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Precise Electrohydrodynamic Direct-Write Micro-Droplets Based on a Designed Sinusoidal High-Voltage AC Power

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Abstract: The precise manufacturing of micro/nano structures is the key to the rapid development of flexible micro/nano systems. In this paper, a sinusoidal high-voltage alternating current (AC) power is designed for electrohydrodynamic direct-writing (EDW) technology. A push-pull converting circuit is utilized as the direct current (DC) voltage regulator power of a full-bridge inverter circuit. A single-phase full-bridge inverter circuit is used to output the controllable AC voltage, which is then boost-filtered to output the high-voltage sinusoidal AC signal. The amplitude of the output sinusoidal voltage is proportional to the input voltage and the modulation degree of the sinusoidal pulse width modulation (SPWM) inverter circuit. Then, the designed sinusoidal high-voltage AC power is used in the AC EDW process to print micro-droplets. The deposition frequency and the average diameter of droplets can be effectively controlled by adjusting the voltage amplitude and the voltage frequency. The design of this sinusoidal high-voltage AC power will promote research on the applications of EDW technology in the field of micro/nano manufacturing.

Keywords: electrohydrodynamic direct-writing; sinusoidal high-voltage AC power; micro/nano structure; precise deposition

1. Introduction

The rapid development of flexible micro/nano systems has enabled higher requirements for the precise integration printing of micro/nano structures [1–4]. Compared to the traditional ink-jet printing technology driven by internal pressure, electrohydrodynamic direct-writing (EDW) technology applies an external electric field force to stretch the polymer solution into a fine jet. The stable motion stage of the jet is achieved by reducing the distance between the nozzle and collector for the accurate controllable deposition of a micro/nano structure, which has become an important potential method for the application of organic and flexible device manufacturing with great advantages of high resolution, simple process and good material compatibility [5,6].

EDW technology uses an external electric field to deform the viscous solution into a Taylor cone for the printing of micro/nano structures. When the electric field force on the Taylor cone is large enough to overcome the surface tension of the liquid, the jet is ejected from the tip of the Taylor cone and deposited onto the collector to achieve micro/nano patterns [7,8]. Coppola et al. [9,10] promoted

pyro-electrohydrodynamic (EHD) technology to achieve the precise fabrication of microfluidic footpaths and applied it to obtain individual polymer droplets. In recent years, the AC electric field has been introduced into the EDW process for the fabrication of precise micro/nano structures [11,12]. The AC electric field can reduce the accumulation of the charges on the nozzle by neutralizing the electric polarity, which can help to suppress the charge repulsion interferences under the DC electric field and promote jet stability and deposition precision [13–15]. Under the AC electric field, the free charges reciprocate along the jet and the polarity of the electric field force on the jet changes periodically, then micro/nano droplets can be obtained at a corresponding frequency [16,17]. Wei et al. [18] applied a new high-resolution AC-pulse modulated electrohydrodynamic (EHD) jet printing technology to obtain the drop-on-demand fabrication. Guo et al. [8] optimized the effect of process parameters on ejection volume in electrohydrodynamic jet printing under the AC electric field to obtain the high printing resolution of the deposition structures. In the previous works of our group [19], the AC power of square-wave voltage has been applied in EDW technology for the printing of micro-droplets and has indicated the effects of voltage frequency on the deposition frequency and the diameter of droplets for polymer solutions with various conductivities. However, the sharp edge of the voltage wave is likely to cause instability in the early stages of the ejection process.

The AC power of sinusoidal voltage without sharp edges has been used for the precise printing of micro/nano structures. Nguyen et al. [20] applied an AC sinusoidal voltage for the continuous ejection of droplets, proving that the sinusoidal alternating electric field provides a good way to suppress the electrical breakdown and solution atomization of the EDW jet. S. Maheshwari et al. [21] and R. Kessicka et al. [22] indicated that the sinusoidal alternating electric field is beneficial in weakening the jet whipping, reducing the instability of jet motion, and improving the deposition accuracy of micro/nano structures. At present, sinusoidal AC power has been widely used in various fields [23]. However, the integrated development of AC power with a high output voltage, low output current, and low power is still an obstacle for its further application in EDW technology.

In this paper, a sinusoidal high-voltage AC power is designed to meet the requirements of AC EDW technology. The effects of the voltage amplitude and voltage frequency on the average droplet diameter and deposition frequency are also investigated.

