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Effects of Ultrasmall Orifices on the Electro-generation of Femtoliter-Volume Aqueous Droplets

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Abstract

The ability to generate individual picoliter- and femtoliter-volume aqueous droplets on-demand is useful for encapsulating and chemically manipulating discrete chemical and biological samples. This paper characterizes the effects of orifice dimensions and material choices on generating such droplets in an immiscible oil phase using single high-voltage pulses with various amplitudes and durations. We have examined microfluidic orifices as small as 1.7 μm in equivalent radii, and found that the electrohydrodynamic jet lengths and the subsequent formation of droplets are affected by the axial aspect ratios of the orifices (length of an orifice divided by its equivalent radius). As higher voltages were used to compensate for the increased capillary pressure and hydrodynamic resistance in ultrasmall orifices, we observed secondary jet protrusions and droplet formations that were not of classical electrohydrodynamic origin. The droplets generated from secondary jets traveled at relatively lower velocities as compared to those of electrohydrodynamic origin, and these slow individual droplets are potentially more useful for applications in microscale chemical reactions.

INTRODUCTION

Aqueous droplets of volumes on the order of picoliters to femtoliters are useful for investigating chemical reactions involving small amount of reactants and the intrinsic biochemical behaviors of cells and subcellular structures at an individual level.^{1,2} The controlled formation of a single droplet on demand is often required to follow accurately the reactions or assays within the droplet. Generating a single sub-picoliter-volume droplet in an immiscible liquid phase is more challenging than in a gas phase because of the complex momentum coupling and the material displacement between the two liquid phases. Aqueous microdroplets are commonly generated in a continuous oil phase in a microchannel format by using T-channel configurations²⁻⁵ or with flow focusing geometries⁶ where the droplet size can be controlled by varying the shear rates of the oil phase relative to the aqueous phase. To generate a single subpicoliter volume droplet on demand using these approaches, however, requires complex and delicate external pressure balance and control.²

We have previously investigated the electro-generation of aqueous droplet in an immiscible phase as a method for forming a single droplet on demand.⁷ By applying high voltage DC pulses across the immiscible fluid interface, we characterized the formation of electrohydrodynamic jets and the associated droplet formation and demonstrated the generation of droplets with sizes ranging between 14 fL to 8 pL in volume. In this paper, we study the effects of orifice dimensions on the formation of electro-generated droplets and report secondary effects that caused additional droplet formation, which is not electrohydrodynamic in origin. The formation of droplets by these secondary effects is reproducible and the corresponding droplets, which are often slightly larger and much slower than those of

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electrohydrodynamic origin, may be manipulated more easily and are thus more suitable for applications in encapsulation and microscale reactions.

EXPERIMENTAL SECTION

Design and Fabrication of Microchannels

Figure 1 shows the schematic of the channel design. The microfluidic channels were fabricated in poly(dimethylsiloxane) (PDMS) by replicating off a silicon molding master produced by two-step photolithography.² The features of the first layer of the Si master (3–6 μm in height) were produced by spin-coating a low-viscosity SU-8 negative photoresist (e.g., SU-8 2002) onto a silicon wafer, pre-baking per manufacturer's recipes, followed by UV exposure under a chrome mask in a mask aligner and post-baking. Then a high-viscosity SU-8 (e.g., SU-8 2025 or higher viscosity) was spin-coated on top of the first layer to define the height of the second layer (20–40 μm) and also baked. Before the exposure of the second layer, the alignment marks on the first layer and the second-layer mask were carefully aligned with a mask aligner. After alignment, exposure, and post-baking, the wafer was developed and the resulting Si master was silanized. To form the microfluidic channels, the patterns on the master were replicated in PDMS. After the outlet holes were punched on the PDMS replica, the replica was sealed against a piece of PDMS-coated coverslip² using oxygen plasma. The complete device with enclosed channels was baked at elevated temperature (120 $^{\circ}\text{C}$) for at least one day to restore the surface hydrophobicity of PDMS microchannels. The channel structure consisted of two main flow channels bridged by a small channel constriction (orifice). The main channels were each connected to a reservoir containing different liquid phases; the main channel on the oil side (right) was also connected to an exit channel for removing unwanted oil or water in the system. The dimensional details of the orifice are illustrated in the dashed enclosure in Figure 1, with the depth (D), width (W), and length (L) labeled for clarification. The dimensions of the orifice were systematically varied to investigate the effect of the channel geometry on the jet protrusion and droplet generation. We used $D \times W \times L$ to represent the orifice geometry in this paper. The oil and water main channels were 500 μm wide and 1.2 cm long in all designs, but the depth varied from 40 μm (if the orifice length was between 60–100 μm) to 20 μm (if the orifice length was 24 μm). The exit channel on the oil side had the same depth and width as those of the main channels. The oil-water interface was maintained at the left entrance of the orifice prior to the initiation of an electrical pulse.

