resonant plasma jet, are discussed.

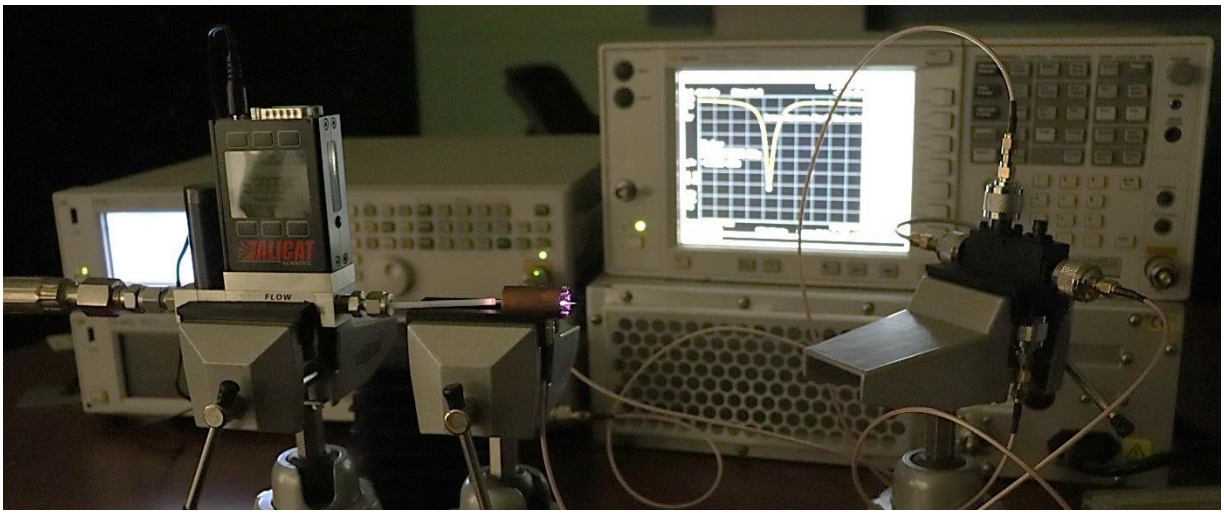

Fig. 1 Picture of the microplasma jet setup.

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Triple Junction-Enhanced Thermally Induced Gas Discharges Using Pyroelectric Crystals

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Thermally cycling a pyroelectric crystal induces a change in its intrinsic spontaneous polarization, leading to very high surface potentials that in turn induce gas breakdown in the vicinity and form a discharge. This property opens the possibility of using pyroelectric crystals to form thermally-induced microdischarges without the need for a high voltage power supply. While this concept has been demonstrated previously [1], the resultant current was too weak to have any practical application. In this work, we perform experimental investigations to enhance the strength of the thermally-induced gas discharges by creating a 'triple junction' on the pyroelectric crystal surface. The triple junction in this work is the junction where the pyroelectric crystal, an aerosol jet printed conductive silver layer, and air meet, at which the electric field is enhanced and gas discharges are formed more easily [2]. Preliminary results show that there is a noticeable increase in the peak current measured with the coated crystals undergoing the same thermal cycle as an uncoated crystal. Peak currents and total charge produced in the microdischarge are compared among crystals printed with different triangular silver patterns from 0° (crystal is fully covered by silver layer) to 360° (no silver layer). The measured results vary with the angles but do not show a symmetry with respect to 180°, indicating that there is possibly a 3D effect in the mechanism of triple-junction enhancement.

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Effects of TiO_2, ZnO, and $SrTiO_3$ on DBD plasmas through N_2/O_2 and $N_2/O_2/CH_4$

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It has been long understood that different dielectric barrier discharge (DBD) configurations can result is significantly different chemical production. Such differences can include electrode gap, electrode material, dielectric thickness, dielectric materials and a host of other design parameters. In this study we examined the impact of photocatalytic dielectric materials on the resultant gas phase chemistry in DBD plasmas. For maximum consistency, semiconductor device grade manufacturing processes were followed. Specifically, photolithography was used to pattern the electrodes on two sides of a 0.5 mm thick alumina substrate. Two μm thick gold electrodes where then deposited using a physical vapor deposition (PVD) tool. For the DBD systems with photocatalytic dielectric materials, prior to the electrode deposition step, a layer of 2 μm of either ZnO or $SrTiO_3$ was deposited and annealed on the alumina substrate. We then operated the DBD plasmas at 6 kV peak to peak and 20 kHz in a closed vacuum system in which we could create highly repeatable atmospheric gas mixtures of O_2/N_2 and CH_4. In comparison to the bare alumina surface, $SrTiO_3$ shows a reduction in NO_x species, while the TiO_2 shows a slight reduction in CH_4. In this report, we will explore the processes behind the observed differences.

Microscale microwave argon discharges: global model and particle-in-cell simulations

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Low temperature microplasma discharges operated in sub-millimeter size have emerging applications in biomedicine, satellite propulsion, environmental remediation, nanomaterial fabrication and more [1, 2]. High-fidelity numerical simulations are key to understanding the fundamental discharge processes due to the time and space resolution limitations of experimental diagnostics. In this work, a global model [3] and particle-in-cell simulations incorporating excited state species as fluids [4] are adapted to address the importance of the fundamental processes involving excited state species (Ar^m, Ar^r, Ar^{4p}, Ar_2^*) in microwave argon discharges sustained between two parallel plates. Firstly, a global model [3] is used to identify the role of different reactions in wide range of pressures (100-760Torr) and to understand the role of Ar_2^* and Ar_2^+, which are often neglected when modelling intermediate and low pressure discharges. Secondly, the effects of the excited state atoms involved in bulk reactions including metastable pooling and photoemission on plasma characteristics are quantitatively evaluated for an argon microplasma driven by a 5×10^4 A/m^2 current source at 1 GHz. The pressure is fixed at 100 Torr and the plate spacing is 0.2 mm. Processes involving excited species including step-wise ionization and metastable pooling enhance the density by a factor of 6 compared to the case in absence of excited state atoms. Due to the relatively small driving current density, the discharge properties are not affected by secondary electron emission from the surface bombardment of ions and excited states, implying that the discharge operates in the alpha mode.

