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論 文 内 容 要 旨

This thesis deals with the development of an experimental method to measure the small transient diffusion fields and the mass diffusion coefficients of several species were derived from the experimental data. The developed experimental set-up was applied to microgravity experiments and a comparison between the different transient diffusion fields under reduced gravity conditions and terrestrial gravitation conditions was investigated. This thesis consists of 7 chapters. Chapter 1 is introduction and the main body of this study is from Chapter 2 to Chapter 6 and Chapter 7 is general conclusions.

Chapter 1. General Introduction and Background

Transport properties, such as the viscosity, thermal conductivity, and diffusion coefficients are essential to the design of chemical processes and equipment involving fluid flow, heat and mass transfer and chemical reactions. In particular, the diffusion coefficient is one of the most important properties from scientific as well as technological viewpoints. One of the significant failing of the historical methodology for the investigation of diffusion phenomena is the lack of experimental data under various conditions. Hence, the existing database cannot satisfy a number of requirements in wide range of studies. In order to make the existing database useful for a wide range of situations, a large amount of experimental work needs to be done. Numerous types of experimental methods exist and many of these methods continue to be modified. However, these conventional methods have critical disadvantage, which is the executive measurement time required. A period of a few hours or even days are normally required to conduct the experiments. In order to make a new, useful database, an experimental technique for short-time measurement of diffusion fields is required. The objectives in this study are firstly to develop the method of derivation for mass diffusion coefficients using short-time measurement technique. The second objective is to carry out microgravity experiment in order to investigate the relationship between body force and diffusion phenomena.

Chapter 2. Theoretical Approach to Mass Diffusion Coefficients

This chapter describes a theoretical approach to mass transport phenomena. The influence of body force upon the diffusion phenomena and expression of mass flux in fluids were considered and a governing equation of transport phenomena in the diffusion region was derived. A theoretical consideration of diffusion phenomena proposes that there is negligible influence by terrestrial gravitation force upon the transient diffusion fields if the elapsed time is less than 300 seconds. Furthermore, a theoretical expression for the derivation of mass diffusion coefficients was proposed from the governing equations. This expression can derive the mass diffusion coefficients by using experimental data for location of the fringes and the elapsed time from the commencement of diffusion. The advantage of this method is that it can derive a large number of mass diffusion coefficients at the same time, so that it ensures an accurate mean value for the mass diffusion coefficient.

Chapter 3. Development of Measurement System for Transient Small Diffusion Fields

This chapter deals with the development of a new experimental method for the measurement of the mass diffusion coefficients, using a real-time phase shifting interferometer and shearing type cells for the measurement of small transient diffusion fields. The mass diffusion coefficients were derived from the experimental data of transient diffusion fields within 300

seconds as shown in Fig.1. The results of all experimental runs suggested that the present method could be utilized for a wide range of concentrations, regardless of the kind of solutes and their concentrations. Although the derived diffusion coefficients were slightly smaller than reference values in the literature, they were considered to be in good agreement with this reference data. The utilization of shearing type diffusion cells leave some disturbances in the

diffusion fields for a period generally less than 200 seconds. These remaining disturbances are not macroscopic such as turbulence, but microscopic. Therefore, only the experimental data whose elapsed time is over 200 seconds are utilized in deriving the mass diffusion coefficients. The measurement uncertainty was quantified as described in the ANSI/ASME Standard. The experimental data of this present method was evaluated by 95% coverage definition, and estimated to be less than $\pm 8.5\%$ uncertainty.

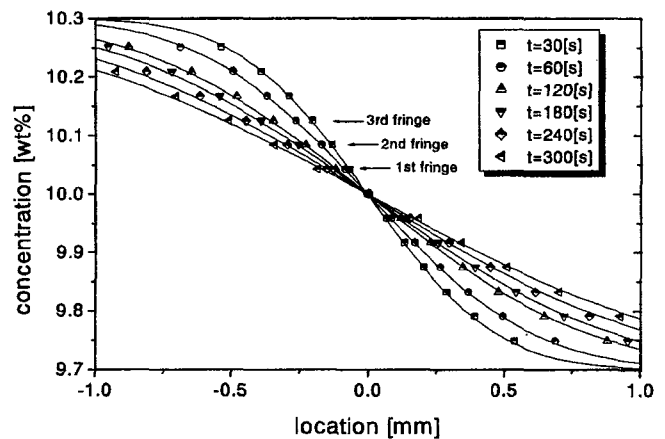


Fig.1 Concentration profile of transient diffusion fields

Chapter 4. Development of Measurement System for Transient Small Diffusion Fields

