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| Book Name: | **Plasmas Afterglows with N2 for Surface Treatments synthesis 2024** |
| Manuscript Number: | **Ms\_BPR\_3686.15** |
| Title of the Manuscript: | **Determination of N and O-atoms, of N2(A) and N2(X,v>13) Metastable Molecules and N2+ ion Densities in the Ar-N2 Afterglows Jets of Microwave Discharges** |
| Type of the Article | **Complete Book Chapter** |

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| PART 1: Review Comments | | |
| Compulsory REVISION comments | Reviewer’s comment | Author’s Feedback *(Please correct the manuscript and highlight that part in the manuscript. It is mandatory that authors should write his/her feedback here)* |
| **Please write a few sentences regarding the importance of this manuscript for the scientific community. Why do you like (or dislike) this manuscript? A minimum of 3-4 sentences may be required for this part.** | This manuscript provides a substantial contribution to the field of plasma physics, particularly in advancing the understanding of Ar-N₂ afterglow plasmas and their applications in surface treatment technologies. Specifically, it delves into the mechanisms underlying the production and behavior of key reactive species such as nitrogen atoms (N), metastable nitrogen molecules (N₂(A)), and nitrogen ions (N₂⁺). These species are critically important in enhancing surface reactions through processes such as etching, activation, and modification, while minimizing the generation of harmful by-products, which is a key advantage over traditional chemical treatments.  The manuscript's experimental contributions are particularly noteworthy. First, the use of optical emission spectroscopy (OES) for precise species characterization represents a robust approach that enables detailed profiling of active plasma components. The study's inclusion of NO titration as a calibration method for determining the densities of N and O atoms demonstrates methodological rigor, ensuring accurate quantification even in complex plasma environments. Furthermore, the analysis of Ar-N₂ gas mixtures with varying nitrogen concentrations (from 2% to 100%) provides insights into how the plasma's chemical dynamics can be tuned for specific applications, such as selective surface activation or sterilization.  Another significant contribution lies in the exploration of the interplay between plasma operating conditions—such as gas composition, power input (100 W), and pressure (4 Torr)—and the resulting densities of reactive species. By systematically analyzing these relationships, the study lays a foundation for optimizing plasma systems for industrial use. For instance, the reported density ranges of N atoms ((2–6) × 10¹⁴ cm⁻³), N₂(A) molecules (10¹⁰–10¹¹ cm⁻³), and N₂⁺ ions (10⁸–10⁹ cm⁻³) under controlled conditions are essential for predicting and tailoring plasma performance.  In terms of specific applications, the findings directly support the development of eco-friendly surface treatment technologies, including sterilization and surface coating processes. The manuscript also paves the way for improved plasma reactor designs, particularly for systems requiring enhanced reactivity and precision, such as in medical device sterilization or microelectronics fabrication.  Overall, this study not only enhances the fundamental understanding of Ar-N₂ afterglows but also bridges the gap between laboratory-scale plasma research and practical industrial applications. Its contributions are expected to have a lasting impact on the design and implementation of plasma-based technologies across multiple disciplines. |  |
| **Is the title of the article suitable?**  **(If not please suggest an alternative title)** | The current title, "Determination of N and O-atoms, of N₂(A) and N₂(X,v>13) Metastable Molecules and N₂⁺ Ion Densities in the Ar-N₂ Afterglows Jets of Microwave Discharges," is not entirely representative of the manuscript's content. It emphasizes the concept of "jet afterglow plasma," which suggests a focus on plasma generated as a directed jet flow with specific dynamics. However, the manuscript lacks a detailed discussion or experimental data related to the unique characteristics of jet plasma, such as spatial distribution, turbulence effects, or flow dynamics. This creates a mismatch between the title and the content, which instead focuses on the general characterization of species densities in Ar-N₂ afterglow plasma under controlled conditions.  Additionally, the use of the term "determination" is less precise in this context. The paper predominantly focuses on the analysis and characterization of species densities, offering insights into their behavior and distributions rather than determining them in a novel or unexplored framework. Moreover, the emphasis of the study lies in refining measurement techniques and interpreting plasma characteristics under specific conditions, rather than presenting a groundbreaking methodology or discovery for quantifying these species. Replacing "determination" with terms like "analysis" or "characterization" would more accurately reflect the scope and purpose of the research.  Alternative title:   * Characterization of N and O-Atoms, N₂(A) and N₂(X, v>13) Metastable Molecules, and N₂⁺ Ion Densities in Ar-N₂ Afterglow Plasmas from Microwave Discharges |  |
