Self-focused acoustic ejectors for viscous liquids

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Self-focused acoustic ejectors using the Fresnel zone plate (FZP) have been developed for ejecting viscous liquids, without nozzle, in the drop-on-demand mode. The FZP is composed of a lead zirconate titanate piezoelectric plate patterned with a series of annular electrodes, with the unelectroded region of the plate removed. Our results show that the acoustic waves are effectively self-focused by constructive interference in glycerin (with a viscosity of 1400 mPa s), giving small focal points with a high pressure. Due to the high attenuation, the wave pressure decreases significantly with the distance from the FZP. Nevertheless, the pressure at the focal points 2.5 and 6.5 mm from the FZP is high enough to eject glycerin droplets in the drop-on-demand mode. Driven by a simple wave train comprising a series of sinusoidal voltages with an amplitude of 35 V, a frequency of 4.28 MHz, and a duration of 2 ms, the ejector can eject fine glycerin droplets with a diameter of 0.4 mm at a repetition frequency of 120 Hz in a downward direction. Droplets of other viscous liquids, such as the prepolymer of an epoxy with a viscosity of 2000 mPa s, can also be ejected in the drop-on-demand mode under similar conditions. © 2010 American Institute of Physics. [doi:10.1063/1.3442526]

I. INTRODUCTION

Various dispensing systems, adopting either the contact mode or noncontact-mode ejection, have been developed to eject high-viscosity liquids, such as epoxies, in a drop-on-demand mode effectively and precisely.1–3 In the contact-mode dispensing systems, such as the Auger pump, piston pump, and time-pressure dispensing systems, a nozzle is moved close to a substrate and the liquid is forced out, either by an “Auger” feed screw, a piston, or a pressure, and placed on a substrate. The nozzle is then retracted and the liquid is broken off to form a droplet. The size as well as the quality of the droplets are affected significantly by the distance between the nozzle and the substrate (i.e., the dispense gap). If it is too narrow, the nozzle will be contaminated with the liquid and the wetting area in the drop breakoff process will be increased. On the other hand, if the gap is too large, the contact area between the liquid and substrate will become small and the drop breakoff process will be prolonged. Both conditions result in liquid residue at the nozzle and then variations in the droplet size. Accordingly, a precise z-directional control system for the nozzle is needed. Unlike the contact-mode systems, the nozzle in the noncontact-mode dispensing systems, such as the jetting dispensing system,3 remained at a fixed distance from the substrate. The nozzle has a ball and seat design. The liquid is pressurized to fill the void left by the ball as it retracts from the seat. As the ball returns (at a high speed), the liquid is “jetted” out the nozzle and onto the substrate. As there is no motion in the z axis, the cycle time can be significantly reduced from 90 (for typical contact-mode dispensing systems) to 15 ms. However, a control system is also needed for the spring-driven ball to shoot precise volume of the liquid. There are also drawbacks of the systems, such as wearing of the seat, clogging of the nozzle, and bulkiness of the nozzle. To achieve optimal and consistent viscosity, the liquid is usually preheated in the chamber located just above the nozzle.

Devices based on piezoelectric transducers have been extensively developed for ejecting liquid droplets.4–7 The liquid is generally forced to pass through a nozzle by the piezoelectric force/motion. Except for those devices using stack piezoelectric actuators, the devices are relatively simple and small, and can eject droplets at high frequencies (up to 16 kHz).5 However, due to the weak piezoelectric force, these devices are used only for the ejection of low-viscosity liquids, such as water, ink, liquid medicine, and photoresists. It has been shown that the energy required for droplet ejection is proportional to the viscosity of the liquid.8,9

