

Ultrasonic treatment of aerosol jet printed traces

Research Article

Marcin Korzeniowski^{1*}, Marcin Winnicki¹, Bartosz Swiadkowski², Wojciech Łapa¹

¹ Wrocław University of Technology: Faculty of Mechanical Engineering, Wrocław, Poland

² Wrocław University of Technology: Faculty of Electronics, Photonics and Microsystems, Wrocław, Poland

Received 22 July 2024; Accepted 30 September 2024

Abstract: Aerosol jet printing (AJP) is a contactless direct-write approach aimed at the production of fine features on a wide range of substrates. The technology has been explored for a variety of applications, including active and passive electronic components, actuators, sensors, and a variety of selective chemical and biological responses. However, the quality of conductive traces printed with nanoparticle inks using AJP can be affected by several factors, including carrier and sheath gas flow rate, ink properties, and substrate material properties. A typical defect present in the prints, such as non-uniform metal particle distribution, solvent local concentration, porosity, delamination, and bubbles, can be reduced by additional ultrasonic post-printing treatment. Therefore, the article investigates the influence of the self-designed sonotrode tool on the quality of conductive traces printed with nanoparticle inks using AJP. An ultrasonic head was fixed in a tandem position behind a printing nozzle at a distance equal to the node of ultrasonic waves. In this article, it was found that ultrasound has a positive effect on the quality of printed traces by improving degassing with simultaneous uniform particle distribution. As a result, the surface open porosity obtained for printed and ultrasonically treated traces was decreased almost seven times from 17.2 to 2.7%, respectively. An improved surface and structural morphology increased electrical resistivity in the prints from 6.85 to 4.57 $\mu\Omega$ cm. The analysis included quantifying the macroscale geometry, electrical properties, and micromorphological characteristics of the traces. The results of this article suggest that the application of ultrasonic-assisted aerosol jet printing with a proper tool improves the quality of AJP prints.

Keywords: Flexible printed electronics • Ultrasonic treatment • Polymer foil • Printed conductive traces • Silver ink

1. Introduction

The field of additive manufacturing has witnessed remarkable advances in recent years, revolutionising traditional manufacturing processes. Central to this transformation is the field of flexible printed electronics, which has emerged as a key enabler of versatile, cost-effective, and environmentally sustainable electronic systems [1,2]. The demand for flexible, lightweight, and customisable electronic devices has never been greater. From portable health monitors to foldable smartphones, the convergence of electronics with flexibility has initiated a new field of applications [3,4]. A fundamental shift in the manufacturing landscape has propelled flexible printed electronics to the forefront of technological progress. One of the most transformative developments within this domain is aerosol jet printing (AJP), a cutting-edge, three-dimensional (3D) additive manufacturing

technique that is revolutionising how we design and produce electronic circuits on flexible substrates [5–7].

AJP involves the deposition of functional materials on a substrate using a precisely controlled aerosol stream [8]. This technique enables the fabrication of complex and intricate structures with fine feature sizes and high resolution, making it suitable for a wide range of applications, including electronics [9], sensors [10], and biomedical devices [11]. Therefore, the potential applications of AJP span across various industries. In the electronics sector, this technique can be used for the fabrication of flexible and stretchable electronic devices, antennas, and interconnects [12]. In the field of sensors, it offers opportunities for the development of high-performance gas sensors, biosensors, and environmental monitoring devices [13]. Furthermore, AJP can facilitate the precise deposition of biomaterials, opening avenues for biofabrication and tissue engineering applications [14]. However, challenges such as limited material compatibility, nozzle clogging, and

* E-mail: marcin.korzeniowski@pwr.edu.pl

low deposition rates have forced modifications to improve process performance and versatility [15]. One of the possible solutions is the integration of the assistance of the ultrasonic nozzle into the AJP. Ultrasonic waves are used to enhance the atomisation and transport of aerosol droplets [16,17]. Another option is subjecting the aerosol stream to ultrasonic vibrations in the nozzle of the printing head (PH) to control the droplet size distribution, resulting in improved print quality, increased deposition rates, and enhanced material compatibility [18]. However, the process limits and does not eliminate the presence of common defects in the printed traces, e.g. non-uniform metal particle distribution, reduced by solvent local metal concentration, pores, bubbles, etc. [19]. To solve the problem, the deposited traces should be treated with ultrasonic waves.