This work supported in part by the Air Force Office of Scientific Research (AFOSR) MURI Grant No. FA 9550-18-1-0062, AFOSR Grant FA9550-21-1-0367, and U.S. Department of Energy Office of Science, Grant No. DE-SC0022078.

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An overview of plasma modeling capabilities in SOMAFOAM using dielectric barrier discharges as a case study

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Low-temperature microplasmas have a number of applications due to their unique physical and chemical properties. Therefore, it becomes extremely important to obtain a better understanding of their characteristics to better control and tune their parameters to achieve desired outcomes. While significant advances have been made in diagnostics and measurements, numerical simulations remain an important component of advancing our understanding of microplasmas. This talk will present an overview of the plasma modeling capabilities, with specific emphasis on microplasmas, offered by the open-source code – SOMAFOAM [1] – that is under active development in our group. SOMAFOAM has several features that are desired in a plasma modeling framework including run-time choice of drift-diffusion or full-momentum equation for each species, arbitrary chemistry, arbitrary geometry, and massive parallelism to name a few. In this work, we use an argon dielectric barrier microdischarge to demonstrate the features of the code including robustness and applicability for a wide range of operating conditions. Specifically, one-dimensional simulations are presented for the ignition and operating characteristics for a wide range of pressures (few Torrs to atmospheric pressure), frequencies (six orders of magnitude ranging from 1 kHz to microwave), and various applied voltages. The simulations reveal various operating characteristics including single-pulse mode, multiple current pulses mode, and glow mode depending on the operating parameters. Modeling results are compared (qualitatively or quantitatively) with experiments when appropriate.

[1] A. K. Verma and A. Venkattraman, *Comp. Phys. Comm.* **263**, 107855 (2021).

Characterization of Helium and Argon Plasmas Under Different Operational Waveform Regimes Operating at Atmospheric Pressure at Radio Frequency

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Helium and argon are used in the characterization and study of plasma phenomena, as in the case of the former, it is easy to ignite, while the latter provides a low-cost alternative to the former. Yet, while ignition characteristics effects on plasmas have been studied at low pressure at the radio frequency (RF) regime, ignition characteristics effects on plasma operating at RF, atmospheric pressure are not well understood. In this study, unipolar and bipolar pulsed ignition waveforms, alongside under-damped ignition waveforms are studied to have a more encompassing understanding of the waveform effects on the plasma characteristics. An in-house numerical solver based on OpenFOAM, SOMAFOAM[1] is used to get a numerical solution to the plasma characterization to the ignition characteristics. A validation for both helium[2] and argon[3], based on previous work is done to confirm the validity of our in-house solver and to have a point of reference on helium and argon ignited. Parameters in the plasma affected by the ignition characteristics, such as number density of species, the voltage within the gap, current density, electron temperature, and energy are presented as a function of the ignition waveform to have a more holistic understanding of the effects of the waveforms.

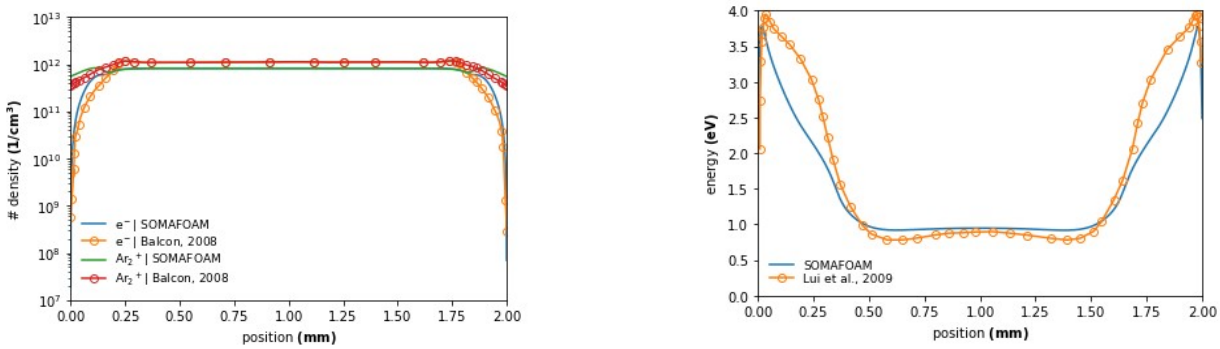

Fig. 1 validation of in-house code for argon and helium cases using timed-averaged number density for argon and timed-averaged energy profile for helium, respectively

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Modeling of a Microplasma within a Photonic Crystal at 43 GHz.

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The electron density (n_e) and argon metastable density ($1s_5$) of a 43 GHz microplasma are obtained using the Zero-Dimensional Plasma Kinetics solver (ZDPlasKin) [1] for an incident power of 100 and 1000 mW and pressures of 10 to 600 Torr. These simulations are compared with the experimental data found in a previous study [2]. To determine the self-consistent electric field in the modeled plasma, the three-dimensional millimeter wave fields are computed as a function of electron density using ANSYS EM19.2, HFSS®. This EM field model is coupled to ZDPlasKin such that any increase in the simulated plasma density correctly attenuates the electric field within the microplasma.

The simulation is found to be very sensitive to argon gas temperature, so a two-zone temperature model was needed to obtain agreement with experimental measurements. The temperature at the central core of the microplasma was determined using the Lorentzian line shape of the absorption profile at 811.53 nm by using laser diode absorption spectroscopy. This temperature was used as input to the model for the simulation of volume recombination losses. The outer regions of the microplasma are assumed to be in equilibrium with the walls (300 K). This second temperature was used in the model to determine diffusion losses.