Focused acoustic ejectors have recently been studied for ejecting liquid droplets from the surface of a liquid, without the need of a nozzle.10–13 Besides the simple nozzleless structure, the other advantages of the ejectors are the capabilities of ejecting viscous liquid as well as liquids with particulars which generally cause clogging of the nozzle. Various acoustic focusing mechanisms have been developed. Amemiya et al.15 demonstrated that a high-viscosity phosphor-dispersed resin (viscosity of 400 mPa s) can be ejected by a nozzleless printhead which utilizes an acoustic lens for focusing the ultrasonic waves. However, the fabrication of the acoustic lenses by mechanical grinding and polishing is difficult and expensive. Recently, the Fresnel zone plate (FZP), which was originally developed for focusing electromagnetic waves, has been demonstrated to focus the acoustic waves for ejecting liquid droplets.14–18 The zone plate is a piezoelectric plate containing a series of annular electrodes of a half-wave-band source.18 The acoustic waves are focused by constructive wave interference. In general, liquid droplets can be ejected in the drop-on-demand mode,
and the size, which is primarily determined by the wavelength of the acoustic waves, can be as small as 10 μm. However, most of the works have focused on ejecting low-viscosity liquids, such as water (1 mPa s), photoreacts (5–100 mPa s), and silicone resin (~400 mPa s).

Recently, we have demonstrated that viscous liquids, such as glycerin (1400 mPa s), can be ejected in the drop-on-demand mode using the FZP. The self-focusing capability of the zone plate has been confirmed by the measurements of the acoustic wave pressure distributions. The pressure distribution at different times has been calculated using a finite volume method in a computational fluid dynamics package (OPENFOAM). A mesh of arbitrary polyhedral cells in three dimension, bounded by arbitrary polygonal faces, is used for the calculation of the pressure distribution inside the liquid. The simulation results have revealed that the acoustic waves are self-focused effectively near the zone plate and along the axial direction (Fig. 1). Owing to the focusing and the corresponding increased intensity, a small volume of glycerin is stretched and deformed significantly to be broken into a fine droplet. Glycerin droplets can be ejected at a repetition frequency of 120 Hz. However, probably due to the hindrance from the unexcited piezoelectric material, a high driving field, about 100 kV/m, is required. In this work, milled FZPs (with the unelectroded region of the piezoelectric plate removed) will be fabricated so as to increase the vibration of the piezoelectric plate and hence reduce the driving field. Self-focused acoustic ejectors using the milled zone plates as the driving elements will be developed and fabricated for ejecting droplets of viscous liquids, such as glycerin, in the drop-on-demand mode. The self-focusing performances of the zone plates will also be investigated by the measurement of the pressure distribution of the acoustic waves in the liquid.

II. EXPERIMENTAL SETUP

A. Design and fabrication of FZPs

The FZP containing a series of annular electrodes of a half-wave-band source is shown schematically in Fig. 2. Acoustic waves are generated at the annular electrodes and propagate toward the designed focal point \( F_0 \). The waves arriving at \( F_0 \) are in phase and constructively interfere with each other. The wave generated at a particular point of an annular electrode should have a path difference in an integral number of wavelengths from the wave generated at a corresponding point of an adjacent electrode (Fig. 2),

\[
R_{i+2} - R_i = m\lambda,
\]

where \( i = 1, 2, 3, \ldots, m \) is an integer and \( \lambda \) is the acoustic wavelength in liquid. To achieve a maximum amplitude/intensity, destructive interference between the waves generated at the same annular electrode should be minimized. Hence, the path difference between the waves generated at the edges of the annular electrodes should satisfy

\[
R_{i+1} - R_i = \frac{\lambda}{2}.
\]

In order to fulfill Eqs. (1) and (2), \( m \) is taken as 1. The inner or outer radii \( r_i \) of the annular electrodes (Fig. 2) are then given as

\[
r_i = \sqrt{i\lambda f_0 + \left(\frac{\lambda}{2}\right)^2},
\]

where \( f_0 \) is the focal length.

Lead zirconate titanate (PZT) ceramic plates (APC 880, APC International Ltd., Mackeyville, PA, USA) with thickness 0.5 mm were used to fabricate the FZP. The thickness mode resonance frequency of the PZT plates was 4.28 MHz (which was the operating frequency of the self-focused acoustic ejectors). The inner and outer radii of the annular electrodes were calculated using Eq. (3), an acoustic wavelength of 0.448 mm and a designed focal length of 10 mm. As glycerin with a high viscosity of 1400 mPa s (Ref. 21) was mainly used as the liquid medium for the investigation, the acoustic wavelength (0.448 mm) was calculated using a wave velocity of 1920 m/s for glycerin and an operating frequency of 4.28 MHz. Silver annular electrodes were patterned on PZT plates by screen printing, followed by firing at 650 °C for 20 min. FZPs with six, five, and four annular electrodes (including the central electrode) were fabricated for the study. As only the piezoelectric material covered by the annular electrodes is excited by the external electric field, the vibrations will be hindered by the unexcited material next to it and hence become smaller. The effect will be more severe at the outer and thinner electrodes. In order to increase the vibration amplitude, the unelectroded region of the PZT plates...
was milled to a thickness of 0.35 mm. A photograph of the milled FZP (with six annular electrodes) is shown in Fig. 3.

