# 博士学位論文審査要旨

２０２３年1月１４日

論文題目： Ultrasonic Atomization—Its Periodic Characteristics Involving Liquid-Fountain Oscillations Associated with Mist Generation
（ミスト発生を伴う液柱振動挙動に立脚した超音波霧化の周期特性）

学位申請者： 王　小璐

審査委員：

主　査： 理工学研究科　教授　土屋　活美

副　査： 理工学研究科　教授　塩井　章久

副　査： 理工学研究科　教授　白川　善幸

要　　旨：

超音波霧化（UsA）は、比較的均一に分散した微小液滴群から成るミストの連続生成プロセスとして、化学工業・環境・医学といった様々な分野での応用が期待されており、特に液自由表面で表面過剰を伴うような溶液に適用した場合、既存の霧化技術ならびに分離技術に代わるグリーンかつ省エネルギーな「微細液滴（ミスト）内濃縮分散技術」として極めて有用である。しかし、その実用化／効率化に不可欠な霧化機構に関しては、依然解明されていないのが現状である。

本論文は、UsA プロセスのメカニズム解明に向けて、その基盤現象の一つである自由表面の液柱化、特にそのダイナミックスを司る周波数特性について検討したものである。論文の序盤で、効率的ミスト分散を応用可能な様々な特定技術について整理し、基礎特性である液柱形成・形態遷移・振動現象解明の重要性を説いた後、データ整理に有用な時系列解析（FFT、離散ウェーブレット DWT 解析等）について解説している。論文の本論では、高速度・高解像度可視化解析を通じて、まず低印加電力での UsA 現象を捉え、画像データおよび理論／モデル解析により「数珠状」液柱の数珠径・振動周波数を、それぞれ基本原理・Rayleigh 理論に基づき、定量的に予測している。続いて、高印加電力下、実用化に即した（通常の）複雑ダイナミック形態をもつ液柱の振動現象を画像解析し、定量的記述が困難な高周波数（≥ 1MHz）励起による（高直進性超音波の伝播方向に垂直な）振動現象を、極簡略化した物理解析モデル「振り子モデル」で定量化すると同時に、超音波の最適照射角度を見出した。いずれの解析も最終的なミス発生の引き金となる振動の位相を提供し、液柱形成の特徴的な４段階（液面隆起、液柱土台成長・振動、ミスト発生源である微細液面形成および液柱の複雑化）の解明も含め、極めて有用な知見を得ている。

よって、可視化実験および解析モデルによる検討を通じて、超音波霧化の基本特性の解明を行なった本論文は、博士（工学）（同志社大学）の学位を授与するにふさわしい十分な価値を有するものと認められる。

# 総合試験結果の要旨

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要　　旨：

本論文提出者は、2019 年 3 月に本学大学院理工学研究科応用化学専攻博士前期課程を修了し、2019 年 4 月より 本学大学院理工学研究科応用化学専攻博士後期課程に在籍している。本論文の主たる内容は、*Ultrason. Sonochem.* **86**, 105997, pp.1–13 (2022)、*Fluids* **7**, 306, pp. 1–19 (2022) に掲載され、十分な評価を受けている。

2023 年 1 月 14 日 午前 10 時より約 1 時間 30 分に亘って提出論文に関する学術講演会（博士論文公聴会）が開催され、種々の質疑討論が行われたが、提出者の説明により十分な理解が得られた。さらに講演会終了後、審査委員により学位論文に関連した諸問題につき口頭試問を実施した結果、十分な学力を確認できた。提出者は、英語による論文発表ならびに国際会議での発表、語学試験にも合格しており、十分な語学能力を有すると認められる。よって、総合試験の結果は合格であると認める。

# 博 士 学 位 論 文 要 旨

論 文 題 目：　Ultrasonic Atomization—Its Periodic Characteristics Involving Liquid-Fountain Oscillations Associated with Mist Generation
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氏　　　名：　王　小璐

要　　　旨：

Ultrasonic atomization (UsA) is often regarded as a means of generating rather uniform distribution of a swarm of small droplets, or mist. In reality, the UsA-generated mist consists of droplets spanning wide ranges of their diameters, from micro- down to nanometers. A variety of its applications are evidenced in the fields including: selective separation or concentration of solute from organic solution, spray cooling, film coating, preparation of micro- and/or nano- sized powder, air purification, medical applications, and applications utilizing sonochemical reactions/reactors associated with cavitation. Among those applications, a typical technique of installing the ultrasonic transducer is to set it, in the form of an oscillating disk, within a liquid and to irradiate ultrasonic wave almost vertically upward. In such a configuration, as the bulk liquid is irradiated at a high driving frequency, a fountain of liquid will emerge from its free surface and micro- to nano- sized droplets could be generated—as mist—from the fountain. However, the current literature does not contain specific information regarding the fountain characteristics other than those of the mist as well as droplets. This study is claimed to be the dynamic elucidation through a sequence of images, captured through high-speed, high-resolution visualization, of the periodically oscillating acoustic fountain, as well as the quantitative evaluation of the effect of acousto-related operating conditions on the structure and dynamics of the fountain and an attempt to reveal factor(s) triggering mist emergence via experimental frequency analysis and theoretical prediction, respectively.