Experimental Procedure

The water reservoir was connected to a micropipette holder/electrochemical half-cell (“micropipette holder” for short, Model: MEH2RW, World Precision Instruments, Sarasota, FL) to allow simultaneous hermetic seal of the water line and electrical connection to the anode of the voltage pulse generator: a 1-mL syringe filled with water was connected to the inlet of the micropipette holder with a syringe needle and polyethylene tubing (PE 100, IntramedicTM, Becton Dickinson, Franklin Lakes, NJ), and another PE 100 tubing was inserted into the outlet of the micropipette holder. Prior to inserting the free end of the PE 100 tubing at the outlet into the water reservoir on the microdevice, the air in the entire line was displaced by manually pushing the 1-mL syringe to fill with water. Compared to the water delivery and electrode placement method in our previous work,⁷ the use of micropipette holder allows better pressure control on the water side, which is important for small orifice where substantial capillary resistance can be expected.

The oil reservoir inlet was connected via PE 100 tubing to a 1-mL syringe mounted on a syringe pump (BSP-1, Braintree Scientific, Braintree, MA). The pump was operated manually by adjusting the advancement screw. The channels were usually filled with oil at first, and then water was introduced into the channel by manually pushing the syringe filled with water. By

carefully balancing the pressure applied to both the water and oil syringes, the water-oil interface was brought to the left aperture of the orifice (water side).

Voltage pulses were applied with a custom-built high-voltage pulse generator with additional circuits to rapidly discharge voltage during the trailing edge of the pulse. A single electric pulse could be initiated by a manual trigger; repetitive pulses could also be applied. The output waveforms of the high-voltage power supply were repeatedly measured and were consistent with the pulse waveform reported earlier⁷ where the rise time of the waveforms was ~ 10 ms and the decay time was ~ 25 ms to reach 5% of the setpoints of the pulse voltages. To avoid too much water flowing into the oil side, we applied pulses with low voltages and short duration time at first, and then gradually increased the voltage or duration time to cause the formation of jets and droplets. The initial protrusion or curvature of the water-oil (W-O) interface toward the orifice influenced the jet and droplet formation as well. A larger protrusion or curvature of the interface toward the orifice led to a smaller voltage required for generation of droplets. To evaluate the influence of the pulses with a particular channel dimension, we kept the degree of the initial protrusions the same.

Materials and Instruments

For the immiscible organic phase, we mainly used soybean oil (Ventura Foods, City of Industry, CA) that had been degummed and saturated with deionized water. Silicone oil AR 20 with a density of 1.008 g/mL and a dynamic viscosity of 20 mPa·s at 25 °C (Fluka, Buchs, Switzerland) was also used as an alternate organic phase. The effect of the latter on W-O interfaces in small PDMS channels is evaluated later in the discussion section. For the organic phase, 3% (w/w) sorbitan trioleate (Span 85, Fluka, Buchs, Switzerland) was added to stabilize the W-O interface. The aqueous phase was tap water.

Images were acquired with an inverted microscope (Nikon Eclipse TE 2000-S, Tokyo, Japan) and a high-speed camera (CPL-MS10K, Canadian Photonic Labs, Minnedosa, Manitoba, Canada). The image of an orifice shows the dimensions of the width and length of the orifice.