2. Materials and Methods

An experimental system for AC EDW is built, as in Figure 1. The AC high-voltage power is connected to the spinneret, and then a high-voltage electric field is generated between the spinneret and the collector. The solution is supplied to the spinneret by a precision syringe pump (Harvard Pump 11 Elite, USA). A silicon wafer is used as the collecting plate and fixed on the XY motion platform. The host computer is used to control the motion trajectory and the speed of the platform. The CCD camera (UI-2250-C, IDS Imaging Development Systems GmbH, Obersulm, Germany) is used to observe and analyze the ejection and deposition process of the charged jet. Polyethylene oxide (PEO, $M_w = 300,000$ g/mol) solution with concentration of 8 wt% is used as the EDW material, where the solvent is a mixture of deionized water and ethanol with volume ratio of 3:1 ($v:v$).

A sinusoidal high-voltage AC power (0~4 kV, 0~2 kHz) is designed, as shown in Figure 2, to meet the demand of AC EDW technology. The voltage of the DC converting circuit serves as the input voltage of the single-phase full-bridge inverter circuit when the switching source sends a signal. Compared with the DC regulated power for the full-bridge inverter circuit, the DC converting circuit can effectively suppress the leakage inductance and the harmonic distortion factors. Thus, the DC converting circuit can be used to achieve the adjustable output voltage and to reduce the total harmonic distortion (THD). The voltage of the DC converting circuit will be the input voltage of a single-phase full-bridge inverter circuit, then the controllable AC voltage is output under the control of pulse width modulation (PWM). After that, the AC signal is boost-filtered to output a sinusoidal high-voltage AC signal. Aiming at the AC power source with low power for EDW, the PWM and sinusoidal pulse width modulation (SPWM) control modes are adopted to achieve a controllable sinusoidal alternating

voltage. The sampling circuit monitors the voltage waveform in real-time and the control signals can be sent to SPWM and PWM through the host computer.

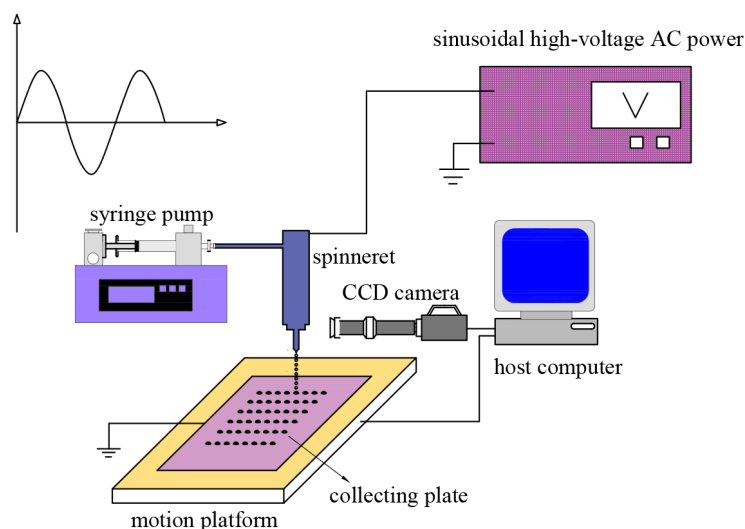


Figure 1. Alternating current (AC) electrohydrodynamic direct-write experimental system.