Ultrasonics affect particles in a liquid suspension through a process called acoustic cavitation [20]. Acoustic cavitation refers to the formation, growth, and implosive collapse of small gas- or vapour-filled bubbles (cavities) within a liquid when subjected to intense ultrasound waves. Rapid expansion and collapse of the cavities generate intense local shear forces and turbulence within the liquid. Mechanical agitation causes a more uniform dispersion of particles throughout the liquid and prevents agglomeration and sedimentation [21]. In addition, the high-pressure waves created during cavitation lead to the mechanical breakdown of agglomerated particles and are known as sonoporation or ultrasonic fragmentation. Ultrasonics generally provide three basic advantages: (i) the force is distributed over the entire surface and not only on a very small region, (ii) adhesion problems do not arise because no contact occurs, and (iii) many particles can be handled at the same time due to the periodic nature of a wave [22]. Taking into account the advantages in combination with the AJP process, smaller particles with a larger surface area and uniform distribution are sinterable well [23]. Furthermore, the additional energy provided by ultrasound can promote chemical reactions by breaking chemical bonds or overcoming activation energy barriers [24]. As a result, surfactants, polymers, and other chemical additives are easily removed.

It is well known that the forces of acoustic radiation acting on particles are larger in an ultrasonic standing wave than in a progressive wave, and therefore, standing waves are mainly used for particle manipulation [25]. Ultrasonic particle manipulation (UPM) technique is a non-contact label-free method for manipulating suspended in fluid micro and nanoscale particles using ultrasound. The UPM based on the standing wave principle has been widely used in many applications. Courtney et al. [26] proposed and demonstrated a method to trap 10 μm polystyrene particles

in a water-filled cavity and manipulate them in a two-dimensional plane using two orthogonal pairs of counter-propagating waves. These traps can be manipulated by appropriate adjustment of the system, e.g. sources of counter-propagating waves. Usually, suspensions with nanosized or submicrometre-sized particles appeared as homogeneous dispersed substances with a tendency to agglomeration. However, when ultrasound is applied, the particles move to the nodes of the standing wave field with a distance between the localised particles equal to half the wavelength [27]. It should be emphasised that aerosol droplets in AJP are non-uniform [28] with potential agglomeration of metal nanoparticles due to changes in ink properties, e.g. zeta potential [29]. Aerosol droplets quickly lose solvent upon contact with a dry carrier gas (CG), which results in polydispersity [30]. The larger droplets generally settle at the bottom of the ink reservoir. Therefore, gravitational settling is a positive feature in AJP, as it imposes a tighter filter on the droplet size [31]. However, a higher atomiser power produces a population of larger droplets, and inks with insufficient volatility will retain a larger droplet size, or worse, without proper metal packing [32]. Additionally, partly dried large agglomerates can be lifted and transported to the PH and further deposited on the trace, locally changing the concentration of metal particles. These phenomena can explain the presence of pores, cracks, and delamination found in traces printed by AJP [33–35]. In the printed graphene interconnects by Pandhi et al. [36], the breakdown of the pattern occurred mainly due to gases and solvents trapped within the interconnect, which expand or vaporise during heating and resulted in mechanical failure of the interconnect. Therefore, applying UPM after AJP can establish a uniform structure with shattered agglomerates at the submicronic or even nanoscale [37] and remove gas bubbles or solvent remnants prior to sintering [38].

Ultrasonic-assisted aerosol jet printing (UA-AJP) provides many benefits. First, it enables the printing of a broader range of materials, including highly viscous or nonatomisable inks, expanding the possibilities for functional material deposition. Second, the improved atomisation and transport efficiency leads to higher printing speeds, reducing the fabrication time of complex structures. Conversely, the enhanced control over particle distribution enables finer resolution and improved accuracy in the printed features. Therefore, in this article, the concept and potential of UA-AJP process with ultrasonic post-printing treatment are presented from a technical point of view. The ultrasonic generator was connected to a booster and a conical sonotrode. Additionally, a special tool with a spherical focussing cup was designed using the 3D finite-

element method (FEM), self-made and joined with the sonotrode. After the assembly was merged, ultrasonic synchronisation was performed to make the system ready. Afterward, an ultrasonic head (UH) was placed in a tandem position behind a printing nozzle. The UH was fixed at a distance equal to the node of the ultrasonic waves. As a result, when the silver trace was printed, ultrasounds were focused at the small point of the printed trace due to the cup shape. The quality of the traces printed with and without ultrasound was assessed by microscopic analysis and resistivity measurements.