RESULTS AND DISCUSSION

Effects of Orifice Geometry on the Formation of Droplets

Figure 2A shows an image sequence of the droplet formation process inside an orifice of $6 \times 6.7 \times 63 \mu\text{m}^3$ after the application of an electrical pulse (1000V, 10-ms duration). This observation is typical in orifices where the diameters of the electrohydrodynamic jets spanned most of the cross sections of the orifices. After the rising edge of the DC pulse, the interface between the aqueous phase and the organic phase became diffuse, and a jet was formed, rapidly protruding into the orifice half-way. At or immediately after the trailing edge of the pulse at 10 ms, a primary droplet was emitted and the jet began to recede toward the water entrance of the orifice. This droplet-emission process is consistent with our earlier observation.⁷ Figure 2B plots the position of the vertex of the jet as a function of time, and the positions of detached droplets are indicated with triangles.

A secondary protrusion occurred significantly later following the trailing edge of the pulse. Since the trailing edge of the pulse decayed at a finite rate (the voltage was ~ 200 V at 10 ms following the onset of the trailing edge of the pulse), some degree of influence from the electric field may still be expected after the pulse. Approximately 10 ms after the trailing edge of the pulse, the vertex of the jet advanced again. This time the advancement of the vertex was significantly slower, and reached an equilibrium position where the vertex lingered for ~ 30 ms prior to droplet detachment. In general, this secondary droplet was larger than the primary droplet, traveled at a much lower velocity, and occurred significantly later following the trailing edge of the pulse.

Table 1 summarizes the experimental results from several different orifice geometries, and we found that secondary droplets were formed consistently among these geometries. Although the period between the trailing edge of a pulse and the detachment of a secondary droplet can vary significantly, sometimes longer than 100 ms, the secondary droplet was clearly distinct from the primary droplet in its larger size and slower velocity.

The primary jet and droplet formation is of electrohydrodynamic origin, and has been discussed elsewhere⁷⁻¹¹. The dominant force to overcome in the formation of an electrohydrodynamic aqueous jet and the subsequent detachment of a droplet inside an ultrasmall oil-wetted hydrophobic orifice is the capillary force, i.e., Laplace pressure related to the interfacial tension. Laplace pressure (P_L) in a rectangular channel can be estimated:¹²

$$P_L \approx \frac{2\gamma}{r_e} \quad (1)$$

where γ is the interfacial tension between the immiscible phases, and $r_e = DW/(D+W)$ is the equivalent radius calculated from the depth (D) and the width (W) of the cross sectional dimension of the orifice. Experimentally r_e is approximately the same as the jet radius in small orifices of nearly square cross sections. Assuming a value of 22.8 mN/m for the interfacial tension γ between soybean oil and water¹³ without correcting for the reductive effect of the added surfactant, the Laplace pressure is 14 kPa for a channel of $6 \times 6.7 \mu\text{m}^2$ cross section. Based on the same assumption of the interfacial tension, we calculated Laplace pressure (P_L) of various cross sectional geometries used in the present study and listed the results in Table 1. These values, invariably on the order of tens of kilopascals, are significantly more than the kinetic energy (which can be estimated from u_{max} in Table 1) of the resulting droplet, suggesting that the electrical energy in these runs was barely above the threshold required to counter the capillary force, leaving very little momentum remaining for the kinetic motion of a detached droplet.

The maximum advancement velocity (u_{max} in Table 1) of a jet ranged between 1–6 mm/sec depending on the size of the orifice. When comparable electrical energy is used to form the jet, the orifice of a small equivalent radius suffers from a high capillary force, and thus the net kinetic energy of the vertex is lower than that formed in the orifice of a large equivalent radius. Thus for a smaller orifice, a higher voltage is needed to accelerate the jet sufficiently to increase the probability of the ejection of a primary droplet. For example, in comparing the data from the $3 \times 10.5 \times 78 \mu\text{m}^3$ orifice to that of the $3 \times 4.1 \times 78 \mu\text{m}^3$ orifice in Table 1, a simple increase in the width of the orifice while keeping the depth and the length the same led to more facile droplet generation. Although in the case of the wider orifice the jet usually did not occupy the full width of the orifice and assumed a radially asymmetric profile, the volume of the ejected droplet was often larger.