B. Measurement of the wave pressure distributions

A needle-type hydrophone was used to study the pressure distribution of the acoustic waves in glycerin generated by the FZPs. The active element of the hydrophone is a poled P(VDF-TrFE) copolymer film with a diameter of 0.5 mm and a thickness of 6 μm. The FZP was mounted at the bottom of a cylindrical container. The container was then filled with glycerin to a depth of 30 mm. A signal generator (Sony Tektronix AFG 310, National Instruments, Austin, TX, USA) was used to generate a continuous sinusoidal voltage at a frequency of 4.28 MHz and a voltage (amplitude) of 10 V to drive the FZP. The voltage applied on the FZP \( V_a \) was measured by a digital oscilloscope (HP Infinium, Agilent Technologies, Inc., Palo Alto, CA, USA). A needle-type hydrophone was immersed in glycerin with its surface aligned parallel to the surface of the FZP. The voltage (amplitude) generated by the hydrophone was amplified by a preamplifier (National Physical Laboratory) and measured by the digital oscilloscope. The acoustic pressure \( P \) was calculated by dividing the measured voltage by the sensitivity (end of cable) of the hydrophone \((0.8 \, \mu\text{V/Pa})\).

C. Fabrication and performance evaluation of self-focused acoustic ejectors

For practical applications, the ejector is designed to eject droplets in a downward direction. The schematic diagram of the ejector is shown in Fig. 4. The FZP was mounted, with an air backing, to a cylindrical metal holder. The assembly was then fastened to a cylindrical tube, in which it can be rotated to move upward and downward so as to adjust the vertical position and then the liquid depth. The other end of the cylindrical tube was sealed with a thin metal plate, at the center of which there is a small orifice of diameter 1.0 mm. A thin metal ring with an inner diameter of 2 mm was adhered concentrically at the outside of the orifice. With the ring, a thin layer of liquid covering the orifice (i.e., meniscus) can be formed which is essential for the droplet ejection. A hole was opened at the side of the cylindrical tube for filling (and refilling during the operation) the chamber with liquid.

A signal generator (Sony Tektronix AFG 320, National Instruments, Austin, TX, USA) was used to generate a wave train comprising a series of sinusoidal voltages with a frequency of 4.28 MHz and a duration of T. The repetition rate of the sinusoidal voltages is \( f_{rep} \). The wave train was amplified by a power amplifier (NF Electronic Instruments, Yokohama, Japan) and then used to drive the FZP. A digital oscilloscope (Sony Tektronix TDX 220, National Instruments, Austin, TX, USA) was used to measure the voltage (amplitude) applied on the FZP \( V_a \). The ejection process was recorded using a high-speed digital camera (Casio FX 1, Tokyo, Japan), which can take 1200 pictures/s.

III. RESULTS AND DISCUSSION

A. Wave pressure distributions

The axial variations in the wave pressure (in terms of \( P/V_a \)) for the milled FZPs with six, five, and four annular electrodes are shown in Fig. 5. It should be noted that the active element of the hydrophone is slightly larger than the wavelength of the acoustic wave \((0.5 \, \text{versus} \, 0.448 \, \text{mm})\), so the measurement only gave an averaged value of the pressure at each position. In general, there is no significant difference between the pressure distributions for the three FZPs. For each FZP, three focal points with a (local) maximum pressure are observed. This may indicate that the contribution from the outer and thinner annular electrodes is not significant, partly due to the strong hindrance from the unmilled and unelectroded materials. As compared with the normal FZP (i.e., without milling), the observed pressure peaks associated with the focal points are higher and clearer. These should be attrib-
uted to the enhanced vibration of the electroded region (resulted from the reduced hindrance by milling) and hence more contribution to the interference from the outer electrodes (e.g., the third and fourth annular electrodes).