2. Materials and methods

2.1 Ultrasonic high-power generator

A Sonic Blaster Plus, high-power 1 kW generator, supplied by the Tele and Radio Research Institute of Warsaw, was used to generate waves of the required ultrasonic frequency. It is a general-purpose power generator used as a power supply for power transducers for the processes of welding metals, plastics, atomisation of liquid metals, or ultrasonic washers.

The generator has a touch-screen graphic display, where parameters such as power and frequency range, as well as ultrasound generation time, can be set. In addition, the generator allows measurement and recording of the transducer's frequency characteristics (phase and impedance as a function of frequency), which makes it possible to control the resonant frequency of the oscillating system.

The useful frequency range is 18–25 kHz, falling within the operating frequency range of the ultrasonic system. The user is informed in real time about the current status of operation, failure, or readiness.

The generator has a built-in digital input and output module, allowing integration with an external control system, increasing flexibility and automatic responses to external signals.

2.2 AJP process

In this article, a custom-built aerosol jet printer was used, as depicted in Figure 1, to illustrate the operational concept of the printing procedure. An aerosol with droplet size in the range of 8–17 μm was generated in an ink reservoir using an ultrasonic transducer immersed in a water chamber and operating at a frequency of 1.7 MHz. A compressed air stream served as the carrier and sheath gas (SG), transporting the generated aerosol to the PH. The ink aerosol flowed through a 0.36 mm diameter orifice and expelled from the nozzle to create fine conductive traces on the foil substrate. The PH, positioned 3 mm above the computer numerical control (CNC) bed, was affixed to a manipulator arm (refer to Figure 2). Precise regulation of compressed air pressure down to 1 mbar was achieved using a microfluidic flow controller (Elveflow OB1 MK3+, Paris, France). Polyimide foil samples, 100 μm in thickness and previously degreased with ethanol, were secured to the high-precision mobile CNC heating bed using magnets. The length of the printed traces was standardised to 40 mm. A sonotrode with a designed and

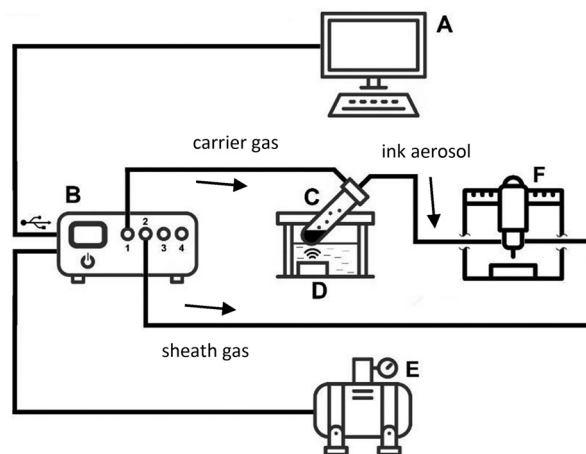


Figure 1. Schematic diagram of the aerosol jet printer with an ultrasonic atomiser equipped with (A) control unit, (B) microfluidic Elveflow controller, (C) ink reservoir, (D) ultrasonic transducer, (E) air compressor, and (F) sonotrode with PH fixed above CNC bed.

self-made UH with cup shape was placed in a tandem position with a PH. The UH was fixed at a multiple length of the node formed by propagating and reflected ultrasonic waves, in this case, 26 mm above the sample. A dedicated cup shape enabled minimisation of ultrasounds region and simultaneously focused and intensified the ultrasound waves. While the printing was finished, the UH moved the same path and modified the structure of still non-dried ink by ultrasounds. The selected samples were printed with an additional inline heating of the substrate material to a temperature of 70°C. In the last step, all of the printed traces were sintered by an near infrared radiation-dot lamp (adphos, Bruckmühl, Germany) in three passes with voltages of 4, 5, and 6 V (from 40 to 60% of the maximum power), with traverse velocity of 10 mm/min. The lamp is equipped with a 150 W emitter halogen lamp that focusses light waves and provides a high heating energy density of 3.9 W/mm².