The modeled electron and metastable density are the order of 10^{18} m^{-3} and 10^{20} m^{-3} respectively. This is in a good agreement with those measured experimentally [2], [3], as long as the two-zone temperature model is used. Specifically, we observe and model that the argon metastable density decreases dramatically as the gas pressure approaches 600 Torr due to strong ionization of the metastables by electrons. In the absence of a hot gas core, the modeled three-body recombination rates are excessive and the simulation severely under-predicts the electron density and over-estimates the metastable density. We conclude that the millimeter wave microplasma has a hot core (~2500 K at 600 Torr) that rarifies the argon gas and effectively stops three-body recombination. This allows one to achieve high electron density on the order of 10^{20} m^{-3} with only 100 mW of wave power. This work was supported by the U.S. Department of Energy under Award No. DE-SC0021249

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http://www.zdplaskin.laplace.univ-tlse.fr (University of Toulouse, LAPLACE, CNRS-UPS-INP, Toulouse, France, 2008)

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Numerical modeling of the intra- and inter-cellular behavior influenced by cold atmospheric plasmas

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Cold atmospheric plasmas (CAPs) have been widely applied in the field of biomedicine. In these works, specific CAP effects on cells and biomolecules via various pathways have been extensively investigated. A number of works have suggested that the CAP-induced reactive species affects cell fate (cell survival/death) [1]. For a further understanding of how CAPs exert their biological effects, the author has developed a zero-dimensional intracellular biochemical reaction model quantifying the influence of CAP-induced reactive species on the mitochondrial redox-mediated functions and energy metabolism [2].

This study examines the effects of CAP-induced reactive species on intracellular cell fate determination (biochemical reactions at the single cell level) and intercellular immune behavior (collective cell behavior) in two separate (but linked to each other) numerical models. Figure 1 shows a conceptual diagram of the current modeling. The intracellular biochemical model focuses on the induction of cell death (apoptosis / necrosis) by CAP-induced stress. The 0D model includes activation of death-receptors, mitochondrial signaling cascade pathways, initiator- and executor-caspases and regulators. The intercellular immune model represents the innate immune system of leukocytes (neutrophils and macrophages) against bacteria. Here, the 2D dynamics of leukocytes, namely migration and phagocytosis, are simulated. In either case, the effects of CAP irradiation can be modeled as extrinsic or intrinsic stress.

This work was partially supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (C) JP19K03813 and Fostering Joint International Research (B) JP20KK0089.

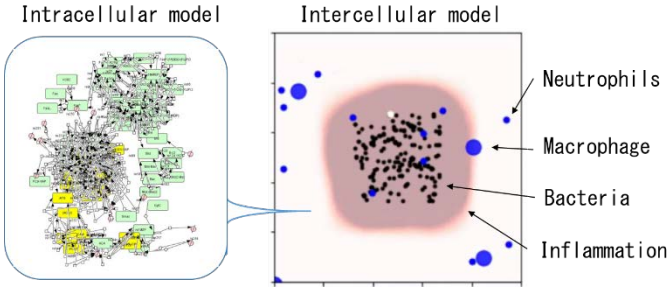

Fig. 1 Conceptual illustration of the present modeling.

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[2] T. Murakami, Scientific Reports, **9**, 17138 (2019).

Numerical modeling of the chemical effects of plasma-induced reactive species on air-saturated saline solution

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Cold atmospheric plasmas (CAPs) have been widely studied in the fields of biomedicine or agriculture. In these applications, plasma-activated liquids are gaining increasing attention because they can produce abundant reactive species, whereas plasma-induced aqueous chemistry is complex and its mechanism is not fully understood.

In this study, we propose a one-dimensional numerical simulation model to clarify the effect of inflow of various plasma-induced reactive oxygen / nitrogen species on the chemistry of air-saturated saline solution. The chemical model includes over 500 reaction processes to explain the reaction of hydrogen, oxygen, nitrogen and chlorine with carbonic acid equilibrium. Figure 1 shows a network diagram that visualizes the chemistry of air-saturated saline solution. Graph-theoretical analysis [1] shows the chemical species at the center of the network, e.g. OH, O_2, H^+, OH^-, O_2^- and Cl^- act as hubs for the reaction network. Detailed numerical simulations examine how plasma-induced reactive species permeate liquids and change their chemistry.

This work was partially supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (C) JP19K03813 and Fostering Joint International Research (B) JP20KK0089.

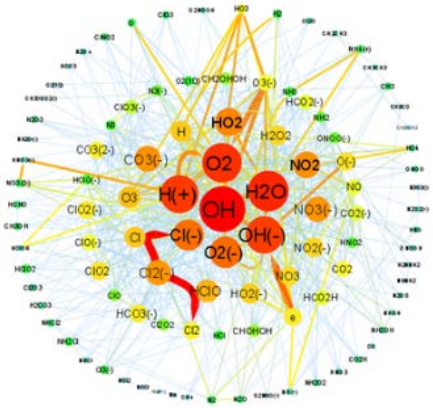

Fig. 1 Visualization of chemical networks as preprocessing of numerical simulations.

[1] T. Murakami and O. Sakai, Plasma Sources Sci. Technol. **29**, 115018 (2020).

Cavity ring-down spectroscopy of the spatial distribution of species in a cold atmospheric pressure plasma jet

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The application of cold atmospheric pressure plasma jets (CAPJs) for the localized treatment of heat-sensitive surfaces and biological tissue has gained high industrial and medical importance. A detailed understanding of the complex chemical reaction network confined to a CAPJ would enable customized compositions of reactive species to be tailored for a specific plasma application. This requires highly sensitive and selective measurements of transient molecules and free radicals, their spatial and temporal distributions, and their transport in non-equilibrium environments. However, some of the key radical species are often present only in trace amounts and for a short period of time, although they often drive the chemistry and therefore their detection is crucial. A challenge for diagnostics of CAPJs is the small dimensions (µm to cm range) over which the reactive species are confined. A common approach to obtain absolute species densities are absorption spectroscopy techniques. To increase the absorption path length, cavity-enhanced spectroscopy methods can be applied. However, with these techniques often line-of-sight densities without any spatial information are obtained. Nevertheless, using continuous-wave cavity ring-down spectroscopy, we were able to determine the spatial distribution of the HO_2 radical [1, 2] and the H_2O_2 molecule in the effluent of the plasma jet CAPJ device kINPen (see Fig. 1), developed at the INP for use in plasma medicine.

