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博士論文の要旨

氏　　名：中澤　淳一郎　NAKAZAWA, Junichiro

論文題目：**Mass Spectrometry Techniques for Sublimated Gas from Organic-Bearing Microparticles by Hypervelocity Impacts**

This dissertation aims to establish a new mass spectrometric approach that suppresses molecular fragmentation for organic analysis in the outer Solar System bodies and icy satellites, by numerically and experimentally investigating the ionization and emission processes of neutral gas and plasma generated by hypervelocity impacts.

Conventional impact ionization dust mass spectrometers onboard spacecraft suffer from severe molecular fragmentation due to plasma interaction induced by hypervelocity impacts of microparticles, which makes reliable identification of molecular structures of organic compounds in the dust grains difficult. On the other hand, incidental measurements of impact-generated gas by neutral gas mass spectrometers in space have suggested that molecular fragmentation associated with dust impacts may be more moderate. However, molecular structure identification remains limited due to fragmentation induced by electron-impact ionization (EI) within neutral gas mass spectrometers, and the effectiveness of neutral gas analysis has not been experimentally validated.

In this dissertation, neutral gas generated by hypervelocity dust impacts is systematically analyzed to experimentally demonstrate that impact-induced molecular fragmentation can be significantly suppressed. Furthermore, mass spectrometric techniques that combine neutral gas analysis with less destructive ionization methods other than EI are evaluated to improve the accuracy of organic analysis in future Solar System exploration missions.

This research consists of two main pillars:
(1) understanding the ionization and sublimation processes of hypervelocity impact products, and
(2) analysis of neutral gas generated by dust impacts.
Through these approaches, neutral gas produced by impacts is treated as the primary analytical target, and the feasibility of suppressing molecular fragmentation is explored by redesigning the entire measurement architecture, including the ionization process.

(1) Understanding the Ionization and Sublimation Processes of Hypervelocity Impact Products

Direct experimental reproduction of ionization and emission phenomena associated with hypervelocity impacts is extremely challenging, as no existing ground facility can accelerate and impact micrometer-scale particles at velocities exceeding 20 km s^{-1}. Therefore, this study treats high-temperature and high-pressure fields generated by laser irradiation as an analogue of hypervelocity impacts and investigates the correspondence between the two using numerical simulations. To account for non-equilibrium ionization processes, a two-temperature radiation hydrodynamics code, P4P (PINOCO for Plasma Processing and Planetary Physics), which was originally developed for laser fusion research, was adapted. P4P solves the conservation equations of mass, momentum, and energy in combination with radiation transport and an equation of state for high-energy plasma, enabling the analysis of high-energy fluid behavior. However, QEOS has limitations in accurately describing transition regimes where solids are rapidly heated and partially ionized. To address this issue, a correction model was introduced to recalculate the electron chemical potential and electron number density, thereby improving the accuracy of electron statistics in partially ionized regions. As a result, we successfuly reproduced the relationship between impact-generated charge (Q) and impact velocity (v), as $Q \propto v^{4.8}$ based on experimental data reported by Ratcliff et al. (1997) and Burchell et al. (1999), with high fidelity.

Using the improved P4P model, the analogy between laser irradiation and hypervelocity impacts was further examined numerically. The correspondence between laser irradiation and hypervelocity impacts depends on the physical quantities used for comparison, such as crater diameter, shock pressure, generated charge, and entropy, and empirical scaling relations have traditionally been applied for individual parameters. In particular, Klenner et al. (2019) investigated similarities in mass spectra between impact-ionized plasma and laser ablation plasma; however, the physical basis for the reason why specific laser conditions reproduce spectra corresponding to specific impact velocity ranges still remained unclear. In this study, the experimental conditions of Klenner et al. (2019) were reproduced with the P4P model, and the temporal evolution of energy density, pressure, and temperature distributions was analyzed. The results demonstrate that specific pulsed-laser conditions can reproduce thermal energy states equivalent to hypervelocity impacts at approximately 5-15 km s^{-1}. This provides a theoretical foundation supporting laser irradiation experiments as an effective surrogate for reproducing plasma generation and ionization processes in hypervelocity impacts.

(2) Analysis of Neutral Gas Generated by Dust Impacts

Based on the findings of (1), laboratory-scale simulated impact experiments using laser ablation were conducted, and the feasibility of analyzing simulated impact-sublimated materials was examined by combining P4P simulations with mass spectrometric observations. Using polystyrene samples, mass spectra of sublimated gas were obtained via laser irradiation followed by electron-impact ionization. The observed spectra exhibited major peaks corresponding to styrene trimer fragments and low-molecular-weight alkyl groups. Compared with experimental results from impact ionization mass spectrometers summarized by Srama et al. (2009) using the same sample material, the degree of fragmentation was significantly reduced, demonstrating that impact-generated gas analysis enables relatively soft mass spectrometric measurements.

As a further extension, photoionization mass spectrometry was developed and evaluated as a non-destructive ionization method alternative to conventional EI. Using a VUV light source, PI and EI spectra of volatile organic molecules such as acetone, o-xylene, and ethanol were obtained. The results show that PI exhibits higher molecular ion retention than EI and provides superior structural preservation. Moreover, by controlling photon energy, molecular fragmentation can be further suppressed, demonstrating the applicability of PI to future spacecraft-borne mass spectrometers.

