様式Ｆ－１９

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研究課題名（和文）　エレクトロスプレーデポジションにおける包括的数値解析手法の開発

研究課題名（英文） Development of a comprehensive numerical simulation technique for ESD

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研究成果の概要（和文）：
エレクトロスプレーデポジションにおける包括的数値解析手法の開発というテーマの研究は計画通り順調に進められた。以下の4つのプログラムは開発されました：
１、試料液用のナビエストークスシステムと、電位用のラプラスの方程式を組み合わせたプログラム；２、液体ジェットフローを含む周囲のガス用のプログラム；３、静電界シミュレーション用のプログラム；４、特定の堆積プロセスをモデル化し、堆積特性を評価するために、液滴の軌跡を追跡するためのラグランジュプログラム。
これまでに，当プロジェクトの得られた結果は第8回日韓熱流体工学会議とWCCM2012のカンファレンスペーパーおよび雑誌 Particuology と Journal of Micromechanics and Microengineering. で出版され，雑誌Japan Journal of Industrial and Applied Mathematicsへの投稿が受理された。

研究成果の概要（英文）：
The comprehensive numerical simulation technique for ESD has been developed successfully. It includes the following four programs: 1. Solver for Navier-Stokes system of the sample liquid coupled with the Laplace's equation; 2. Navier-Stokes solver for modeling surrounding gas with liquid jet flow; 3. Program for electrostatics field simulation; 4. Modeling Coulomb fission and tracking droplet trajectory. Up to now, the results from this project have been published in the conferences 8th JSME-KSME Fluid Engineering Conference and 10th World Congress on Computational Mechanics (WCCM 2012), as well as international journal Particuology and Journal of Micromechanics and Microengineering. Furthermore, one paper has been submitted to Japan Journal of Industrial and Applied Mathematics.

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１．研究開始当初の背景

Electrosprays are of great interest for technical application as well as tool for scientific research. Electrospray deposition (ESD) is a method to fabricate thin films of micro/nanoparticles directly from solution. Compared with other deposition methods, ESD has following merits: (1) the ring stain effect does not occur during deposition process; (2) high resolution and uniform patterning are possible; (3) size of dried particles can be controlled by concentration of liquid; (4) thick film or multi-layered structure can be fabricated by the control of deposition time; (5) deposition efficiency is intrinsic high. ESD has been used in various processes, such as sample preparation in mass spectrometry, modification of silicon surfaces, semi-conductive ceramics, polymer coatings, DNA and protein films for scanning tunnelling microscopy, and functionally and biologically active protein deposition.

The fundamental concepts of ESD are considered to be the following four steps: (1) by applying a high voltage between a counter electrode and a thin capillary where the sample solution is stored, liquid forms a cone-like shape called a Taylor cone; (2) the electric field concentration makes the solution spray and form a jet; (3) the jet is often divided into mother droplet, and the mother droplets break up into secondary droplets and satellites by electric charges on its surface (coulomb fission); (4) small droplets dry rapidly through evaporation to form charged nano-particles and accumulate on the conductive substrate that is connected to the ground to release their charge. Using an appropriate nonconductive stencil mask with surface charge and pre-patterned substrate, electrodynamic focusing process and precise deposition can be implemented.

To obtain an optimum deposition condition using electrospray device, many parameters (applied voltage, capillary-substrate distance, aspect ratio of stencil mask, shape of substrate pattern, surface charge density, solvent property, deposition time, liquid feed rate, particle charge, substrate temperature, configuration of capillaries in the case of multiple spaying and et al.) have to be considered carefully, and a lot of efforts have to be made if it entirely relies on experiments. Numerical simulation techniques have been developed greatly and become an indispensable tool in both academic field and industrial field. However, due to the complexity of the multiphysics phenomenon during the whole ESD process, the several studies on ESD simulation only considered the step 4 described above, which means that getting an initial droplet size distribution used for simulation still had to rely on experimental data and some assumptions such as Gaussian distribution assumption, and the influence of jet flow to the trajectories of droplets was ignored. Furthermore, those simulation results were only compared with experimental data qualitatively, and error analysis was not carried out and validity was not verified. Therefore, the above studies cannot serve as a strong tool for ESD operation.

２．研究の目的

Electrospray deposition (ESD) as a patterning method of dried micro/nano-particles on the substrate has attracted great attentions recently. However, obtaining an optimum deposition condition for ESD relies heavily on many trial experiments due to the lack of a reliable numerical simulation technique. The purpose of this study is to provide an accurate and comprehensive numerical simulation technique which can be used as quantitative analysis for ESD operation.

３．研究の方法

The developed numerical simulation technique for ESD includes four programs which are the program of Navier-Stokes system for the sample liquid coupled with the Laplace's equation for the electric potential to get cone-jet velocity and mother droplet information, the solver for surrounding gas with liquid jet flow to evaluate velocity and temperature distributions, electrostatics field simulation code and Lagrangian solver for tracking the trajectory of droplets to model a specific deposition process and evaluate the deposition characteristics. The numerical simulation technique is verified by comparing with experimental data.