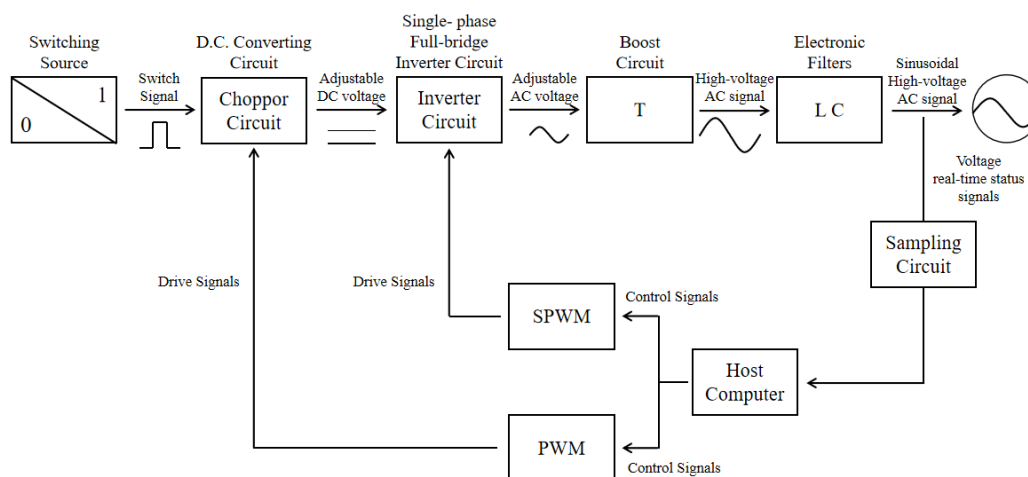


Figure 2. Design of the sinusoidal high-voltage AC power system.

The push-pull DC converting transformer core has the features of a high utilization rate, a large output power and a small output ripple voltage, which can meet the requirements for the application in AC EDW. The schematic diagram of the push-pull DC converting circuit is shown in Figure 3. A switching power (48 V, 250 W) is used as an input power. VT1 and VT2 are two metal-oxide-semiconductor field-effect-transistor (MOSFET) power switches of the same type with the characteristics of IRF640N, which are switched in turn under the PWM signals high-side output (HO) drive signal and low-side output (LO) drive signal with a different phase of 180° and a duty ratio of less than 50%. Fast recovery diodes (FRD) with the characteristics of MUR1100E are selected to function with the rectified effect and freewheeling effect. The energy storage inductor L is 0.5 H and the filter capacitor C_o is $18.8 \mu\text{F}$, realizing the continuous inductor current and low output voltage ripple.

The inverter is used to convert the DC electric energy into AC electric energy, and a single-phase full-bridge is used as the topology of the inverter circuit, as shown in Figure 4. The MOSFETs of IRF840 with low on-resistance and fast on-off response are selected as the power switching tubes. An inductance of 100 mH and two parallel polypropylene capacitors of 10 nF are used to make up the inductance-capacitance (LC) low-pass filter to filter out the high frequency harmonics. SPWM signals

HO_L and HO_R with dead time are complementary in phase. The alternating switching of the power switching tubes is controlled by the SPWM signal, and the straight-through phenomenon is eliminated due to the presence of the dead time. Then, the sinusoidal AC voltage can be obtained through the low-pass LC filtering.

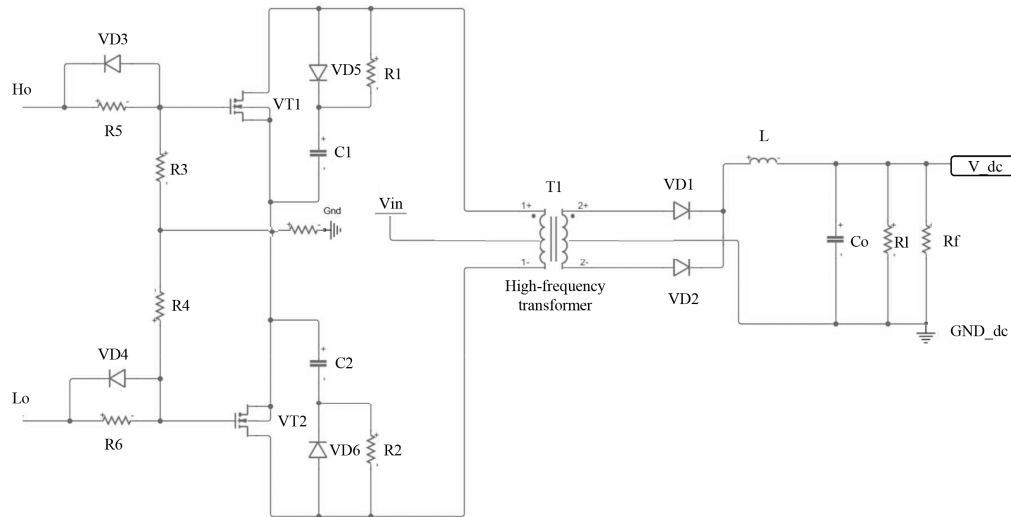


Figure 3. Schematic diagram of the push-pull direct current (DC) conversion circuit.