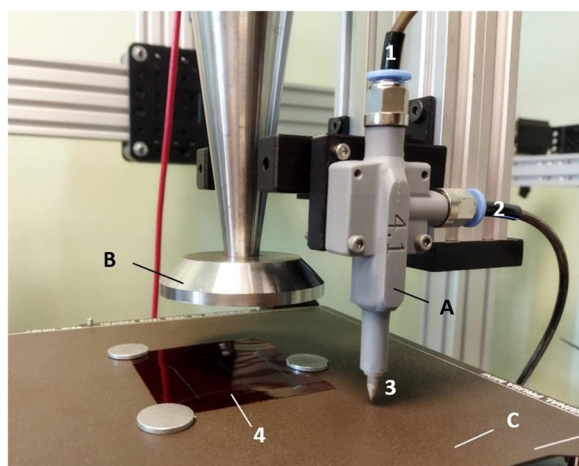


Figure 2. PH (a) and UH (b) fixed and attached to manipulator arm, (c) CNC heating bed. 1 – aerosol inlet, 2 – shielding gas inlet, 3 – exchangeable nozzle with an inner orifice diameter of 0.36 mm, 4 – sample (polyimide foil) with printed traces.

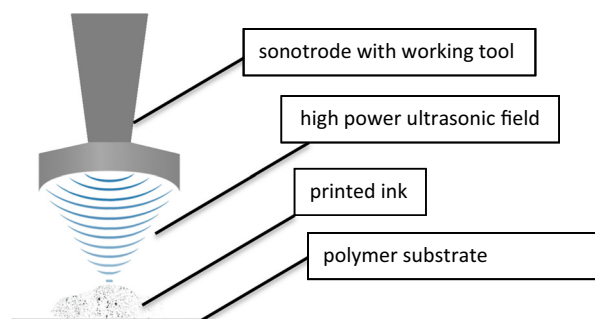


Figure 3. Schematic view of high-power ultrasonic field impact on ink particles.

Figure 3 presents a schematic diagram of the impact of the ultrasonic field on the ink particles applied on the substrate.

The research used commercially available silver nanoparticle suspension ink (Amepox Microelectronics, Ltd., Łódź, Poland), consisting of silver nanoparticles with a median size (d₅₀) of 6 nm, suspended in a solvent, predominantly tetradecane or a mixture of ethanol and glycol, with a silver concentration of 45% by volume. Ink properties are summarised in Table 1. The pressure of the CG and the SG was 80 and 240 mbar, respectively. The printing velocity was fixed at 73 mm/min. The parameters were selected in our previous study [39]. All printing operations were carried out under controlled room conditions at a temperature of 22°C and a humidity level of 50%. In the research, the following samples were produced: printed and sintered (P + S); printed with bed heating and sintered (P + H + S); printed, ultrasounds treated and sintered (P + U + S); and printed with substrate heating, ultrasounds treated and sintered (P + H + U + S).

2.3 Printed trace characterisation methods

The examination of printed and sintered traces involved the use of the VHX-6000 digital microscope (DM) (Keyence VHX-6000, Osaka, Japan) and the atomic force microscope (AFM; NT-MDT NTEGRA Prima, Apeldoorn, The Netherlands). AFM scans were conducted in resonant, non-contact mode, employing NANOSENSORS PPP-NCLR cantilevers. Cross sections of the traces were performed by focused-ion beam (FIB) polishing and imaged with the use of high-resolution scanning electron microscope (SEM) Felios Nanolab 600i DualBeam SEM/Ga-FIB (FEI, Waltham, USA). The width of the samples was measured with the use of DM, analysing ten images at a magnification of 300×. The surface open porosity (SOP), height and roughness of each sample were determined by analysing five images taken with an AFM at a scan area of 0.12 × 0.12 mm². The resistance of the samples was measured using the precise measure unit B2901BL, Keysight Technologies (Santa Rosa, USA), with the four-point probe method. The measurements were conducted for three samples of each type of printed trace at a distance of 40 mm. The resistivity of the traces was calculated on the basis of the electrical measurement combined with the trace length and area of cross-section according to the following equation:

$$\rho = \frac{R \cdot A}{l}, \quad (1)$$

Dynamic viscosity (mPa·s)	Surface tension (dynes/cm)	Density (g/cm ³)	Silver content (%)	Silver powder particle size range (nm)
7.5–10.5	28.5–32.5	1.1–1.3	45	3–8

Table 1. Properties of the utilised ink provided by manufacturer [40].

where ρ is the resistivity (Ω mm), R is the resistance (Ω), A is the area of cross section (mm^2), and l is the length (mm).