This chapter deals with a new experimental technique, which utilized the phase shifting interferometer and the counter flow type (CFT) diffusion cell as shown in Fig.2. The cell has several unique properties: extremely weak initial turbulence, giving a sharp step change of concentration field as the initial condition, and giving continuous diffusion fields. In the experiments, the thickness of the initial boundary layer of concentration was estimated to be less than 0.1 mm, which corresponded to the analytical results for designing the CFT cell. No visible disturbances

have appeared at the beginning of diffusion, and the diffusion fields in the small channel develop one-dimensionally. The transient diffusion fields were produced by this new cell type and were observed experimentally. The diffusion coefficients of several species were derived from experimental data, whose elapsed time were within 20 seconds, and evaluated by means of measurement uncertainty. However, the experimental data within 10 seconds could not be utilized for derivation of mass diffusion coefficients because of remaining initial disturbances. The derived data showed good agreement with reference values.

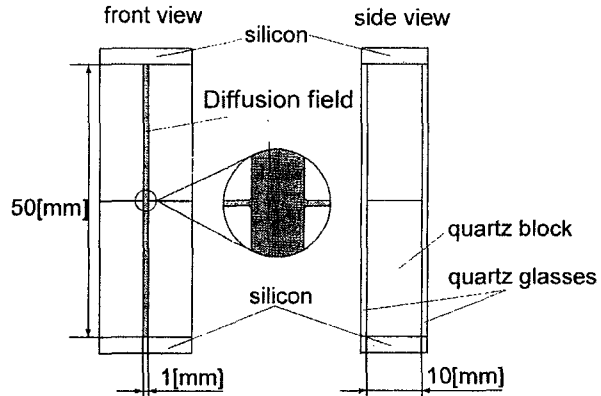


Fig.2 Schematics of counter flow type diffusion cell

Chapter 5. Application to Microgravity Experiments

This chapter describes the application to microgravity experiments by means of terrestrial facilities (an airplane for parabolic flight and a drop shaft). A combined damping system, which was developed for parabolic flight experiments, was used to reduce the external force condition. The quantitative evaluation of several microgravity conditions were carried out.

The drop shaft facility could produce the environment of reduced acceleration level of the order of 10^{-4} G without any kind of damping system. The parabolic flight, however, could produce the environment of reduced acceleration levels of the order of 10^{-5} G with the combined damping system. From the experimental data, the diffusion coefficients were estimated to be much smaller in comparison to that under normal gravitational conditions as shown in Fig.3. The difference of the values between the

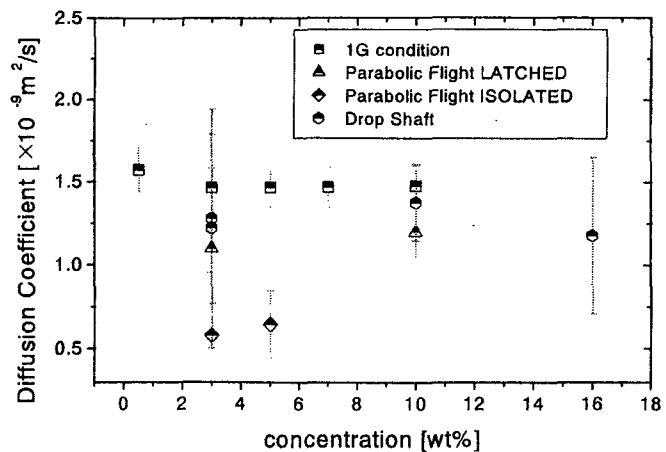


Fig.3 Summary of both normal and microgravity experiments

1G data and the low gravity data strongly depends on its acceleration level and g-jitter. From these results, the anomalous diffusion phenomena has been observed under the low acceleration level conditions. There is a possibility that an external force such as terrestrial gravitation and g-jitter have great influence upon the diffusion phenomena and transmute the diffusion phenomena based on the fundamental principle of mass transfer.