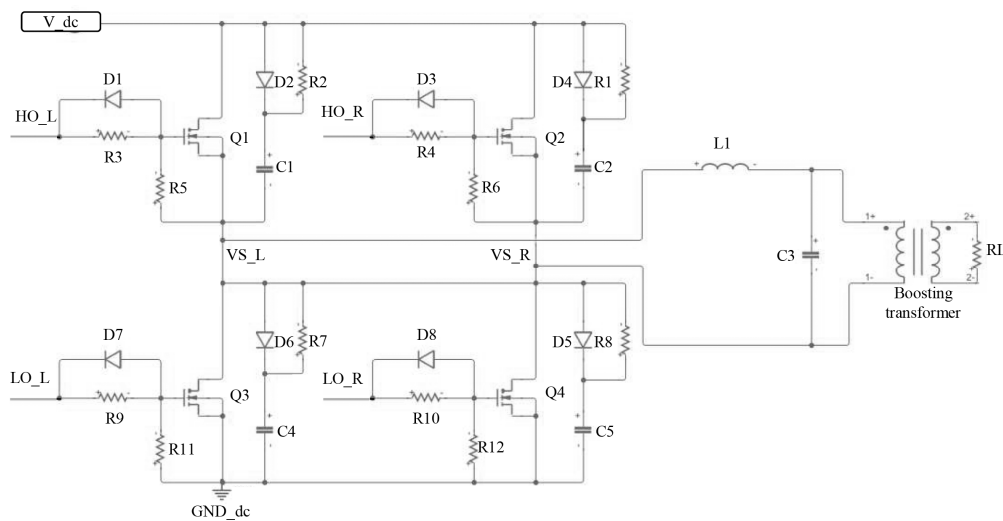


Figure 4. Schematic diagram of the single-phase full-bridge voltage type sinusoidal pulse width modulation (SPWM) inverter circuit.

A controllable sinusoidal high-voltage AC voltage can be obtained successfully by transforming the input voltage through the inverter circuit, the boost circuit and the control circuit. The influence of modulation degree on the sinusoidal alternating voltage is shown in Figure 5. The sinusoidal output voltage increases with the increase in modulation degree at a certain proportion.

The effects of the modulation degree on the peak of sinusoidal output voltage and THD at different input voltages and frequencies have been investigated, as shown in Figure 6. The results demonstrate that the peak value of sinusoidal output voltage is proportional to the input voltage and modulation degree of the SPWM inverter circuit. The controllable output voltage can be realized by changing the input voltage or modulation degree.

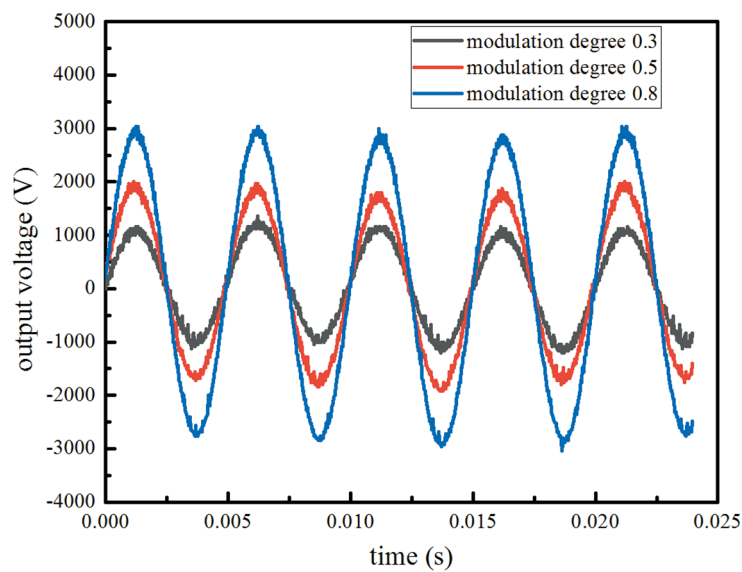


Figure 5. Sinusoidal output voltage waveforms at different modulation degrees.

