MEMS-Based MHz Integrated Ultrasonic Nozzles
With Applications to Micro/Nano Technologies

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ABSTRACT

This paper reports on results of atomization of water and gold colloid solutions using micro electro mechanical system (MEMS)-based 3-Fourier horn 0.5 MHz ultrasonic nozzles. The resulting droplets are much smaller and more uniform than those obtained by ultrasonic nebulizers at much higher frequency. The gold nanoparticles spray-coated on silicon substrates are seen to remain well dispersed.

Keywords: atomization, drops, Fourier-horn ultrasonic nozzles, MEMS, gold nanoparticles

1 INTRODUCTION

Atomization is the breakup of a volume of liquid into drops, resulting in a dramatic increase in the surface area available for heat and mass transfer in a chemical reaction. Ultrasonic atomization (liquid atomization by ultrasound alone) and two-fluid atomization (liquid atomization by air) are commonly used in industrial applications including combustion, spray coating, spray drying, and spray pyrolysis. The former produces much narrower drop-size distribution than the latter. Also, ultrasonic atomization involves Taylor-mode jet breakup (different from Rayleigh-mode jet breakup in ink-jet printing). While the drop diameter resulting from Taylor-mode jet breakup is much smaller than the jet diameter [1], the nozzle is less prone to channel plugging as well. Conventional ultrasonic atomization utilizes either a bulk-type nozzle or a nebulizer. Because of manufacturing difficulty, the highest frequency of the former that is commercially available is 120 kHz [2], which yields a peak drop diameter as large as 55μm [1]. With the addition of air, an ultrasound-modulated two-fluid (UMTF) atomizer [1] is capable of reducing the peak drop diameter by a factor of two.

This paper reports on the novel design [3] and MEMS-based fabrication of 0.5 MHz ultrasonic nozzles, and their applications to atomization of gold nanoparticles dispersions. Such silicon-based ultrasonic nozzles possess a number of advantages over the existing metal-based bulk-type ultrasonic nozzles, such as MEMS-based micro-fabrication technology for mass production, much higher ultrasonic frequency than the 120 KHz limitation referred to earlier, much smaller and more uniform drops, and much lower electric drive power requirements.

2 EXPERIMENTAL

The ultrasonic nozzle is made of a piezoelectric drive section and a silicon-resonator consisting of three Fourier horns as shown in Fig. 1. Each horn is of half-wavelength design with a vibration amplitude magnification of two. 3-D simulation was carried out using the commercial ANSYS program [4]. The simulation results for the 0.5 MHz 3-Fourier-horn nozzle used in the atomization experiment are also shown in the figure. Specifically, a pure longitudinal vibration occurs at the resonant frequency of 495 kHz. At this resonant frequency, the vibration amplitude gain at the nozzle tip is equal to the theoretical value of 2^3.

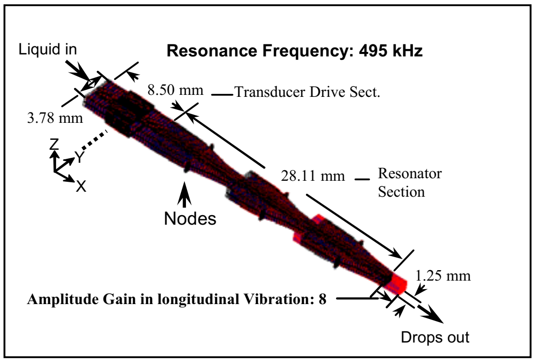

Figure 1: Dimensions and 3-D simulation results on the amplitude gain at the nozzle tip of a silicon-based 3-Fourier-horn 0.5 MHz ultrasonic nozzle.

2.1 Micro Fabrication of Nozzles

Major fabrication steps for the silicon resonator halves (including the base sections where PZT plates are to be bonded) using MEMS techniques are illustrated in Fig. 2. For simplicity, the horn profile is not shown in the figure. Inductive Coupled Plasma (ICP) etching, instead of wet etching, was used because of its capability to produce a smooth and precision finish in cutting through the 530 μm-thick wafer to make resonator halves with a rectangular trough 100μm deep and 200μm wide. Subsequently, two resonator halves were glued together to form a central rectangular channel (200μm x 200μm) for liquid flow. Two PZT plates, one on each side, were then bonded to the resonator at the base section using silver paste.