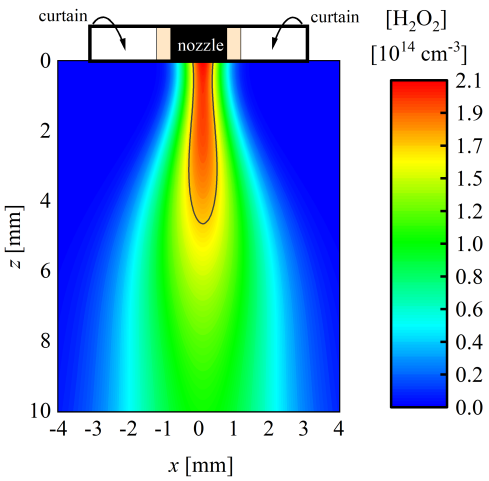

Fig. 1 A lateral contour plot of H_2O_2 densities in the effluent of the kINPen plasma jet.

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Optical diagnostics of micro-thrusters: spatial and temporal behaviour of the plasma

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In this presentation, the research on temporal evolution and spatial distribution of plasma parameters in two micro-thrusters is presented. The presentation consists of two parts. In the first part, temporally-resolved optical diagnostics is performed on a micro-cathode arc thruster (μCAT). A spectra acquisition system is used to obtain the 1-us-resolved emission spectra of the μCAT along a discharge pulse. The system consists of an ICCD detector, a Czerny–Turner spectrometer, and a high-precision high-synchronization signal source. Temporally resolved electron temperature and electron density are determined from emission spectra based on a collisional-radiative model of Ti^{+} and Ti^{2+}. The influence of operating conditions on these parameters has also been studied. In the second part, a miniaturized electron cyclotron resonance ion thruster (ECRIT) is studied using spatially-resolved optical diagnostic method. Emission spectra along the radial direction of the discharge chamber of the ECRIT are obtained, and electron density is determined by using a comprehensive collisional-radiative model. The influence of gas flow rate and microwave power is also studied. Finally, the prospect of spatiotemporal resolved optical diagnostics and its application in micro-plasma research are discussed.

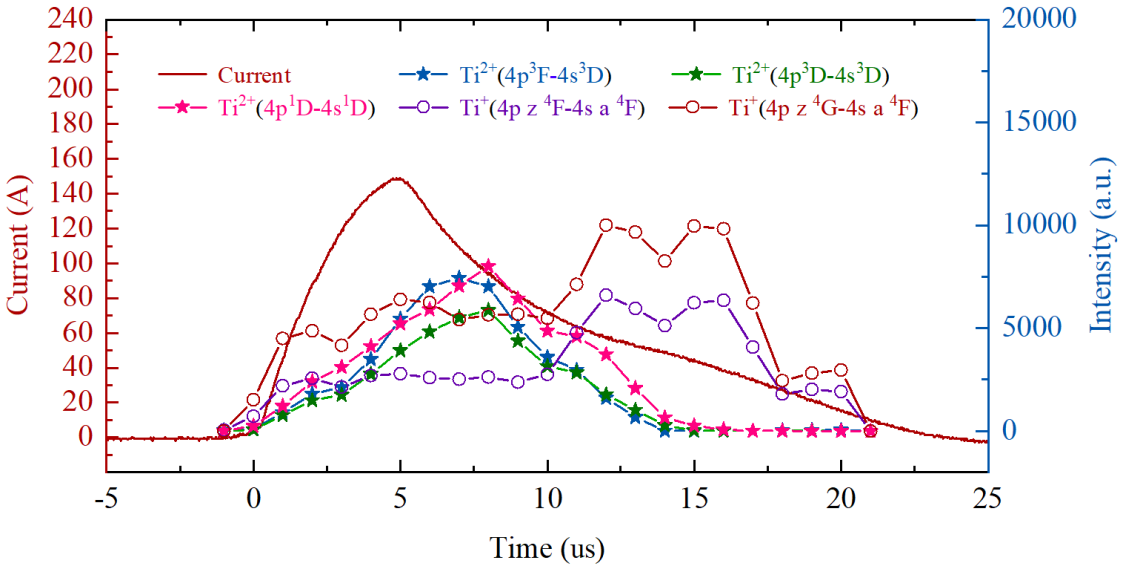

Fig. 1 Time evolution of discharge current and emission intensities of Ti^{+} and Ti^{2+}.

[1] Ji T Y, Wei L Q *et al* Influence of pulse frequency on discharge characteristics of micro-cathode arc thruster, Vacuum **196** 110748 (2022).

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Dielectric barrier discharge at low voltage using carbon nanotube electrodes in high density nitrogen including supercritical fluids and liquids

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In recent years, many studies have been conducted on microplasmas in high-density media including supercritical fluids and liquids. Our group previously reported that the use of carbon nanotubes (CNTs) as discharge electrodes enables discharges at relatively low voltages in such dense media [1]. In nitrogen, the color of the emission changed significantly with fluid density (high-density: green, low-density: violet) and the intensity of the emission became stronger with the density increasing. This color change suggests that the reaction system in the discharge changes with the density. However, many aspects of the discharge mechanism with CNTs, including this color change, have not been revealed yet.

In this study, surface dielectric barrier discharges (DBDs) using CNTs as discharge electrodes in high-density nitrogen were evaluated by photographic and spectroscopic measurements. The experiments were performed in gases, supercritical fluids, and liquids: at temperatures ranging from 80 to 200 K and pressures from 0.1 to 7.0 MPa.

Figure 1 shows the dependence of emission color on temperature, pressure, and density. The violet-green transition was not clear, and a transition regime of coexisting violet-green emission existed. The most important parameter that determines the transition conditions is presumably density. Unlike the previous study [1], green emission was observed even in gas. In the presentation, we will report details of the discharge phenomenon at low voltages with the DBD electrode using CNTs, including this color change.