The four programs and their relationships are described in details as follows:

(1) Solver for Navier-Stokes system of the sample liquid coupled with the Laplace's equation

To develop a comprehensive ESD simulation technique, the first thing we should do is to analyze how a cone-jet forms and how the thin jet that emanates from the cone's tip break up into mother droplets (step 1 and 2 described in "Purpose of the Research"). A coupling numerical scheme of Navier-Stokes system of the sample liquid and the Laplace's equation for the electric potential, which successfully simulated the formation of cone-jet and mother droplet, was adopted in this study and used to evaluate the jet velocity and the mother droplet's diameter, charge, position and velocity.

(2) Navier-Stokes solver for modeling surrounding gas with liquid jet flow

Drag force from the surrounding gas is one of the factors determining the trajectory of droplet. To evaluate drag force, velocity distribution in the gas flow field was calculated by solving the continuum-assumption based Navier-Stokes equations. The jet flow of sample liquid from the capillary modeled in program (1) has important effect on the velocity distribution, and was considered as the boundary condition of the surrounding gas simulation. Droplet evaporation is one of key processes for ESD. To make the droplet evaporate fast, sometimes the substrate needs to be heated. Therefore, the temperature distribution in the gas field was modeled for droplet evaporation calculation by solving energy conservation equation together with continuity equation. The finite element method was used to solve those governing equations.

(3) Program for electrostatics field simulation

Electrical force has an important impact on the droplet trajectory and deposition characteristics. An accurate simulation results on electrical field distribution is extremely important. The Laplace's equation for the electric potential will be solved using finite element method.

(4) Modeling Coulomb fission and tracking droplet trajectory

The mother droplet modeled at program (1) will break up into a secondary droplet and satellites, and the break-up will repeat with the process of evaporation until the ratio of the electric normal stress to the surface tension stress on the newly generated droplet less than unit. The course is usually called Coulomb fission. By combining the secondary droplet calculation model and the experimental data on Coulomb fission, the size distribution and charge of the small droplets (satellites) are obtained. To evaluate the deposition characteristics i.e., our final aim, such as deposition region, deposition size, the relationship between deposition thickness and time, particle size distribution on the substrate and et al., the trajectories of those droplets are tracked by using three-dimensional Lagrangian method through the transport equation $m_p(dV_p/dt) = F_D+F_B+F_E$, where m_p is particle mass, F_D is drag force, F_B is Brownian force, and F_E is external force. F_E describes the forces on a particle and is expressed by $F_E=F_C+F_I+F_{Di}+F_{vdW}$, where F_C is the Coulomb force that is induced by charges and electric fields, F_I is the image force acting on a metal particle, F_{Di} is dipole force caused by the polarization of particle by the electric field, and F_{vdW} is van der Waals force. The modeling of Coulomb fission and droplet trajectory are accompanied with the evaporation calculation.

The above four programs were combined as an organic whole after developing each one. The validity and accuracy of the software kit was verified by comparing with experimental data.

４．研究成果

The comprehensive numerical simulation technique for ESD has been developed successfully. It includes the above four programs. Up to now, the results from this project have been published in the conferences 8th JSME-KSME Fluid Engineering Conference and 10th WORLD CONGRESS ON COMPUTATIONAL MECHANICS (WCCM 2012), as well as an international journal Particuology and Journal of Micromechanics and Microengineering. Furthermore, one paper has been submitted to Japan Journal of Industrial and Applied Mathematics.

５．主な発表論文等
（研究代表者、研究分担者及び連携研究者には下線）

〔雑誌論文〕（計３　件）

① Wei Wei, Zhaolin Gu, Sheng Wang*, Takeshi Fukuda, Kiwamu Kase, Jungmyoung Ju, Yutaka Yamagata and Yusuke Tajima , Numerical simulation of nanoparticle pattern fabricated by electrostatic spray deposition, Particuology, Volume 11, Issue 1, 2013, P. 20-24. (peer reviewed)

② Wei Wei, Zhaolin Gu, Sheng Wang, Yunwei Zhang, Kangbin Lei and Kiwamu Kase, Numerical simulation of the cone-jet formation and current generation in electrostatic spray - modeling as regards space charged droplet effect, J. Micromech. Microeng., 22, 2012. (peer reviewed)

③ Wei Wei, Sheng Wang*, Zhaolin Gu, Yunwei Zhang, Kangbin Lei and Kiwamu Kase, A comprehensive numerical simulation t echnique for electrostatic spray dep osition, submitted to Japan Journal of Industrial and Applied Mathematics(to be peer reviewed)

〔学会発表〕（計　２ 件）

① Wei Wei, Sheng Wang*, Zhaolin Gu, Kiwamu Kase, Yusuke Tajima, Takaaki Orii, Hideaki Takaku, Noboru Imai, Jungmyoung Ju and Yutaka Yamagata, Numerical computation of cone-jet formation and deposition characteristics in electrostatic spray, 10th World Congress on Computational Mechanics (WCCM 2012), Sao Paulo, Brazil, July 8-13, 2012.

② Wei Wei, Sheng Wang*, Zhaolin Gu, Kiwamu Kase, Yusuke Tajima, Takaaki Orii, Hideaki Takaku, Noboru Imai, Jungmyoung Ju and Yutaka Yamagata, Development of comprehensive numerical simulation methodology for electrostatic spray deposition, The 8th KSME-JSME Thermal and Fluids Engineering Conference, Inchon, Korea, March 18-21, 2012.

〔図書〕（計　０　件）

〔産業財産権〕
○出願状況（計　０ 件）

○取得状況（計　０ 件）

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