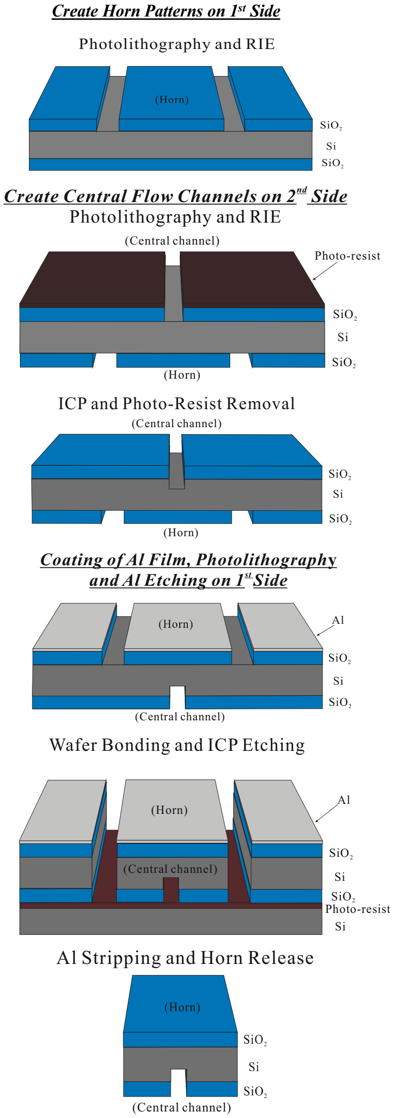

Figure 2: Micro fabrication steps for manufacture of the silicon-based ultrasonic nozzles

2.2 Preparation of Gold (Au) Sol

Au colloid solution (nanoparticles dispersion) was prepared as follows. In a 1-L Erlenmeyer flask, 500 mL of 1 mM $HAuCl_4$ was brought to a boil, with vigorous stirring on a magnetic stirring hot plate. 50 mL of 38.8 mM Na_3citrate was then added to the solution. The solution changed color from yellow to clear, to dark blue, to purple, and then to burgundy red within a few minutes. Boiling and stirring continued for 22 minutes. The solution was then removed from heat but kept stirring for 17 minutes. Subsequently, de-ionized (DI) water was added to make up 500 mL solution, resulting in 1 mM Au concentration. The Au colloid solution was then stored in a brown bottle at 5^{o}C.

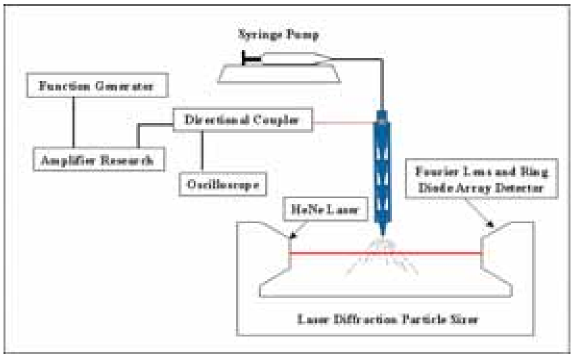

Figure 3: Schematic diagram of the atomization setup

2.3 Atomization Setup

A schematic diagram of the atomization setup is shown in Fig. 3. Major components of the setup are: (1) a PZT drive system to provide an alternating current (AC) electrical signal to the Si-based ultrasonic nozzle, (2) a Syringe Pump (Model 101, *Kd*-Sientific) to provide a constant liquid flow rate, and (3) a CCD camera to take pictures and movies or a Malvern Particle Sizer (Model 2600c) for analysis of drop sizes and size distribution. As shown in the figure, the pair of PZTs of the nozzle is driven by the AC electrical signal.

3 RESULTS AND DISCUSSION

3.1 Atomization Results

As water or Au colloid solution (nanoparticles dispersion) is pumped into the 150 µm-diameter channel of the nozzle at a constant flow rate, ranging from 10 µl/min to 150 µl/min, a liquid drop forms at the nozzle tip but no atomization takes place when the frequency of the drive signal differs significantly from the resonant frequency. In contrast, a thin film of liquid forms at the nozzle tip and a 0.4 mm-wide sheet of drops is seen when the drive signal is at the actual resonant frequency of 484.5 kHz. Due to resonance effect of the multiple Fourier horns, the drive voltage (7 V) required is one order of magnitude lower than that required in atomization using conventional nebulizers. The resulting drop-size distribution measured by the Malvern Particle Sizer 2600c is shown in Fig. 4. This figure shows that over 60% of the droplets have a diameter as small as 7 µm.