Based on these results, the numerical modeling and experimental findings are integrated to clarify the following two points:

(1) The ionization behavior of hypervelocity impact plasma was successfully reproduced using the P4P code, and the analogy between laser irradiation and hypervelocity impacts was quantitatively established.

(2) Electron-impact mass spectrometry of impact-sublimated gas enables molecular structure analysis while relatively preserving the integrity of the original molecules.

Furthermore, building on these fundamental insights, this study conceptually organizes two mass spectrometric approaches for future spacecraft implementation: Impact-generated gas Electron-Impact Ionization Mass Spectrometry (I-EI-MS) and Impact-generated gas Photo-Ionization Mass Spectrometry (I-PI-MS), and examines their respective feasibility and system-level requirements. I-EI-MS, based on existing neutral gas mass spectrometers such as Cassini INMS, enables relatively low-resource implementation by intentionally introducing and controlling impact-sublimated gas. In contrast, I-PI-MS is expected to further enhance molecular structure preservation by incorporating non-destructive photoionization, although the power consumption and space qualification of VUV light sources are identified as major design challenges. Building on this framework, future work will focus on integration with high-resolution mass spectrometers (e.g., MULTUM and Orbitrap) and experimental validation under low-temperature and low-pressure conditions, toward establishing spacecraft-compatible mass spectrometric techniques that control molecular fragmentation induced by hypervelocity impacts.

In conclusion, this dissertation establishes that mass spectrometric analysis using sublimated gas generated by hypervelocity impacts constitutes a practical and scalable strategy for organic analysis under hypervelocity impact conditions. Additionally, this work presents a systematic framework for organizing the trade-offs between analytical performance and spacecraft resource requirements within the new observational paradigm of impact-generated sublimated gas analysis. The outcomes of this study provide new guidelines for the design and calibration of spacecraft-borne mass spectrometers and serve as foundational knowledge contributing to higher-accuracy organic analysis in future planetary and astrobiological exploration missions.

（別紙 Separate Sheet）

Results of the Doctoral Thesis Defense

博士論文審査結果

Name in Full
氏　　名　中澤　淳一郎

Title
論 文 題 目　Mass Spectrometry Techniques for Sublimated Gas from Organic-Bearing Microparticles by Hypervelocity Impacts

Mr. Nakazawa's Ph.D. thesis aimed to establish a new in-situ mass spectrometry technique that significantly suppresses molecular fragmentation of organic components in neutral gas sublimated from micrometeoroids by hypervelocity impacts.　To achieve this goal, both numerical modeling originally developed from the P4P code and laser irradiation experiments were combined, resulting in the following outcomes:
(1) The ionization behavior of hypervelocity impact plasma was successfully reproduced, and the analogy between laser irradiation and hypervelocity impacts was quantitatively established.
(2) Electron-impact mass spectrometry of impact-sublimated neutral gas enabled molecular structure analysis while relatively preserving the integrity of the original molecules.　This study also proved that photo-ionization mass spectrometry for organic-bearing neutral gas would be even less destructive than conventional electron-ionization.　It defined a new guideline for the design and calibration of space-borne mass spectrometers, especially for higher-accuracy organic analysis in future planetary and astrobiological exploration missions.

All the viva examiners agreed that this work has a great academic value in the field of space science and engineering; it helps explain the physical processes behind impact ionization, which is an established technique in space missions to measure dust particle composition and has produced numerous science papers in Nature, etc., so finally having a sound model-based theory to explain those results is an important milestone for planetary science.

Mr. Nakazawa's originality lies in evaluating the similarity and quantification of hypervelocity impact experiments and laser experiments through computational simulations.　It was novel to apply the radiation-hydrodynamics code to investigate plasma conditions during hypervelocity impacts for the first time, permitting a non-equilibrium-based approach to the problem of impact plasma production and evolution.　The outcome was exceptional, and it finally explained the experimental results addressed by Ratcliff et al. a quarter century ago.　The viva panel also applauded that this work answered major questions about future instrumentation for planetary exploration, e.g., which method is best suited to analyse organics for astrobiological questions? What are the limits in the comparison between individual measurement methods? How do the known experimental spectrometer results fit with the latest computer simulations?

The viva panel noted that the only way to conduct this highly interdisciplinary research was to engage in close collaboration with world-class researchers within the respective disciplines.

Mr. Nakazawa made clear their roles in this work, adapting the code, running the calculations, and designing and performing new experiments, all with original contributions.

As for the English proficiency in this thesis, Mr. Nakazawa fulfilled international standards of writing skills. A solid background knowledge of the field was demonstrated throughout the thesis. Notably, he reviewed and characterised four major methods of in-situ mass spectrometry in the last 30 years: impact ionization, electron impact ionization, laser ionization, and photo ionization. The future work proposed in the last chapter indicated that Mr. Nakazawa was well capable of conducting creative research independently.

In conclusion, all the viva panel examiners agreed that this thesis is well deserved for a Ph.D. degree.

(EOD)