The axial aspect ratio (L/r_e) of the orifice affected the jet length and the subsequent formation of a droplet. Using just the minimal voltage and pulse duration sufficient for jet formation and primary droplet ejection, the jet length observed in a long orifice ($L=63\text{--}78 \mu\text{m}$) was just shorter than the full length of the orifice. Because the walls of the orifice confined the boundary of the jet, an orifice with high axial aspect ratio led to the formation of an elongated thin jet, which is unstable and highly susceptible to droplet detachment.

On the other hand, if the axial aspect ratio of the orifice was small, the droplet-generation process was less regulated. For example, when a pulse (800V, 20 ms) was applied across an orifice of $24\text{-}\mu\text{m}$ length ($3 \times 4.1 \times 24 \mu\text{m}^3$, Table 1), the jet quickly reached the full length of the orifice at 12 ms and instantly shed small emulsion droplets into the oil reservoir. There was a noticeable breakup at the end of the pulse (20 ms) and then the jet had two separate slow breakups. For this type of orifices, the water jet either rapidly reached the exit of an orifice and

formed multiple droplets when the electric pulse was powerful enough, or stayed intact inside the orifice without any breakup with the pulses of insufficient power. It was therefore difficult to produce small droplets singularly in such orifices. The amplitude of the voltage, pulse duration, and the initial protrusion of the water phase into a 24- μm -length orifice as required to induce droplet formation were lower compared to those required for a 78- μm -length orifice. We attribute the difference to a smaller hydrodynamic resistance associated with a shorter length from the Hagen-Poiseuille equation:¹⁴

$$P_h \propto \frac{l}{r_e^2} \quad (2)$$

where P_h is the hydrodynamic pressure drop and l is the length of the jet. The displacement effect of the oil phase on water was discussed in greater detail by Chan and Yang.¹⁵

Regardless of the axial aspect ratio of an orifice, we found experimentally that as long as the cross section of the orifice was sufficiently small, large capillary pressure ($P_L > 12 \text{ kPa}$) provided sufficient resistance to easily position and stabilize the location of the water-oil interface.

Secondary Jet Protrusions and Formation of Droplets

Secondary jet protrusion and droplet formation after the termination of the voltage pulse was observed when we used small orifices of sufficient hydrodynamic resistance and voltage pulses of amplitude or duration in excess of the threshold for primary droplet formation. When the applied voltage was barely enough to eject a primary droplet, only a primary jet was formed, which is illustrated in Figure 3 (the dimensions of the orifices in Figure 2 and 3 are identical). Following the trailing edge of the pulse, the primary jet receded back to the entrance of the orifice. Note that the droplet formed in this case did not move toward the exit of the channel, but instead drifted back toward the entrance in the same fashion as the jet vertex, suggesting that the droplet was carried by a flowing oil phase moving in synchronization with the receding vertex. The dominant mechanism for the recession of the vertex (interface) is mostly capillary action, or the preferential wetting of channel walls by the soybean oil.¹⁵

At higher voltages secondary jet protrusions were observed. Figure 2A shows that, following the ejection of the primary droplet at the trailing edge of the pulse (10 ms, see frame 11.3 ms), the droplet and the vertex moved in opposite directions. As the droplet moved toward the exit, the vertex retracted briefly. The space between the droplet and the vertex must be filled by additional oil flowing in either from the orifice exit (right), flowing past the droplet, or from the orifice entrance (left) if some oil had remained on the orifice walls next to the jet prior to initiating the pulse. The vertex then stopped retracting and began to protrude into the orifice again, although moving at a much slower velocity than the primary electrohydrodynamic jet. The rapidly decaying pulse (less than 200 V at more than 10 ms after the termination time of the pulse) led to a fast-decreasing electrostatic stress exerted on the jet. In the absence of any additional external driving force, such secondary protrusion is impossible; the capillary force (preferential wall-wetting by oil) discussed earlier would have preferred to push the aqueous phase out of the orifice as the electrostatic stress diminished. We believe that the secondary protrusion is due to a time-lagged electrowetting of the PDMS walls of the orifice and the change in the charge density on the liquid jet, because this response was usually observed when a voltage higher than that of the minimum voltage required for the formation of a primary droplet was applied. The subsequent droplet formation from a secondary jet is due to the unfavorably high surface energy from the sustained elongation of the jet.¹⁶