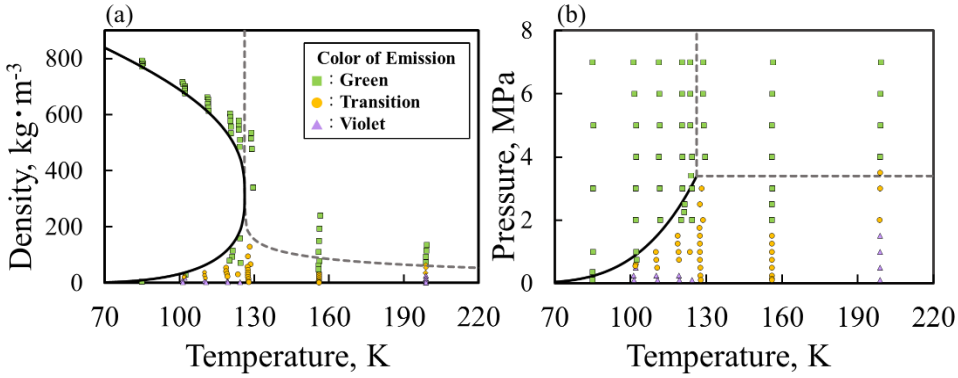

Figure 1. Experimental conditions and emission colors plotted on (a) Temperature-density and (b) Temperature-Pressure diagrams of N_2

[1] Hitoshi Muneoka *et al*, Plasma Sources Sci. Technol. **28**, 075014 (2019).

Laser Spectroscopy of Plasmas

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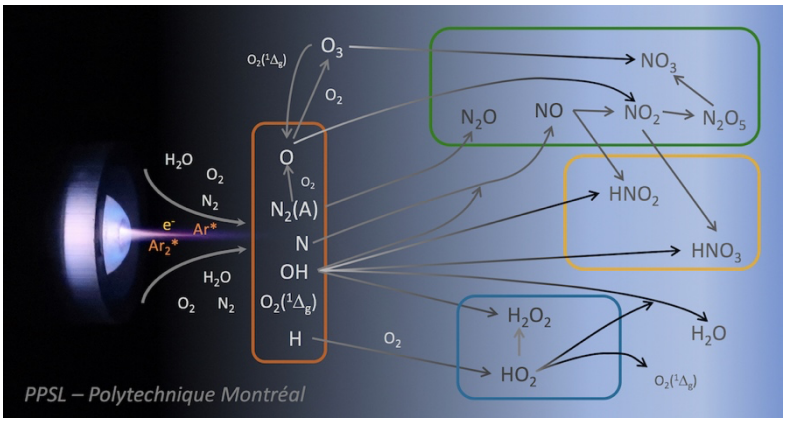

Fig. 1: cold plasma jets require advanced diagnostics to identify their complex reaction chemistry

Within the past decade micro-plasma jets in contact with liquids have been the focus of international research. They have shown great potential in applications ranging from surface treatment to medicine. To be able to control these jets for precise application, a fundamental understanding of the underlying processes is required. For this, detailed diagnostics need to be performed, which are challenged by the plasma jet's high gradients, multiphase transport processes and interfaces of plasma and liquid or solid.

Most conventional plasma diagnostics fail in cases of non-equilibrium processes at atmospheric pressure. Ultrafast laser spectroscopy, however, permits the diagnostic of fundamental plasma properties such as reduced electric field or flow properties and gas composition at timescales much shorter than collisional processes.

The talk presents current development in the field of ultrafast laser diagnostics and the challenges that single shot measurements have. A compromise to gain information from single shot measurements and high signal to noise from averaging measurements can be gained from data post processing or advanced averaging methods.

Periodic forced flow in a nanosecond pulsed cold atmospheric pressure argon plasma jet

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Plasma-flow interaction in cold atmospheric-pressure plasma jets is of importance due to the direct influence on the reactive species production. It has been shown that the effect of plasma heating and/or ionic wind in the plasma plume is too weak to explain the observed flow modification due to the plasma [1]. This work is devoted to the study of the argon flow modification in a cold atmospheric pressure plasma jet driven by nanosecond high voltage (HV) pulses, from single to multiple HV shots applications [2]. A schlieren optical bench has been designed in order to visualize the argon flow downstream expansion in quiescent air, for moderate flow rates below 1 standard liter per minute. A coupled approach is used between charge coupled device (CCD) schlieren imaging and intensified CCD (ICCD) plasma plume imaging, both time-resolved. It is shown that the application of only one HV pulse (i.e. single HV shot) is enough to disturb the flow, as predicted by a numerical model in helium [3]. The disturbed flow exhibits ripple propagation, on a timescale similar to the flow velocity. When operating in double HV shots, the second ionization wave can be used as a probe, to instantly visualize the flow structure any time after the first HV pulse application. For some flow rates, the ripple can increase in amplitude up to the point when it strongly deforms, or even stops, the plasma plume expansion, after which it is entrained by the flow and the plasma plume retrieves its full usual expansion. When a series of HV pulses are applied, the maximal disturbance of the flow is achieved for a certain pulse repetition frequency (PRF), specific of each flow rate. It is associated with ripples alternation in the plasma plume, in a 3D helical-like arrangement. For greater PRF, the ripples progressively vanish, and the flow is clearly less disturbed. Once the ripples have vanished, increasing further the PRF does not change the plasma plume and flow structures. We suggest that the repetitive plasma ignition mechanically forces the flow inside the capillary with consequences on the global flow structure, similarly to a forced backward-facing step flow with actuator [4].

[1] E.R.W. Van Doremaele *et al.*, Plasma Sources Sci. Technol. **27**, 095006 (2018).
[2] T. Darny *et al.*, Plasma Sources Sci. Technol. **30**, 105021 (2021).
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[4] N. Benard *et al.*, Int. J. Heat Fluid Flow **61**, 158 (2016).