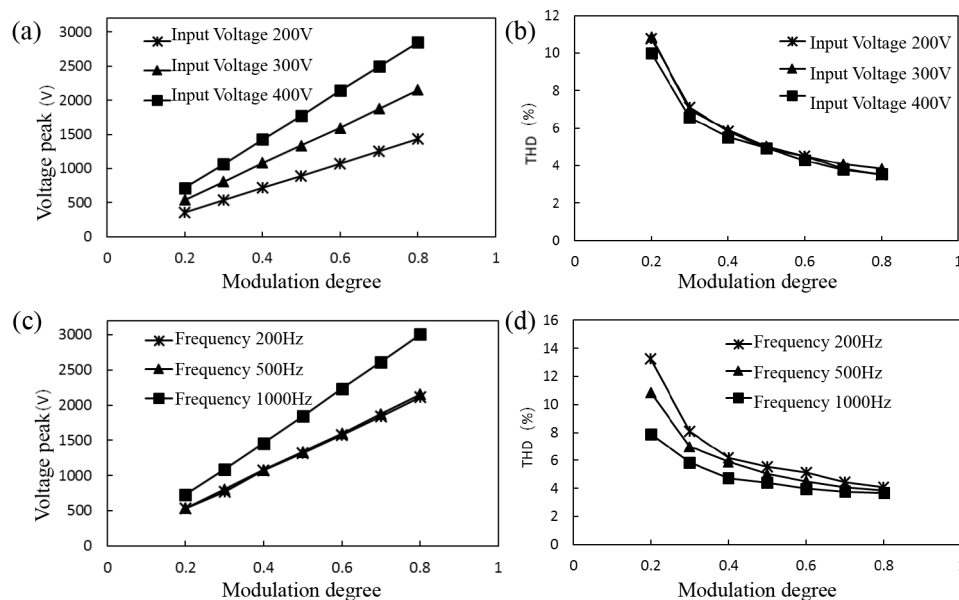


Figure 6. The peak of sinusoidal output voltage and total harmonic distortion (THD) vary with modulation degree. (a,b) Frequency 500 Hz, load 50 M Ω ; (c,d) Input voltage 300 V, load 50 M Ω .

3. Results and Discussion

The designed sinusoidal high-voltage AC power is utilized in the AC EDW experiments to verify its application in the accurate printing of micro-droplets. The effect of the amplitude of AC voltage on the average diameter and deposition frequency of droplets is investigated, as shown in Figure 7. Under the AC electric field, the solution is stretched into a Taylor cone. When the electric field force is strong enough to overcome the solution surface tension, a charged jet will be ejected from the tip of the cone. The Taylor cone will be stretched and shrunk periodically with the transformation of the positive voltage and negative voltage of the AC electric field, then the stable printing of micro-droplets can be realized on the silicon substrate. The relationships between average diameter, the deposition frequency of printed droplets and voltage amplitude are shown in Figure 8. As the voltage amplitude increases, the average diameter of the droplets will rise in a curve. As the voltage increases from 1520 to 1790 V, the periodic shrinkage of the jet experiences relatively significant changes, resulting in the

deposition frequency of droplets decreasing from 150 to 130 Hz, while the average diameter of the droplets increases from 100 to 125 μm under the same variable of the voltage.

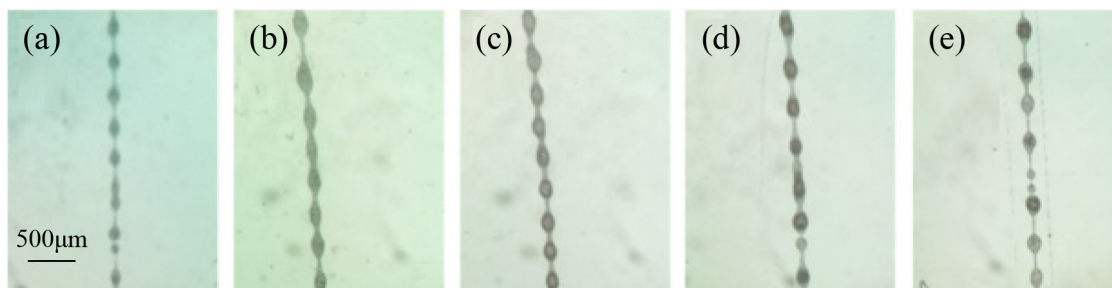


Figure 7. Direct-write micro-droplets on a silicon substrate under different voltage amplitudes: (a) 1523; (b) 1583; (c) 1647; (d) 1719; and (e) 1792 V. Voltage frequency, solution supply rate, nozzle inner diameter, distance between the nozzle and collector, and platform moving speed are 200 Hz, 60 $\mu\text{L/h}$, 60 μm , 3 mm, and 50 mm/s, respectively.