Electrowetting on PDMS surface has been reported previously in the literature.¹⁷ The resulting fluid motion can be enhanced if an immiscible liquid preferentially wets the PDMS surface

and acts as a lubricant for the other liquid. In this case, the oil phase would have served as a lubrication layer while the aqueous jet skated above, subject to a continuous-momentum interfacial boundary condition. In addition, we note that the interfacial curvature between the secondary aqueous jet and the oil phase was neither symmetrical nor stable, suggesting that the driving force may be of charge-related origin. The redistribution of ions at the W-O interface can affect the local electric field and interfacial energy: the ejection process of a primary droplet causes a sudden loss of cations in the primary jet and the temporal response from the electrostatic imbalance can lead to additional interactions with the orifice walls, electrified from electrowetting. Although it appears illogical that electrowetting should occur after the pulse termination while the voltage continued to decay, both the accumulation and the dissipation of electrical charges are expected to be significantly slower than those of an aqueous solution since PDMS is a dielectric material of high permittivity ($\epsilon = 2.75$ at 100 Hz). If the secondary protrusion was not associated with a surface-related phenomenon, we would not have observed it more consistently in channel constrictions of small equivalent radii, which have high surface-to-volume ratios. Indeed, in our previous report⁷ where an orifice of much larger cross section was used and the jet protrusion typically occupied only a fraction of the orifice diameter, the secondary protrusion was not observed under similar conditions. In addition, when the amplitude of a voltage pulse was increased, the magnitude of the secondary protrusion in a small orifice also increased, and multiple protrusions and breakups of the jet were sometimes observed. These multiple protrusions may be associated with capillary waves¹⁸ and the instabilities of the aqueous phase after an excitation, i.e., a sudden input of high energy from an electric pulse.

Using an orifice of high hydrodynamic resistance for electro-generation of droplets therefore has an important trade-off: because the primary jet must counter more hydrodynamic resistance, higher voltage and/or longer pulse duration is required, which leads to a corresponding increase in the likelihood of electrowetting behavior. The secondary jet protrusion as well as the associated droplet detachment became common when an orifice of a small equivalent radius, typically less than 5 μm , was used.

Effects of Long Pulse Duration on Vertex Motion and Droplet Formation

Figure 4A shows the trajectory of the vertex as a function of time when voltage pulses of 50-ms duration but of various amplitudes (200–450 V) were applied. The dimension of the orifice was $6 \times 4.5 \times 67 \mu\text{m}^3$. The voltages in Figure 4A were much lower compared to the breakup voltage for the same orifice listed in Table 1, owing to the longer duration of the applied pulses; breakup of the jets was observed at 500 V. Upon the initiation of the pulses, the vertices steadily advanced into the orifice. For low voltages (200–300 V) the displacements of the vertices eventually reached equilibrium, and the vertices remained at those positions until the pulses were terminated at 50 ms. Upon termination of the pulses, the vertices retracted back toward the water entrance of the orifice as expected. For high voltages (400–450 V), however, within the pulse duration of 50 ms the vertices did not reach their equilibrium positions.

Figure 4B shows a manifestation of the interaction of an electrified jet with the walls of an orifice. The advancement of the vertex in Figure 4B, which was obtained using an orifice of $3 \times 4.1 \times 78 \mu\text{m}^3$ and a 700-V pulse (50-ms duration), showed two small plateaus. These plateaus were caused by a “wiggling” motion of the vertex as the cusp pointed towards the sidewalls of the orifice temporarily rather than in the axial direction of the orifice. This observed temporal instability is likely caused by electrical charging and discharging of the jet and the PDMS walls of the channels.