Poster Abstracts
Poster Session June 7th, 2022

Simulations of atmospheric-pressure micro-plasma jets flows using SPARK

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Atmospheric-pressure micro-plasma jets are complex systems that exhibit coherent structures generated by discharges [1], and where admixtures through advection or diffusion can affect the plasma dynamics. To model these systems, a high-fidelity on the flow's advection-diffusion-reactions mechanisms is required. In this work, we follow this path by adapting the Software Platform for Aerothermodynamics Radiation and Kinetics (SPARK) [2] to the low temperature and low-speed flow conditions observed in APMPJs. We lowered the high numerical dissipation by including the multispecies version of the Simple Low-dissipation AUSM solver (SLAU) [3]. An efficient reduced argon kinetic scheme was included by uniform grouping a more complete scheme, and run times were further decreased by including 5th order WENO [4] variable reconstruction and OpenMP parallelization. Perfect-gas simulations of an argon APMPJ (see Figure 1) revealed a transition, with increasing flow, from short to long stable laminar jets, to unstable laminar jets, qualitatively similar to what is observed in Schlieren imaging experiments [1] (see Figure 1). Different excitation sources are tested including electron energy source terms and direct argon metastable sources.

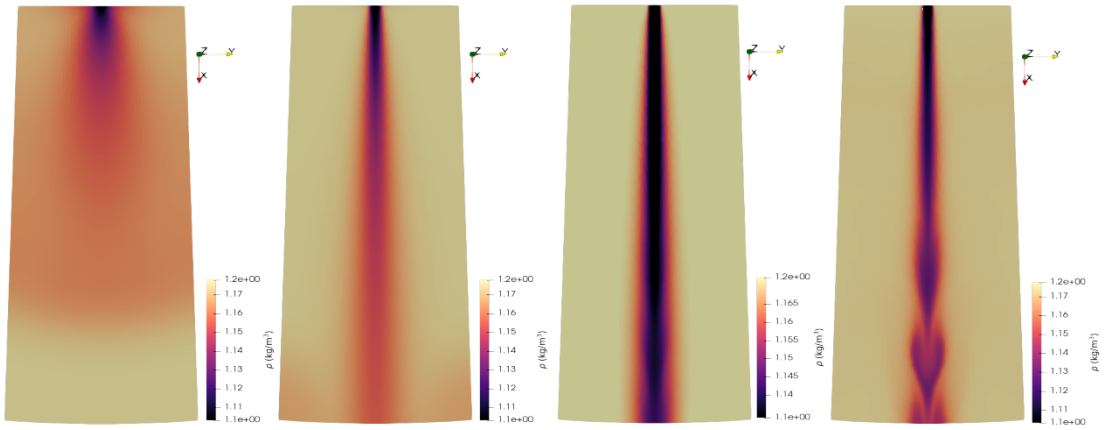

Figure 1. Mass density profiles of perfect-gas simulations at 50, 300, 700, and 900 sccm (from left to right).

This work was partially supported by the Portuguese FCT UIDB/50010/2020 and UIDP/50010/2020, and grant PD/BD/142972/2018 (PD-F APPLAuSE).

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Numerical modeling on apoptosis induction by low-temperature plasma

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Cell death induction is one of the important subjects in the medical application research of low-temperature plasma. For a better understanding of how plasmas exert biological effects, we have developed a zero-dimensional intracellular biochemical reaction model quantifying the influence of reactive oxygen and nitrogen species (RONS) on the mitochondrial functions [1]. In this study, we develop a numerical model of physiochemical reaction that describes an apoptosis induction system, and perform a numerical simulation to clarify the effects of RONS generated by low-temperature plasma.

Figure 1 shows a conceptual diagram of intracellular reaction model. which involves activation of death-receptors, mitochondrial signaling cascade pathway including the disruption of mitochondrial membrane potential, initiator- and executor-caspases and regulators. The effects of CAP irradiation can be modelled as exogenous or endogenous influence. The biochemical system is kinetically solved through a time-dependent 0D numerical simulation involving about 100 agents and about 300 reactions.

This work was partially supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (C) JP19K03813 and Fostering Joint International Research (B) JP20KK0089.

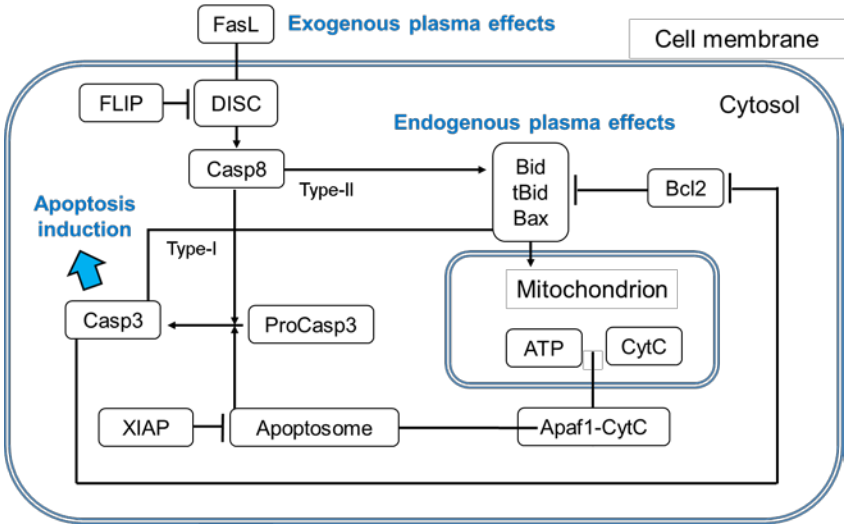

Fig. 1 Conceptual illustration of essential pathways for apoptosis induction.