The observed focal point located at 0.95 mm from the FZP should be the designed focal point $F_o$, (with a designed focal length of 10 mm). The other two focal points should be resulted from the constructive interference at higher orders. The conditions given by Eqs. (1) and (2) (with $m=1$) are those for the first-order constructive interference, and the corresponding first-order (principle) focal point is the farthest one from the FZP. As the (axial) distance from the FZP decreases, the path difference between the waves increases. As a result of interference, the pressure (amplitude) of the resultant wave varies, giving a series of maxima and minima. For FZPs satisfying the conditions $r \ll z$ and $r \gg \lambda$, where $z$ is the axial distance from the FZP, Eq. (3) becomes

$$r_i = \sqrt{k_i L_{f_o}},$$

Equation (4) is generally applied for FZPs used in optical applications.23 Following similar procedures described above, higher-order focal points, at which the path difference between the waves from the (adjacent) edges of the annular electrodes equals an odd number of half-wavelength [refer to Eq. (2)], can be located. For example, the third-order focal length $f_3$ is given as

$$r_i = \sqrt{3k i L_{f_o}},$$

$$f_3 = f_o/3.$$  

It can be seen that each annular electrode gives the focal points of different orders at the same positions. As a result of multiple interference, there is a number of (principle) focal points located at $f_k$ (where $k$ is an odd number) and a number of secondary maxima in between of them. The principle focal points are sharp and have a very large pressure. However, for the FZPs studied in the present work and others used in an acoustic applications, the conditions $r \ll z$ and $r \gg \lambda$ are not satisfied. Owing to the second term in Eq. (2), the higher-order focal points for each annular electrode will be formed at different positions. As a result, the principle focal points may become broadened and the pressure may become smaller. It is hence suggested that the observed focal point ($F_3$) located at 0.255 mm should be the third-order (principle) maxima and the one ($F_4$) located at 0.62 mm should be the secondary maxima.

As also shown in Fig. 5, the pressure at $F_o$ is much smaller than those at $F_3$ and $F_4$. This should be attributed to the high attenuation of the acoustic waves in viscous liquids. It has been shown that the attenuation of an acoustic wave propagating in a liquid is dependent on its viscosity and density. A calculation has shown that the penetration depth for a 4 MHz acoustic wave traveling in glycerin is about 8 mm.24,25 The high attenuation may also cause a decrease in the contribution of the waves generated from the outer electrodes to the interference, making the axial pressure distributions of the three FZPs become similar (Fig. 5).

Because of the higher wave pressure, the focal point $F_3$ (at 0.95 mm) was chosen in this work for the ejection of viscous liquids. Figure 6 shows the variations in the pressure (in terms of the $P/V_o$) along a line passing through $F_3$ and normal to the axis for the FZPs with six, five, and four annular electrodes. It can be seen that the pressure for all the FZPs decreases drastically from the central, having a very small value at distances larger than 1 mm from the axis. This indicates that the acoustic waves are effectively focused and the corresponding focal point is very small, about 1 mm in diameter. It is also noted that the observed focal point is smaller than that produced by the normal FZP.19 This should be attributed to the enhanced vibration and then more contribution to the interference from the outer electrodes. As discussed before, the observed wave pressure is an averaged value over a small area of 0.5 mm, so the actual focal point should be smaller than 1 mm. In general, the dimension of the focal point is determined by and has the same order of magnitude of the wavelength which is about 0.448 mm.

B. Upward ejection

The ejectors, without the thin metal plate, were first operated in an upward direction so as to study the self-focusing capability of the FZPs and to locate the focal points. The chamber of the ejector was filled with glycerin (with a viscosity of 1400 cP) to different depths, in a step of 0.125 mm. It has been observed that glycerin droplets can be ejected only at a liquid depth of 2.5 (Fig. 7) and 6.2 mm, respectively, which agree with the observed focal points ($F_3$ and
$F_s$) from the wave pressure measurements (Fig. 5). This indicates that the liquid-air interface does not have significant effect on the focusing of the acoustic waves. The voltage (amplitude) $V_o$ used for the ejection at both focal points is the same, about 35 V. However, the duration $T$ used for the ejection at $F_3$ is shorter than that at $F_1$ (2 versus 3 ms). This may be attributed to the slightly larger pressure at $F_3$ (Fig. 5). At liquid depths other than $f_3$ and $f_o$, the liquid is deformed or displaced, forming a crest moving outward, as shown in Fig. 8.