Figure 5 is a sequence of images showing the production of a single droplet in an orifice before the termination of a long voltage pulse (1000 V, 100 ms). At ~ 21 ms, the vertex reached its maximum displacement, and a neck started to form on the jet. A water plug was detached from

the jet at 23 ms. By maintaining the amplitude of the pulse at the threshold of droplet formation, a very weak secondary protrusion was observed: the jet had a very small secondary advancement from 106 ms (6 ms after the trailing edge of the pulse) to 119 ms (19 ms after the pulse). The formation of a droplet in Figure 5 was not due to the artificial disturbance introduced by terminating the pulse since the droplet was formed at 1/5 of the pulse duration, but the intrinsic instability of an elongated jet.¹⁶ The voltage applied was insufficient either to drive an excess supply of water into the jet such that the jet could continue the cycle of re-growth and droplet shedding, or to cause enough electrowetting effects for a strong secondary protrusion. Thus there was neither a breakup of the jet upon termination of the pulse nor the formation of a secondary droplet afterwards. At a higher voltage, there can be multiple oscillations and breakups (even flooding) of a jet before and after the termination of the pulse. The periodic fluctuations of a jet before the end of a pulse can be caused by either (1) an imbalance between the fluid flowing in and out of the Taylor cone or the cone vertex,¹⁹⁻²¹ or (2) jet breaking into discrete droplets²⁰ as a result of Rayleigh instability.¹⁶

Substrate and Phase Selection

Although PDMS is a highly versatile material for prototyping, it is not universally compatible with organic solvents.²² Soybean oil was used in this study in part for its negligible swelling effect (~2% over 24 hr). Silicone oil (AR 20) was found to cause ~10% swelling over 24 hr, and this swelling can lead to issues of channel constriction if small channels ($r_c \leq 5 \mu\text{m}$) are used. This is not a serious issue for large orifices.

Figure 6 shows the evolution of the water-oil interface when silicone oil AR 20 (with 3% Span 85) was used as the organic phase. The orifice dimension in this case is $6 \times 7.7 \times 80 \mu\text{m}^3$. In the absence of any electrical pulse, the W-O interface at the left side of the orifice changed from very well-defined and symmetric (0–11 min) to asymmetric (21 min) to very diffuse and rough (30 min). Attempts to reposition the interface (30 min) by balancing pressures resulted in water jetting into the orifice from the upper left corner of the entrance to the orifice. The walls of the orifice also exhibited distortion.

CONCLUSION

Aqueous droplets generated from the secondary protrusions may be more suitable for applications in microscale chemical reactions. The droplets ejected from primary jets tend to have relatively high velocities, and thus may be difficult to capture and manipulate. In addition, the droplets formed from primary jets can be electrically charged. The formation of secondary droplets was reproducible provided that orifices of sufficiently small equivalent radii were used and when appropriate pulse amplitudes and durations were applied. The volume of a secondary droplet was from femtoliters to a few picoliters. Although the volume of a secondary droplet was in general larger than that of the primary droplet, it is sufficiently small for encapsulating subcellular structures and for microscale reactions and assays. The velocity of a secondary droplet can be further controlled by applying additional pulses at voltages below the threshold of droplet formation, in essence using the voltage-induced displacement of the immiscible interface as a pressure source to push the droplet out of the orifice.

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Figure 1.

Schematic of the experimental setup. A micropipette holder/electrochemical half-cell was used in the water line to directly connect the anode of the electric pulse generator to the water phase. Due to the insulating nature of the oil phase, experimentally we did not observe any difference whether the ground electrode (cathode) of the pulse generator was directly in contact with the oil phase. The dashed enclosure shows the dimension of the orifice with depth of D , width of W , and length of L .

Figure 2.