Chapter 6. Diffusion Phenomena and External Force

In this chapter, the experimental results from Chapter 4 and Chapter 5 were summarized on the same chart and the anomalous diffusion phenomena described in Chapter 5 was discussed. An empirical expression, which indicates the relationship between mass diffusion coefficients and the acceleration level of an external force on an environment undergoing diffusion, was proposed from experimental data as shown in Fig.4. It was shown that the empirical expression was applicable

within the gravitational range of 10^{-4} G to 1 G. The theoretical expression for the mass diffusion coefficients in liquid has been proposed and evaluated. In order to explain the anomalous diffusion phenomena which could be observed under the reduced external force condition, one hypothesis was proposed. This hypothesis treated the concept of micro-convection, which occurs under the gravitational fields and prevents the H₂O molecules from associating. Hence, the mass of molecules changes subject to the external force. By using the concepts of the “hydrodynamic theory” and “association phenomena”, an hypothetical expression for the mass diffusion coefficients in liquid are proposed, with several assumptions. The proposed expression was evaluated using experimental data, and it was suggested that the expression may describe the mass diffusion coefficients of a 3.0 wt% NaCl-H₂O binary system in liquids. However, a further investigation should be conducted to determine the general mechanism of the anomalous diffusion phenomena.

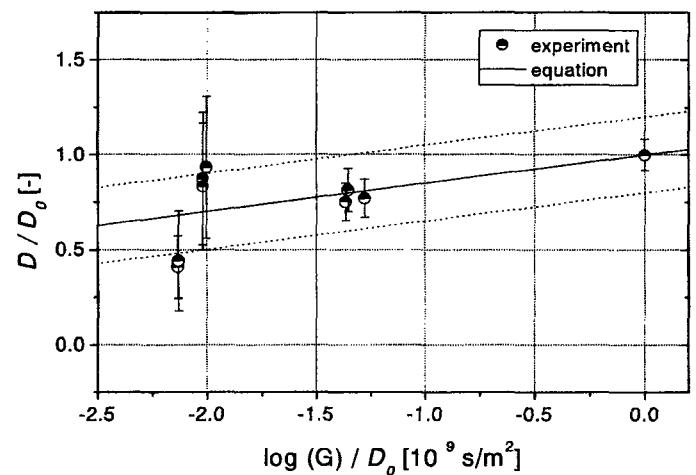


Fig.4 Relationship between external force and diffusion coefficients

Chapter 7. General Conclusions

In this study, the experimental method to measure small transient diffusion fields has been developed and the mass diffusion coefficients were derived using experimental data. The developed experimental set-up was applied to microgravity experiments and the influence of external force upon the diffusion phenomena was investigated. The contributions and new understandings of each chapter were summarized.

論文審査結果の要旨

物質拡散係数は熱物質移動現象を論ずる上で重要な物性値であり、様々な分野においてデータベースの確立が期待されている。従来の測定法では測定時間に長時間を要していたため、多様な物質の測定には困難を伴うことから、高効率な測定法が望まれている。本論文では、微小領域を高精度に測定することで測定時間の短縮化が可能であることに着目し、光干渉法および微小非定常拡散セルを用いた物質拡散係数の高精度短時間測定装置を確立した。また、近年の宇宙環境利用に伴う微小重力実験結果として報告されている拡散異常現象に着目し、短時間微小重力実験を行った。これらのデータから外力が拡散現象に及ぼすミクロな視点での影響について考察を行った。本論文は、以上の研究成果をまとめたもので、全編7章より成る。

第1章は序論であり、本研究の背景および目的について述べている。

第2章では、拡散現象の理論的記述について述べており、これまで考慮されなかった重力の拡散現象に対する影響について明らかにした。また、光学干渉計を用いた高精度な物質拡散係数の導出式を提案している。これは画期的な導出法である。

第3章では、微小非定常拡散場の高精度測定法に関する実験装置・手法を述べ、既存の物質拡散係数と比較することによって、その装置特性を明らかにし、さらには未だデータが得られていない物質拡散係数の測定を行った。

第4章では、新たな拡散セルを設計し、20秒以下の測定時間で物質拡散係数を測定できる手法を提案し、その評価を行った。この手法は、短時間で高精度に物質拡散係数が測定可能で、データベースの確立に大きな寄与をもたらす。

第5章では、第4章で確立した短時間測定装置を地上で得られる微小重力実験に応用し、外力がミクロな視点での拡散現象に影響を及ぼしていることを実験的に明らかにしている。これは高く評価できる知見である。

第6章では、第4章・第5章で得られた実験結果を整理し、外力の流体现象におよぼす影響に関する詳細な検討を行っている。また、流体内での分子の挙動について一つのモデルを提案している。

第7章は本論文で得られた成果をまとめた総括である。

以上要するに本論文は、物質拡散係数の高精度短時間測定法を確立し、その評価を行うとともに、その測定を微小重力実験にまで発展させ、外力と拡散現象の関係を示唆したものであり、熱工学および宇宙工学の発展に寄与するところが少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。