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Surface-Launched Plasma Bullets

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Engemann's He-APPJ formed by the propagation of plasma bullets is generally generated by applying a high voltage AC voltage to a part of the body of a glass tube in which helium gas is filled [1]. On the other hand, we have found that an APPJ can be effused from the surface of a glass plate in contact with helium gas when we apply high-voltage pulse on the other side of the glass plate, as shown in Fig. 1. Imaging with an ICCD camera revealed that the APPJ in this case was formed by plasma bullets which were launched from the surface of the glass plate and propagating along the helium flux (opposite to the flux). There are many reports on the plasma-bullet propagation [2], and also on the plasma-bullet transfer (as if plasma bullets pass through the dielectric wall) [3]. On the other hand, as far as we know, there were no reports of surface-launched plasma bullets. We investigated voltage-rise-rate dependence of this phenomenon and found that 30 MV/s or faster rise rate is necessary for launching plasma bullets from the surface of the glass plate. Details will be presented and discussed in the workshop, together with application of this phenomenon to hydrophilic treatments of the inner surfaces of a bone-regeneration scaffold (a continuous porous dielectric material).

Acknowledgements: This work has been supported by the KAKENHI (Grant No. H19H01888, K19K03811, 20K20913) and Center for Low-Temperature Plasma Sciences (cLPS), Nagoya University.

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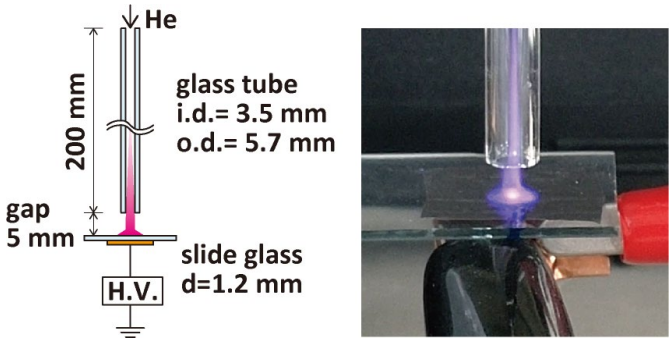

Fig. 1 Schematic illustration of the experimental setup for launching plasma bullets from the surface of a glass plate, and overview of the consequent APPJ.

Maze-Solving Visualized by Log-Path Microchannel Plasmas and Reproduced in Reinforce-Learning Model

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Maze-solving is a task which has attracted for a long time and still currently attracts scientific attentions since it is a typical question with NP (Non-deterministic Polynomial-time) hardness [1]. Recently, we reported an experimental method for searching for a classical two-dimensional maze path; configuring a trench network with the similar spatial pattern to the maze, generation of long-path microchannel plasmas visualizes the true path in a maze pattern [2]. In this study, we accumulate acquired data of successful maze-solving in plasma experiments, and compare them with predictions by a reinforcement learning method. Reinforcement learning [3] is an automatic algorithmic approach in which an optimized solution can be derived by taking into account of *rewards* for a virtual agent in varying environments.

Figure 1 shows a solved path by microchannel Ar plasma using a high-frequency (20-kHz) power supply. Between two discharge electrodes, a visible discharge path successfully forms spontaneously. We also confirmed validity of our reinforcement learning model, which simulates hypothetical potential profiles similar to those in this experimental setup.

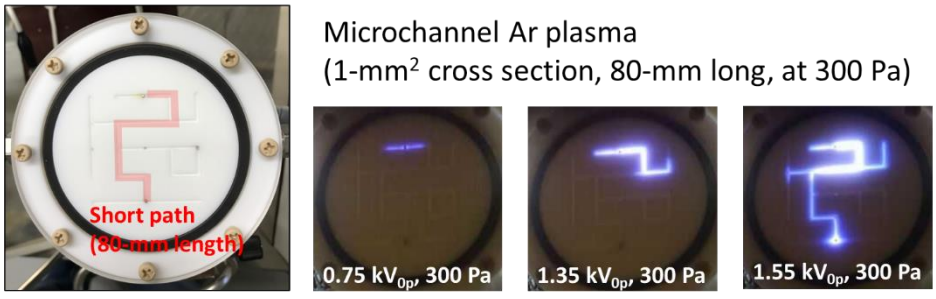

Fig. 1 Visualization of solved path by visible plasma emission.

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Complex network analysis of low-temperature plasma chemistry

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The importance of plasma chemistry is reaffirmed as low temperature plasma research extends beyond physics to biology and medicine. In the field of numerical research, we consider both expanding the target chemical reaction system as much as possible to construct a huge integrated reaction system and reducing it as much as possible to construct the most efficient minimum reaction system. Identifying the unique properties of the reaction system is important for the scale transformation. Therefore, in this research, we analyze the plasma chemical system based on the complex network theory [1,2] and perform scale conversion of the reaction system.

Figure 2 shows a network diagram that visualizes the chemical reaction structure of the He + O_2 plasma [3]. Chemical species in the central part of the network, such as electrons, atomic oxygen, ozone, He, and O_2, are involved in many reaction processes. The thick directed line connecting them indicates that the reaction probability (reaction coefficient) is relatively high.

This work was partially supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (C) JP19K03813 and Fostering Joint International Research (B) JP20KK0089.

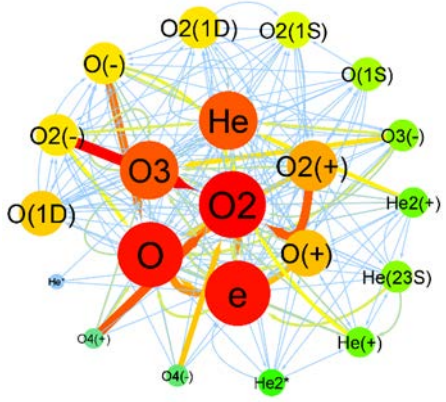

Fig. 1 Network diagram of He+O_2 plasma chemistry

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Influence of Submerged Bubble Proximity on Electric Field in Water

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Gas bubbles in water have been a target of extensive study for plasma discharges in liquid environments. The mechanism of discharge initiation in liquid environments remains an arena of contest hypotheses from the electrostrictive cavitation, in which nanoscopic voids are formed; to rapid vaporization points in which the liquid phase must initially undergo phase change before initiating discharge. Ansys Maxwell simulation models were constructed around electrode/water/bubble configurations imaged experimentally to investigate the influence macroscopic bubbles have on the local field enhancements near the electrodes. Sharp and flat electrode geometries were modeled, and macrobubble positions were altered in relation to the position of the electrodes. Simulated electric fields suggest that gas bubbles enhance the field directly at the electrode tip, but a critical liquid gap is required before the field enhancement becomes dominant at the liquid/gas boundary interface.