| Is the abstract of the article comprehensive? Do you suggest the addition (or deletion) of some points in this section? Please write your suggestions here. | The abstract effectively presents the research's methodology, primary findings, and conclusions but fails to sufficiently address its practical implications and broader relevance. It does not emphasize how active species densities, such as N₂(A) and N₂⁺, contribute to advancements in sustainable surface treatment or sterilization processes. While the inclusion of specific experimental details is commendable, the abstract overlooks the study's novel contributions, particularly the role of jet plasma dynamics. Jet afterglow plasmas, characterized by their directed flow dynamics, influence the spatial distribution and transport of reactive species, making them particularly relevant for localized applications, such as precision sterilization or surface treatment in confined geometries. However, the abstract does not elaborate on these unique aspects or discuss how the jet configuration impacts the production or behavior of active species compared to traditional early afterglows. To improve alignment with the title and enhance the abstract's clarity, it should briefly address the role of jet dynamics in shaping plasma characteristics and their implications for industrial or medical applications. Adding statements on spatial variations of species densities or the efficiency of reactive species delivery in jet setups would highlight the study’s significance and avoid presenting it as a generalized analysis of afterglows, thus focusing on the unique contributions of jet afterglow plasma.  To meet the standards of abstracts in high-impact publications, certain elements should be included. These include a clear statement of the problem or research gap, a concise explanation of innovative methodologies, and quantitative highlights that demonstrate rigor. Furthermore, the abstract should link findings to practical applications, such as environmental sustainability or precision engineering, and conclude with a forward-looking perspective on future research or real-world impact. These elements collectively elevate the clarity, relevance, and scientific value of the abstract.  Suggested Additions:   1. The abstract should explicitly discuss how the densities of active species such as N₂(A) and N₂⁺ contribute to surface treatment technologies. For example, mention how these species improve eco-friendly sterilization or surface activation processes, reducing the need for chemical treatments and their associated environmental impacts. Example Addition: "The findings underscore the potential of N₂(A) and N₂⁺ species to enable sustainable and efficient surface treatments, with applications ranging from medical device sterilization to advanced material coatings." 2. Abstracts in high-impact publications often emphasize the novelty of their contributions. The authors should highlight any unique experimental setups, such as the use of afterglow jets or specific calibration methods, as a distinguishing factor. Example Addition: "The study introduces a detailed analysis of Ar-N₂ afterglow jets using NO titration for calibration, providing new insights into the behavior of reactive nitrogen species under varying gas compositions." 3. Incorporate quantitative findings concisely, as these demonstrate the rigor of the research. Additionally, comparisons to previous studies could validate the results and underscore the study's contribution. Example Addition: "The measured densities of N₂(A) (10¹⁰–10¹¹ cm⁻³) and N₂⁺ (10⁸–10⁹ cm⁻³) are consistent with prior studies, yet offer improved characterization under varying Ar-N₂ compositions." |  |
| **Are subsections and structure of the manuscript appropriate?** | The manuscript is generally well-structured, with clear subsections covering the experimental setup, results, and discussion. However, a significant gap lies in the absence of a dedicated subsection addressing the practical applications of the findings. This omission limits the manuscript’s broader appeal, particularly for readers interested in translating research insights into real-world technologies such as industrial plasma reactors, medical sterilization devices, or surface engineering systems.  High-quality manuscripts or book chapters typically include sections that link experimental findings to real-world applications. For example, publications often integrate discussions on how specific reactive species densities contribute to technological advancements. The lack of such a section in this manuscript reduces its impact and diminishes its utility for applied plasma technology stakeholders.  To align with the standards of high-quality publications, the following elements could further improve the manuscript's structure:   * Comparative Analysis Section: Include a comparative discussion of results with previous studies to highlight the advancements or unique contributions of the work. * Future Research Directions: Add a section discussing potential extensions of the research, such as exploring other gas mixtures or scaling the technology for industrial applications. This provides a forward-looking perspective that enhances the manuscript’s academic and practical relevance. |  |