(A) A sequence of images showing the electro-generation of water droplets with a pulse amplitude of 1000 V and a duration of 10 ms in an orifice of $6(D) \times 6.7(W) \times 63(L) \mu\text{m}^3$. The droplet at the exit (right) side of the orifice in the first image was formed by the previous pulse. The scale bar represents 10 μm and the volume of the droplet in the last image is 420 fL. (B) The profile of the displacement of the vertex (circles) and the droplet/plug (triangles). The start of the voltage pulse was at 0 ms and the termination was at 10 ms as indicated by the dashed line. The immiscible phase was soybean oil with 3% wt Span 85.

Figure 3.

(A) A sequence of images showing the electro-generation of water droplets with a pulse amplitude of 800 V (10-ms duration) in an orifice of $6(D) \times 6.7(W) \times 63(L) \mu\text{m}^3$. The voltage of the applied pulse was barely enough to eject a primary droplet. No secondary protrusion or secondary droplet ejection was observed under this condition. The scale bar in the last image represents 10 μm . (B) Profile of the displacement of the vertex (circles) and the droplet/plug (triangles). The pulse started at 0 ms and ended at 10 ms as indicated by the dashed line. The immiscible phase was soybean oil with 3% wt Span 85.

Figure 4.

Plots of the displacement of the vertex versus time in: **(A)** an orifice of $6(D) \times 4.5(W) \times 67(L) \mu\text{m}^3$ at various voltages (50-ms duration), and **(B)** an orifice of $3(D) \times 4.1(W) \times 78(L) \mu\text{m}^3$ at 700 V (50-ms duration). The immiscible phase was soybean oil with 3% wt Span 85.

Figure 5.

A sequence of images showing the electro-generation of droplets using a long voltage pulse (1000 V, 100 ms) in an orifice of $15(D) \times 9(W) \times 92(L) \mu\text{m}^3$. The immiscible phase was silicone oil AR 20 with 3% wt Span 85. The water plug looks larger at 23–26 ms than those in later frames due to the non-square geometry of the orifice (the water plug may not have occupied the full depth of the orifice) and the rearrangement of the detached droplet into a sphere/spheroid. The scale bar in the last image represents 20 μm .

Figure 6.

Observed changes in the water-oil interface due to the slight swelling of PDMS by silicone oil. The immiscible phase was silicone oil AR 20 with 3% wt Span 85 and the orifice dimension was $6(D) \times 7.7(W) \times 80(L) \mu\text{m}^3$. The two phases were brought to contact at 0 min and the meniscus of water remained sharply defined and symmetric up to 11 min. The interface became asymmetric at 21 min, with the vertex being closer to the lower wall. At 30 min, the interface was very rough and could not form a meniscus. Attempts to re-position the interface by balancing the pressures resulted in water jetting into the orifice from the upper left corner of the entrance. No electric pulses were applied in this experiment.

Summary of experimental conditions and observations of electro-generation of aqueous droplets in orifices of different geometries. The organic phase in all experiments listed was soybean oil with 3% Span 85. r_e is the equivalent radius, P_L is the Laplace pressure, and $u_{\max}$ is the maximum advancement velocity of the vertex.

Orifice dimension ($D \times W \times L$, μm^3)	r_e (μm)	P_L (kPa)	Ease of stabilizing interface	Representative pulse	$u_{\max}$ (mm/s)	Breakup before or at end of pulse	Breakup long after pulse
$6 \times 10.5 \times 68$	3.8	12	not very easy	1000 V, 10 ms	6.1	yes	yes
$6 \times 6.7 \times 63$	3.2	14	easy	1000 V, 10 ms	4.7	yes	yes
$6 \times 4.5 \times 67$	2.6	18	easy	1000 V, 10 ms	4.7	yes	yes
$3 \times 10.5 \times 78$	2.3	20	easy	1000 V, 10 ms	2.1	yes	yes
$3 \times 8.5 \times 78$	2.2	21	easy	1000 V, 10 ms	1.7	sometimes	yes
$3 \times 4.1 \times 78$	1.7	26	very easy	1000 V, 20 ms	0.95	sometimes	yes
$3 \times 4.1 \times 24$	1.7	26	easy	800 V, 20 ms	3.1	many	yes