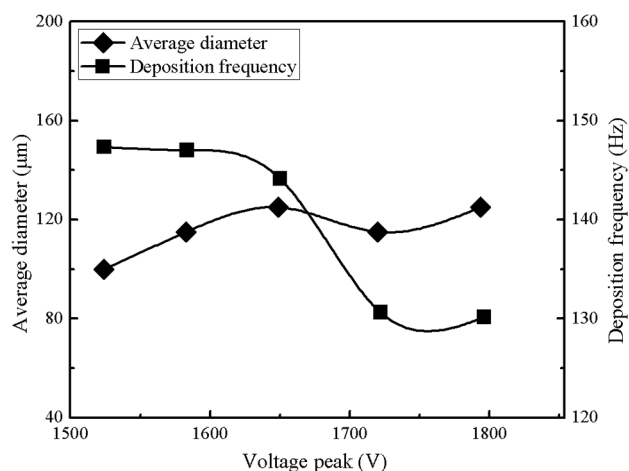


Figure 8. The effects of average droplet diameter and deposition frequency by voltage amplitude. Voltage frequency, solution supply rate, nozzle inner diameter, distance between nozzle and collector, and platform moving speed are 200 Hz, 60 $\mu\text{L/h}$, 60 μm , 3 mm, and 50 mm/s, respectively.

Then, the direct-write micro-droplets under different AC voltage frequencies are also explored, as shown in Figure 9. With the increase in voltage frequency, the alternation of electric field polarity is accelerated, and the Taylor cone suffers a shorter period of stretching and shrinking. Thus, the ejecting frequency of the droplet increases with the voltage frequency. The effects of voltage frequency on the average diameter and deposition frequency of droplets are analyzed in Figure 10. As the voltage frequency increases from 200 to 500 Hz, the average diameter of the droplets decreases from 207 to 143 μm under the constant solution supply rate, which is negatively correlated to the voltage frequency. While the deposition frequency of the droplets increases from 175 to 375 Hz, which is proportional to the voltage frequency. The effects of AC voltage frequency on printed micro-droplets correspond to the AC EDW experiments conducted by the AC power of square-wave voltage, verifying the applicability of the designed sinusoidal high-voltage AC power for the precise printing of micro-droplets [19].

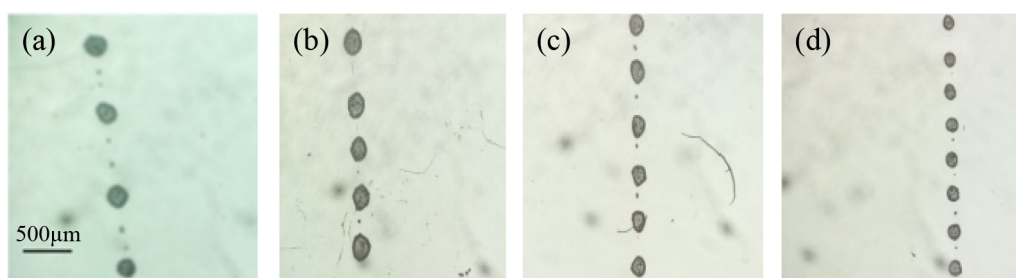


Figure 9. Direct-write micro-droplets on a silicon substrate under different voltage frequency: (a) 200; (b) 300; (c) 400; and (d) 500 Hz. Voltage amplitude, solution supply rate, nozzle inner diameter, distance between nozzle and collector, and platform moving speed are 1807 V, 60 $\mu\text{L/h}$, 60 μm , 3 mm, and 50 mm/s, respectively.

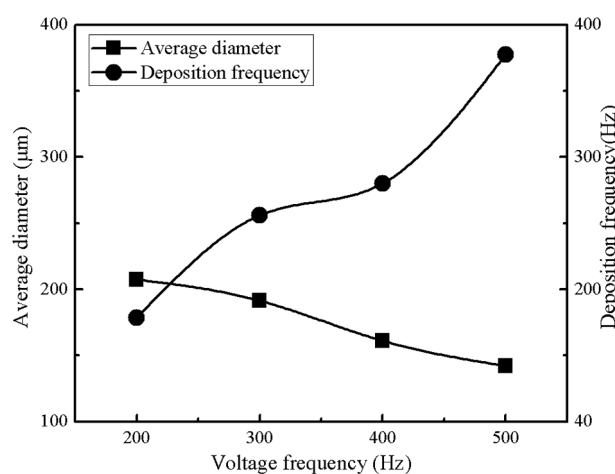


Figure 10. The effects of average droplet diameter and deposition frequency by voltage frequency. Voltage amplitude, solution supply rate, nozzle inner diameter, distance between the nozzle and collector, and platform moving speed are 1807 V, 60 $\mu\text{L/h}$, 60 μm , 3 mm, and 50 mm/s, respectively.