Numerical modeling on the dynamic behavior of immune cells

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In recent medical application research of low-temperature atmospheric pressure plasma, the immune response activation effect has been attracting attention [1,2]. While many experimental studies on immune response activation using plasma have been conducted, few mathematical approaches have been attempted. Here, we develop a time-dependent two-dimensional mathematical model to simulate the most basic functions of immune cells, i.e. migration and phagocytosis. As a primary stage, this model describes the dynamic behavior of leukocyte bactericidal action, in which neutrophils and macrophages play important roles as innate immunity (cell-mediated immunity) agents.

Figure 1 shows a conceptual illustration of the current model. The neutrophils and macrophages autonomously find the affected area affected by bacteria, moves toward it, and exhibits phagocytic action. These collective behaviors are influenced by the presence or absence of plasma irradiation.

This work was partially supported by JSPS KAKENHI Grant-in-Aid for Scientific Research (C) JP19K03813 and Fostering Joint International Research (B) JP20KK0089.

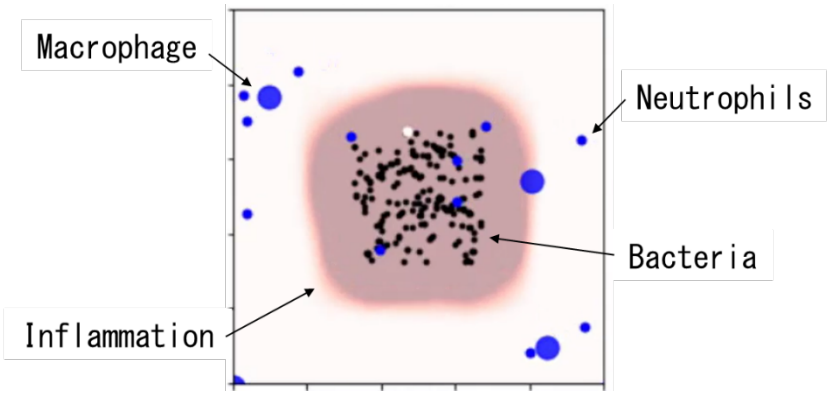

Fig. 1 Conceptual illustration of the present modeling.

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Investigating the Effect of Low Temperature Atmospheric Pressure Plasma Jets on the Cold Hardiness of Sweet Basil

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Plasma treatment in agriculture is a new and rapidly growing field. Low temperature atmospheric plasma jets specifically have been proven to be a promising method of treatment for both seeds and plants. We know that when plants are introduced to abiotic stressors, the resiliency of the plant is increased. With this in mind, a cold plasma jet presents a new form of stress for plants [2]. When applied to sweet basil seeds, they have been shown to increase the physical growth of seedlings. When the plants were treated throughout the growth cycle, the yield of essential oils was increased, as the plants grew larger and more robust [1]. Crop resilience to stressors been studied in the plasma agriculture field, considering only the cold plasma as the stressor. The field is lacking in studies addressing the plants resilience to factors such as cold, heat, or drought. This study seeks to understand if sweet basil plants treated by a plasma jet have an affected temperature hardiness or resiliency. The plants will be grown in a hydroponic loop, ensuring all plants experience the same conditions. The plants will be exposed to stressful, cold water, and plasma treated group will be studied against the control group.

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Thermocatalytic Plasma-Assisted Dry Reforming of Methane Over Heterogeneous Ni/Al_2O_3 Catalyst.

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Plasma catalysis is a relatively new hybrid technique in which a catalytic material is used alongside a gas discharge to yield an enhanced performance in gas processing applications. A typical example of such application is the removal of pollutants and volatile compounds, as well as the production of a range of chemicals by the reforming of hydrocarbons. Another very promising area of plasma-assisted catalysis is the dry methane reforming to selectively generate syngas. The complex endothermic process requires the symbiotic use of CH_4 and CO_2, which mitigates the environmental impact of both greenhouse gases by transforming it into valuable syngas, which may be further used to produce valuable petrochemicals. Furthermore, unlike wet methane reforming, dry reforming of methane provides a higher concentration of active species in the produced syngas as compared to other reforming processes. However, because of the high carbon content in the feedstocks and harsh industrial reaction conditions, the dry reforming of methane is usually much more prone to coke formation, making the catalyst deactivation a serious problem for larger scale industrial application. The process has thus been extensively studied in recent years with the intention of creating more coke-resistant catalysts.

The current presentation describes the results of the plasma-assisted dry reforming of methane carried out over supported Ni/Al_2O_3 catalyst in a fixed bed reactor system. The reaction parameters studied were the catalyst bed temperature, the CO_2 to CH_4 ratio, the methane space velocity, the plasma supply voltage and frequency. The resulting methane conversion and hydrogen selectivity was monitored by an on-line GC analysis using a dedicated Varian 4900 system.

In these initial studies, each experiment was carried out in two distinct regimes using the same charge of catalyst. According to the established protocol, the first stage of each run was started in "No-Plasma" regime to record the baseline process performance. In the second stage called "Plasma-On" regime, the plasma was initiated, and the power supply voltage was varied stepwise in a pre-programmed mode. Under the same process conditions, the reaction was repeated four times to determine the degree of catalyst deactivation from run to run.

The results of these initial studies show that by the time of the fourth experiment, a near-complete deactivation of the catalyst was recorded for the first "No-Plasma" regime. However, in all four consecutive runs, the catalyst, after being placed in a highly deactivated state from the "No-Plasma" regime, showed rapid recovery in the second "Plasma-On" stage. While there are several possible explanations for the unexpected in-situ reactivation of the catalyst, the finding justifies further studies using already deactivated Ni/Al_2O_3 catalyst to determine the most efficient plasma-assisted reactivation protocol.