3. Results

3.1 Design of the ultrasonic system

The effective vibration system that affects the ink particles (Figure 4) consists of three parts: an ultrasonic transducer (1), a truncated cone-shaped sonotrode (2), and a disc (3) that directly transmits vibrations to the air.

The main consideration when designing the geometry of the sonotrode was to achieve the highest possible vibration at the end of the sonotrode. The first step in selecting the geometry was to find the main dimensions such as length and diameter. The diameter of the sonotrode was selected experimentally, whereas the length of the sonotrode corresponds to half the length of the longitudinal wavelength in order to achieve the largest possible

amplitude of displacement from the equilibrium position. Equation (2) can be used to compute the wavelength based on the ultrasonic wave generator's operating range of 18–25 kHz:

$$\lambda = \frac{c}{f}, \quad (2)$$

where λ is the wavelength, c is the speed of sound (5,000 m/s – dilatation wave for rode), and f is the frequency (20,000 Hz).

The desired frequency of the generated ultrasonic waves has been determined by the resonant frequency of the transducer, close to 20 kHz. Using the expression mentioned above, the wavelength is 250 mm. Based on the theory of standing waves, the wave in the middle of its length shows the greatest amplification or the greatest attenuation; for this reason, assuming maximum deflection on the transducer surface, the length of the sonotrode should be close to 125 mm.

The next stage of the designing process was to establish the concept of the sonotrode shape. The designed system consists of two elements: a sonotrode and a disc, the surface of which has the shape of a lens with a radius of curvature of 75 mm. Due to intended use, the sonotrode should transmit the vibrations generated by the transducer and amplify them while maintaining sufficient rigidity and strength. The task of the sonotrode coupled with the plate is to interact with the externally fed ink, grind it, and distribute it evenly on the surface. During the design and prototyping process, a modal analysis of the vibration system was carried out to obtain the desired vibration modulus

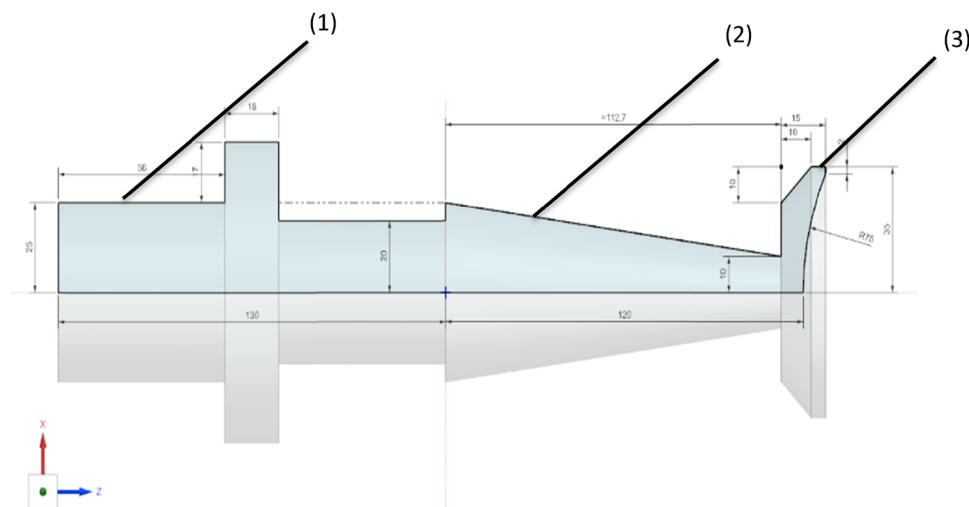


Figure 4. Cross section of the model of the designed ultrasonic system: transducer (1), sonotrode (2), and plate (3).

(longitudinal vibration) at the transducer resonance frequency of 20,000 Hz.