4. Conclusions

In this paper, a sinusoidal high-voltage AC power is developed for the AC EDW. The SPWM control method is adopted to obtain the AC power with a high output voltage and low power. The output sinusoidal voltage can be controlled by changing the input voltage and modulation degree. The AC EDW experiments are carried out and the influences of voltage amplitude and voltage frequency on the printed micro-droplets are investigated to verify the applicability of the designed sinusoidal high-voltage AC power. The deposition frequency of the droplets increases with the AC voltage frequency and decreases with the voltage amplitude. The average diameter of the droplets is negatively correlated to the AC voltage frequency and the voltage amplitude. This work helps to build the foundation for AC EDW technology and facilitate the future application of printed micro/nano structures for flexible electronic systems.

Author Contributions: H.C., G.Z., J.L., J.J., X.W., G.K., W.L., Y.L. conceived and designed the experiments; H.C., J.J., G.K. and X.W. performed the experiments; H.C., G.Z., Y.L. and J.J. analyzed the data; H.C., G.Z., J.J., X.W., W.L. and Y.L. wrote the manuscript. All authors contributed to the writing. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Nagle, A.R.; Fay, C.D.; Wallace, G.G.; Xie, Z.; Wang, X.; Higgins, M.J. Patterning and process parameter effects in 3d suspension near-field electrospinning of nanoarrays. *Nanotechnology* **2019**, *30*, 495301. [[CrossRef](#)] [[PubMed](#)]
2. Zheng, G.F.; Li, W.W.; Wang, X.; Wu, D.Z.; Sun, D.H.; Lin, L.W. Precision deposition of a nanofibre by near-field electrospinning. *J. Phys. D Appl. Phys.* **2010**, *43*, 6. [[CrossRef](#)]
3. Lopez-Covarrubias, J.G.; Soto-Munoz, L.; Iglesias, A.L.; Villarreal-Gomez, L.J. Electrospun nanofibers applied to dye solar sensitive cells: A review. *Materials* **2019**, *12*, 3190. [[CrossRef](#)] [[PubMed](#)]
4. Varga, M.; Morvan, J.; Diorio, N.; Buyuktanir, E.; Harden, J.; West, J.L.; Jáklí, A. Direct piezoelectric responses of soft composite fiber mats. *Appl. Phys. Lett.* **2013**, *102*, 153903. [[CrossRef](#)]
5. Chen, J.Z.; Wu, T.; Zhang, L.B.; Feng, X.W.; Li, P.; Huang, F.L.; Zuo, C.C.; Mao, Z.P. Fabrication of flexible organic electronic microcircuit pattern using near-field electrohydrodynamic direct-writing method. *J. Mater. Sci. Mater. Electron.* **2019**, *30*, 17863–17871. [[CrossRef](#)]
6. Zheng, G.F.; Jiang, J.X.; Wu, D.Z.; Sun, D.H. Near-field Electrospinning. In *Electrospinning: Nanofabrication and Applications*; William Andrew Publishing: Norwich, NY, USA, 2019; Volume 5, pp. 283–319.
7. Li, K.; Wang, D.; Yi, S.; Jia, H.; Qian, J.; Du, Z.; Ren, T.; Liang, J.; Martinez-Chapa, S.O.; Madou, M. Instrument for fine control of drop-on-demand electrohydrodynamic jet printing by current measurement. *Rev. Sci. Instrum.* **2019**, *90*, 115001. [[CrossRef](#)]
8. Guo, L.; Duan, Y.; Huang, Y.; Yin, Z. Experimental study of the influence of ink properties and process parameters on ejection volume in electrohydrodynamic jet printing. *Micromachines* **2018**, *9*, 522. [[CrossRef](#)]
9. Coppola, S.; Nasti, G.; Todino, M.; Olivieri, F.; Vespini, V.; Ferraro, P. Direct writing of microfluidic footpaths by pyro-ehd printing. *ACS Appl. Mater. Interfaces* **2017**, *9*, 16488–16494. [[CrossRef](#)]