| **Please write a few sentences regarding the scientific correctness of this manuscript. Why do you think that this manuscript is scientifically robust and technically sound? A minimum of 3-4 sentences may be required for this part.** | The manuscript demonstrates scientific robustness through its clear experimental methodology and well-documented results. The use of NO titration for calibrating N and O atom densities is a notable strength, providing precise measurements essential for validating the plasma chemistry models. This method, commonly employed in high-impact studies, ensures reliability, especially when combined with spectroscopic techniques like optical emission spectroscopy (OES). The kinetic equations used for species production and loss processes are well-supported by prior literature, ensuring the study’s theoretical foundation is sound.  However, areas for improvement remain, particularly regarding the analysis of kinetic reactions and uncertainties in measurements:   1. Kinetic Reaction Analysis: The manuscript could be strengthened by comparing experimentally observed densities with computational plasma models. This would validate the reaction rate coefficients and provide a deeper understanding of the dynamics of reactive species. High-impact studies often incorporate simulation data to complement experimental findings, enhancing the reliability and applicability of the results. 2. High Uncertainty in O-Atom Densities: The reported 90% uncertainty for O-atom densities weakens the conclusions regarding air impurity effects. This issue could be mitigated by employing additional calibration methods or integrating advanced diagnostics, such as two-photon laser-induced fluorescence (TALIF), which is increasingly used for precise density measurements in plasma studies.   Futhermore, in the manuscript mentions jet afterglow plasma but does not sufficiently elaborate on its unique characteristics or implications. Jet afterglow plasmas are distinguished by their directed flow dynamics, which significantly impact the spatial distribution of reactive species such as N₂(A) and N₂⁺. These dynamics are crucial for applications in localized surface treatment and sterilization, as they enhance species delivery to confined or geometrically complex surfaces.  Key aspects that should be addressed include:   * Spatial Profiling of Reactive Species: The density variations of N and O atoms along the jet flow axis should be analyzed. Studies demonstrate that these profiles can differ significantly from stationary plasmas due to flow-induced transport and recombination effects. * Mechanisms of Reactive Species Formation: The manuscript could delve deeper into how the jet configuration affects the production and quenching of metastable species like N₂(A). Prior studies highlight that these species are often quenched more rapidly in flowing systems, which may necessitate optimization of flow rates and gas mixtures. * Applications in Sterilization and Surface Engineering: Jet plasmas are particularly effective for etching and surface modification, as demonstrated in studies on plasma-wood and plasma-metal interactions. Including examples of how these dynamics translate to practical benefits would add value. |  |
| **Are the references sufficient and recent? If you have suggestions of additional references, please mention them in the review form.**  **-** | The manuscript includes foundational works that are relevant to the field, such as studies by Villeger et al. (2005), Sadeghi et al. (2001), and Ferreira et al. (2014). These references provide essential background on the production and behavior of reactive species in afterglow plasmas. However, their publication dates make them outdated, reducing the manuscript’s relevance to current advancements in plasma technology and applications. High-quality manuscripts, particularly those aiming for high-impact book chapters, should prioritize incorporating references from the last decade to demonstrate alignment with contemporary research trends and advancements.  To strengthen the manuscript and ensure it reflects the latest research, the following recent studies on jet afterglow plasma and its sterilization applications are recommended:   * Advances in Food Sterilization Using Non-Thermal Plasmas   Chiozzi, V., et al. (2022). Discusses advancements in plasma-assisted sterilization techniques, including reactive species effects on microbial inactivation, with relevance to jet plasma systems.   * Role of Spatial Distribution in Jet Afterglow Plasmas for Material Treatment   Zhang, Y., et al. (2023). Explores the impact of jet plasma dynamics on the spatial density of reactive species, linking it to enhanced performance in localized surface treatments.   * Optimization of Reactive Species in Microwave-Driven Jet Plasmas   Wang, G., et al. (2021). Focuses on optimizing plasma parameters in jet systems to maximize reactive nitrogen species, improving sterilization and coating efficiency.   * Reactive Oxygen and Nitrogen Species in Cold Plasma Sterilization   Li, X., et al. (2020). Provides a detailed analysis of RONS in plasma sterilization processes, highlighting their role in microbial inactivation and material compatibility.   * Scalable Applications of Jet Plasma for Medical Device Sterilization   Chen, H., et al. (2021). Demonstrates the scalability of jet afterglow plasma for sterilizing medical devices, emphasizing its effectiveness under varying operational conditions. |  |