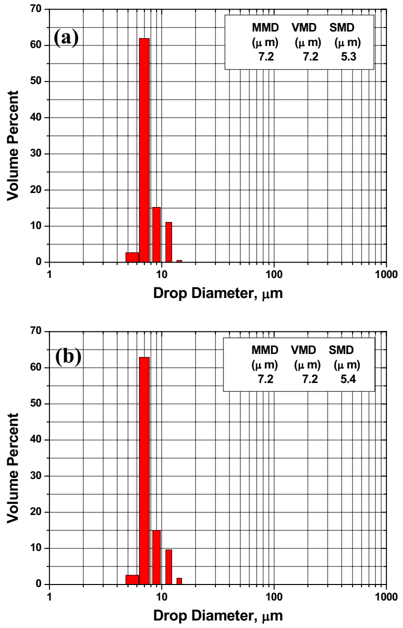

Figure 4: Drop size distributions of atomization using a Si-based 3-Fourier horn nozzle at the ultrasonic frequency of 484.5 kHz and the drive voltage of 7 V for (a) DI water and (b) Au colloid solution.

Atomization of Au colloid solutions shows that Au nanoparticles have no significant effect on the atomization conditions. The resulting drops are also similar to water drops in both mean diameters and size distributions.

In addition, the Au colloid solution was sprayed on clean silicon substrates for 5 seconds. Since the drops were so small, the sprayed substrates were air dried rather quickly. They are stored in a covered clean box for characterization using a Scanning Electron Microscope (SEM).

3.2 Characterization of Sprayed Au Nanoparticles on Silicon Substrates

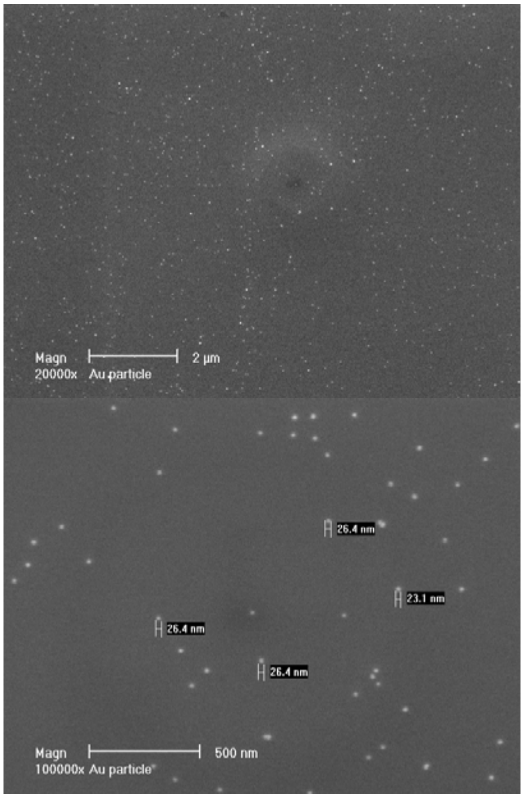

Figure 5: SEM micrographs of Au nanoparticles on silicon substrates

The morphology of the Au nanoparticles on the silicon substrates was examined using a Scanning Electron Microscope (SEM, Philips Model XL 30) with an EDAX energy dispersive X-ray detector for micro analysis of elements. As shown in Figure 5, the Au nanoparticles range from 23 to 26 nm in diameter. They are larger than 15 nm reported in the literature [5], possibly due to a slightly longer reaction time and the fact that the solution has been stored for two weeks before atomization on the substrates. Nevertheless, the SEM micrographs show that the nanoparticles are well dispersed on the silicon substrates with no pretreatment. Even the Au nanoparticles within a single drop remain dispersed with no agglomeration. Further study is in progress to determine the limit of such monolayer layout of Au nanoparticles on silicon substrates using such high frequency ultrasonic nozzles.

4 CONCLUSIONS

Actual atomization using silicon-based high-frequency ultrasonic nozzles has been achieved. The narrow bandwidth of the resonant frequency of these multiple-Fourier horn MHz nozzles facilitates generation of narrowly sized droplets. In fact, the drop diameter obtained at 0.5 MHz is as small as the peak diameter generated by the conventional ultrasonic nebulizers that operate at a much higher ultrasonic frequency (1.65 MHz).

In addition, nanoparticles such as Au in colloid solution (at least in dilute concentrations) appear to have no significant effects on atomization using such MHz ultrasonic nozzles. Furthermore, the nanoparticles remain dispersed on silicon substrates. Thus, the MEMS-based high-frequency ultrasonic nozzles may hold promise for application to nano and bio technologies. Other potential applications are: thin-film spray coating for micro- and nano-electronics processing and alveolar delivery of medicines.

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