10. Coppola, S.; Mecozzi, L.; Vespini, V.; Battista, L.; Grilli, S.; Nenna, G.; Loffredo, F.; Villani, F.; Minarini, C.; Ferraro, P. Nanocomposite polymer carbon-black coating for triggering pyro-electrohydrodynamic inkjet printing. *ACS Appl. Mater. Interfaces* **2015**, *106*, 261603. [[CrossRef](#)]
11. Lyu, H.; Zhang, X.; Liu, F.; Huang, Y.H.; Zhang, Z.; Jiang, S.; Qin, H.T. Fabrication of micro-scale radiation shielding structures using tungsten nanoink through electrohydrodynamic inkjet printing. *J. Micromech. Microeng.* **2019**, *29*, 9. [[CrossRef](#)]
12. Soldate, P.; Fan, J. Controlled deposition of electrospun nanofibers by electrohydrodynamic deflection. *J. Appl. Phys.* **2019**, *125*, 15. [[CrossRef](#)]
13. Stanishevsky, A.; Yager, R.; Tomaszewska, J.; Binczarski, M.; Maniukiewicz, W.; Witonska, I.; Lukas, D. Structure and mechanical properties of nanofibrous zro2 derived from alternating field electrospun precursors. *Ceram. Int.* **2019**, *45*, 18672–18682. [[CrossRef](#)]
14. Farkas, B.; Balogh, A.; Cselko, R.; Molnar, K.; Farkas, A.; Borbas, E.; Marosi, G.; Nagy, Z.K. Corona alternating current electrospinning: A combined approach for increasing the productivity of electrospinning. *Int. J. Pharm.* **2019**, *561*, 219–227. [[CrossRef](#)]
15. Schubert, M.; Rasche, J.; Laurila, M.M.; Vuorinen, T.; Mantysalo, M.; Bock, K. Printed flexible microelectrode for application of nanosecond pulsed electric fields on cells. *Materials* **2019**, *12*, 2713. [[CrossRef](#)]
16. Liu, J.; Lin, Y.H.; Jiang, J.X.; Liu, H.Y.; Zhao, Y.; Zheng, G.F. Bead-on-string structure printed by electrohydrodynamic jet under alternating current electric field. *Appl. Phys. A Mater. Sci. Process.* **2016**, *122*, 6. [[CrossRef](#)]
17. Qin, H.T.; Dong, J.Y.; Lee, Y.S. Ac-pulse modulated electrohydrodynamic jet printing and electroless copper deposition for conductive microscale patterning on flexible insulating substrates. *Robot. Comput. Integr. Manuf.* **2017**, *43*, 179–187. [[CrossRef](#)]
18. Wei, C.; Qin, H.T.; Ramirez-Iglesias, N.A.; Chiu, C.P.; Lee, Y.S.; Dong, J.Y. High-resolution ac-pulse modulated electrohydrodynamic jet printing on highly insulating substrates. *J. Micromech. Microeng.* **2014**, *24*, 9. [[CrossRef](#)]
19. Jiang, J.X.; Zheng, G.F.; Wang, X.; Zheng, J.Y.; Liu, J.; Liu, Y.F.; Li, W.W.; Guo, S.M. Printing of highly conductive solution by alternating current electrohydrodynamic direct-write. *J. Phys. Conf. Ser.* **2018**, *986*, 012027. [[CrossRef](#)]
20. Nguyen, V.D.; Byun, D. Mechanism of electrohydrodynamic printing based on ac voltage without a nozzle electrode. *Appl. Phys. Lett.* **2009**, *94*, 3. [[CrossRef](#)]

21. Maheshwari, S.; Chang, H.C. Assembly of multi-stranded nanofiber threads through ac electrospinnig. *Adv. Mater.* **2009**, *21*, 349–354. [[CrossRef](#)]
22. Kessick, R.; Fenn, J.; Tepper, G. The use of ac potentials in electrospraying and electrospinning processes. *Polymer* **2004**, *45*, 2981–2984. [[CrossRef](#)]
23. Gao, N.; Zhang, M.; Zhang, J.C. Ac electroluminescent processes in pr³⁺-activated (ba_{0.4}ca_{0.6})tio₃ diphas polycrystals. *Materials* **2017**, *10*, 565. [[CrossRef](#)] [[PubMed](#)]



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