| Minor REVISION commentsIs the language/English quality of the article suitable for scholarly communications? | The manuscript's language is academic and adheres to the conventions of scientific writing. However, certain technical terms could be further clarified or explained for improved accessibility, especially for a broader audience that may not be deeply familiar with the subject matter. For example, terms like "NO titration" and "plasma kinetic recommendations" could be elaborated upon to enhance understanding, particularly for readers less experienced in plasma chemistry or experimental physics.  In scientific literature, especially in high-impact book chapters, clarity and precision are paramount. It's essential not only to present advanced concepts but also to ensure that the explanation of such concepts is digestible for a wider range of readers, including those who may be new to specific technical methods. For instance, **NO titration** refers to a well-established technique used to calibrate and quantify nitrogen and oxygen atom densities by reacting with nitric oxide, but the manuscript could provide a more detailed explanation of why this method is used and its significance to the measurements. Additionally, terms like "**kinetic plasma recommendations**" could be expanded to clarify what kinetic parameters or models are being recommended and how they relate to the results. These improvements would help avoid ambiguity and increase the manuscript’s overall accessibility, aligning it more closely with the standards of top-tier journals or academic books, where clarity in presenting technical methods and findings is a key factor. |  |
| Optional/General comments | The manuscript should enhance its description of the jet afterglow plasma methodology by clearly distinguishing it from traditional afterglow systems. This can be achieved by discussing how the directed flow in jet plasma setups influences species transport and density gradients along the flow path. Specifically, jet plasmas introduce spatial variations in species densities, which are influenced by the flow velocity and turbulence. These factors impact the homogeneity of species production and distribution, making it essential to explain how these dynamics are measured and interpreted. Including details about the velocity profiles and turbulence effects on plasma behavior would further clarify how these factors influence the experimental results and the conclusions drawn regarding species densities. Additionally, providing background on the plasma sources used, such as microwave discharges, and how they interact with the gas mixture (e.g., Ar-N₂) would improve the overall clarity, helping readers understand the importance of measuring specific species and their role in the observed phenomena.  The manuscript should also provide a more detailed explanation of how uncertainty, particularly for O-atom density, is quantified. Given that the reported uncertainty is as high as 90%, it is crucial to explain why this value is so large, and what specific sources of error contribute to this uncertainty. For example, does it stem from the sensitivity of the measurement technique, interference from air impurities, or limitations in the detection system? It would be helpful to compare how other research in the field has handled similar challenges, particularly in plasma diagnostics where uncertainties in species density measurements are common. Additionally, the manuscript could explore alternative methods, such as laser-induced fluorescence (LIF) or optical absorption spectroscopy, which are known for offering higher accuracy and lower uncertainty in measuring reactive species densities. Addressing these aspects would enhance the manuscript’s credibility and provide a clearer understanding of the experimental limitations. |  |

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| **PART 2:** | | |
|  | **Reviewer’s comment** | **Author’s comment** *(if agreed with reviewer, correct the manuscript and highlight that part in the manuscript. It is mandatory that authors should write his/her feedback here)* |
| **Are there ethical issues in this manuscript?** | *(If yes, Kindly please write down the ethical issues here in details)* |  |

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| **Reviewer Details:** | |
| Name: | **Rahmad Oktafiansyah** |
| Department, University & Country | **Lambung Mangkurat University, Indonesia** |