This doctoral thesis consists of seven chapters. Chapter 1 serves as a preface to introduce the research background and applications of ultrasound-assisted technology. Chapter 2 provides the overview and applications of ultrasonic atomization, including 1) the chemical and physical effects that can be induced by ultrasonic irradiation in liquid media, such as heating reactions, acoustic cavitation, acoustic streaming and acoustic fountains, summarizing their main industrial applications, respectively; 2) the mechanisms of ultrasonic atomization formation: capillary-waves, cavitation (boiling) and conjunction hypothesis. 3) two important scientific impacts in ultrasonic atomization—the droplet size distribution (DSD) of generated mist, the selective separation/concentration of solute into the mist with its extent and the possible mechanism; 4) Summary of the surface structure and dynamics of ultrasonic fountains. After giving useful introductory literature information, Chapter 3 is devoted to the principles of experimental data analysis methods, frequency analysis (fast Fourier transform), and time-frequency analysis (discrete wavelet variation).

Chapter 4 deals with quantitatively elucidating the effects of the UsA driving frequency on the structure [including characteristic dimension(s)] and dynamics [viz., characteristic frequency(ies)] of a chain-of-beads fountain realized under lower input power intensity. Upon ultrasonic irradiation under confined operating conditions—driving frequencies of 1–3 MHz and input power densities of 3–6 W/cm$^2$, the UsA fountain takes the form of a chain of "beads" in contact, recurring steadily. The recurring beads themselves are characterized by the UsA *wave-inherent*, "effective" dimension. This *effective beads diameter* is found to almost coincide with *half* the UsA wavelength, which can be specified by *half* the UsA excitation/driving frequency. This predictive scheme is realized via physical principle that the wavelength decreases in inverse proportion to the increasing frequency for a given liquid (thus for a fixed ultrasound speed). The scheme appears to hold as long as the beads diameter will not exceed the capillary length. The cyclic properties of the UsA beads fountain, visually exhibited via high-speed imaging, are characterized by the dominant frequency obtained (FFT-evaluated) experimentally. This primary periodicity is found to be well predicted based on the simple theoretical model proposed by Rayleigh (1879) with a proper assignment of the mode of cyclic deformation of the fountain beads themselves. The secondary dynamic characteristics associated with the primary beads-fountain periodicity—droplets bursting and/or mist spreading—involve some limited probability and triggering. While the present time–frequency analysis, via DWT, failed to reach their quantitative elucidation due to their complexity, some requirement(s) are identified in terms of higher probability for the droplets bursting to be triggered.

In Chapter 5, we apply high-speed imaging/image processing to observe/analyze detailed dynamics of the acoustic fountain by changing the angle of ultrasonic irradiation. Here, a transducer operating at 2.4 MHz is used, which is commonly used in ultrasonic atomization technology. For ultrasound irradiated out of the transducer used with its installation angles ranging 0–6° or higher (up to 10° examined) and input power densities of 3.5–6.5 W/cm$^2$, the UsA fountain will exhibit a sequence of oscillating/intermittent characteristics/events: its vertical/axial growth and breakup; its lateral "compound swinging"; and its associated dynamics of mist formation and spreading. Through high-speed visualization, it was found qualitatively that as the extent of tilt (from the vertical direction) in the irradiation angle was increased, the degree of occurrence of mist generation and the amount of identifiable mist generated would decrease. This trend was associated with reduction in both the growth rate and breakup frequency of the fountain on the tilt. It is further found, through the analysis of time variation in the resulting angle of liquid-fountain inclination, that the swinging fountain fluctuates periodically in an asymmetric manner and its periodicity can fairly be predicted based on a simple "pendulum" model proposed. In addition, the optimum value should be recommended to be slightly tilted 2° from the viewpoint of stability of the UsA fountain and not to exceed 5° from that of effective mist generation.

Chapter 6 focuses on the process of ultrasonic atomization involves a series of dynamic/topological deformations of free surface realized by changing some acousto-related operating conditions, including ultrasound excitation frequency, acoustic strength or input power density, and the presence/absence of a "regulating" nozzle. By utilizing high-speed, high-resolution images, UsA fountains could be qualitatively identified as four representative transitions/demarcations: 1) the onset of a protrusion on otherwise flat free surface; 2) the appearance of undulation along the growing protuberance; 3) the triggering of emanating beads fountain out of this foundation-like region; and 4) the induction of droplets bursting and/or mist spreading. On the basis of Chapter 4, quantitatively examined were the two parameters specification—for the degrees as well as induction—of the periodicity in the protrusion-surface and beads-fountain oscillations, observed over wider range of driving/excitation frequencies (0.43–3.0 MHz) and input power densities (0.5–10 W/cm$^2$) of ultrasonic transducers with or without the stabilizing nozzle equipped. The resulting time variations for the extended operating ranges in the "measured" fountain structure, associated with the recurring-beads size "scalable" to the ultrasound wavelength, confirm the wave nature predictable from simple physical principle. Specifically, the thresholds in acoustic conditions for each of the four transition states of the fountain structure have been identified; in particular, the onset of *bifurcation* in the chain-beads diameter below a critical excitation frequency.

A summary and an outlook are discussed in Chapter 7. Upon lower-power ultrasonic irradiation, the steady formation of beads fountain would allow us to apply Rayleigh's theory. It is also found that the atomization of the fountain should be closely related to the higher-order deformation of the beads, which is not a completely random phenomenon and should concur with the fountain periodicity with a limited extent of probability. In experiments at higher power, variations in the ultrasonic irradiation angle improved the stability of the liquid fountain at the optimal tilt angle of 2°, as well as mist generation. Additionally, the proposed pendulum model can predict the periodicity of the fountain swinging and is considered to be able to provide further predictions regarding mist generation.