### C. Downward ejection

After the evaluation of the ejection capability, the ejectors were sealed with the thin metal plate and evaluated for the downward ejection. Based on the results of the upward ejection, a wave-train signal with a $f_{op}$ of 4.28 MHz, a $V_o$ of 35 V, a $T$ of 2 ms, and a $f_{rep}$ of 1 Hz was used to drive the FZP. Using a liquid depth of 2.5 mm as a reference, the liquid depth was adjusted for the ejection. It has been found that the liquid depth for the downward ejection is the same as that for the upward ejection, i.e., 2.5 mm. Similarly, this indicates that the thin metal plate does not affect the focusing of the acoustic waves significantly. To evaluate the capability of the ejection in the drop-on-demand mode, wave-train signals of different $f_{rep}$ were used to drive the FZP. It has been found that the ejectors can eject glycerin droplets one by one, following exactly the repetition of the sinusoidal voltages. The maximum repetition frequency is 120 Hz, which is fast enough for practical applications, e.g., printed circuit board assembly and semiconductor packaging. A photograph showing the downward ejection of glycerin droplets at a repetition frequency of 120 Hz is shown in Fig. 9, while the operation parameters are summarized in Table I. It can be seen that the observed diameter of the droplets is about 0.4 mm, which is slightly smaller than the wavelength of the acoustic wave (0.448 mm). This indicates that the acoustic waves are effectively focused to a small focal spot. It has been shown that the focal spot area and then the droplet diameter are determined by the acoustic wavelength.\textsuperscript{10,14,18} As also shown in Table I, the voltage and duration of the wave-train signal are 35 V (or 70 kV/m) and 2 ms, respectively, which are lower and shorter than those for the normal FZP (100 kV/m and 3 ms).\textsuperscript{19}

The ejectors have also been used to eject droplets of a prepolymer of an epoxy (EE-7132A, Rapid Chemical, Taiwan) successfully. Except using a higher voltage ($V_o = 40$ V), the other operation parameters are the same as those for ejecting glycerin (Table I). The epoxy, which is designed for light-emitting diode encapsulation molding and fiber optical applications, has a higher viscosity of $\sim 2000$ cP compared with glycerin. As the wave velocity for the epoxy is not known, an optimum FZP cannot be designed and fabricated for it. Nevertheless, the successful trials have demonstrated that the ejectors can eject viscous liquids in the drop-on-demand mode for practical applications.

### IV. CONCLUSION

Milled FZPs with different numbers of annular electrodes have been designed and fabricated using PZT piezoelectric plates. The self-focusing capability of the FZPs has been evaluated by the measurements of the wave pressure distribution. Our results show that the acoustic waves are effectively focused in glycerin, giving a series of focal points with a high pressure and a small area. Although the waves are highly attenuated, the pressure at the focal points 2.5 and 6.5 mm from the FZP is high enough to eject glycerin

| TABLE I. Operation parameters for the self-focused acoustic ejector to eject glycerin droplets in the drop-on-demand mode. |
| Liquid viscosity (glycerin) | 1400 mPa s |
| Liquid depth (i.e., focal length) | 2.5 mm |
| Operation frequency ($f_{op}$) | 4.28 MHz |
| Voltage amplitude ($V_o$) | 35 V |
| Duration ($T$) | 2 ms |
| Maximum repetition frequency ($f_{rep}$) | 120 Hz |
| Droplet diameter | 0.4 mm |
droplets in the drop-on-demand mode. Ejectors fabricated using the FZPs have been fabricated. Driven by a simple wave train comprising a series of sinusoidal voltages, the ejectors can eject fine droplets of viscous liquids, such as glycerin (1400 mPa s) and the prepolymer of an epoxy (2000 mPa s), at a high repetition frequency in a downward direction. The droplets are small, having a diameter of 0.4 mm which is close to the wavelength of the acoustic waves.

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