The main requirement formulated for waveguides is to adjust their natural frequency for the axial form of vibration to the frequency of vibration of the transducer. Under such conditions, the system operates in resonance, resulting in large vibration amplitudes. Waveguides are designed to be half-wave resonators. This means that their effective length should be equal to a multiple of half the wavelength. Then, under resonance conditions, a standing wave is formed in them. The antinodes of this wave are located on the mounting surface and, crucially, on the working surface.

At the points of the antinodes, the vibration amplitude reaches the highest values. The points where the nodes of the standing wave occur are places of zero amplitude. They determine the places where the whole system can be fixed in rigid clamps. The distribution of stresses in such a waveguide is reversed. At the antinodes, the stresses are zero and reach their highest value at the wave's node. The form of the standing wave and the stress distribution along the axis of the waveguide are shown in Figure 5.

The wavelength in a waveguide depends on the frequency of the wave, the speed of propagation of the wave in a given material (equation (1)), and the shape of the waveguide. The speed of sound c in a given material can be determined from the following equation:

$$c = \sqrt{\frac{E}{\rho}}, \quad (3)$$

where E is the Young modulus and ρ is the density of rod.

Formula (2) is correct for a rod with a circular cross section (cylindrical sonotrodes). For more complex shapes, the waveguide length must be determined individually based on analytical formulas or computer simulations. Table 2 provides formulas for estimating the appropriate length of sonotrodes so that their geometry allows them to operate in resonance at a given frequency. These formulas

are applicable to sonotrodes with small cross sections. They were derived from the wave equation, the applicability of which applies to waveguides whose cross-sectional dimension does not exceed a quarter of the wavelength in a given material.

For larger waveguide cross sections, the contribution of transverse deformation associated with the Poisson effect increases, and the amplitude distribution is no longer so homogeneous that the wave equation can be used to describe the displacement field [41,45].

Sonotrodes can be designed on the basis of the formulas cited above and then adjusted after they are made. The FEM is successfully used in the design process. With the support of simulations, it is possible to determine the natural frequencies as well as the stress and strain distributions of sonotrodes. The analytical formulas presented can be used to estimate the geometry of the designed tool followed by verification in simulation. For more complex geometries for which there are no suitable formulas, the use of FEM is recommended. For modal analysis supported by FEM, Siemens NX software was used.

The calculations were carried out under the assumption that the sonotrode and disc would be made of AISI 410 series stainless steel. The material was chosen because of its easy availability and anticorrosion properties. As soon as the boundary conditions (limiting vibrations to 1 degree of freedom in order to analyse vibrations in the sonotrode axis) have been determined, the type and type of analysis was selected. The structural modal analysis of the frequency response was chosen because of the accurate determination of the model vibration frequency.

Figure 6 shows the results of the simulation (modal analysis) of the vibration system designed for a frequency of 20,000 Hz. Figure 6a presents the idle state of the ultrasonic system, while Figure 6b and c correspond to the different phases of tool displacement. It should be noted that the RGB colour scale is a dimensionless normalised value of the amplitude displacement.

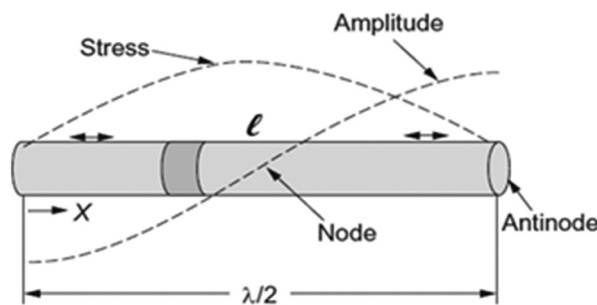


Figure 5. Distribution of vibration amplitude and stress along the waveguide [41].

Sonotrode shape	First resonance length
Cylindrical	$L = \frac{c}{2f}$ [41]
Stepped cylindrical	$L = k_1 \frac{c}{4f} + k_2 \frac{c}{4f}$ [42]
Exponential	$L = \frac{c}{2f} \sqrt{1 + \left(\frac{\ln \left(\frac{D_1}{D_2} \right)}{\pi} \right)^2}$ [43]
Conical	$L = \frac{2c \operatorname{atan} \left(\frac{2\pi f}{ca} \right)}{2\pi f}$ [44]

where

- k_1, k_2 represent coefficients depending on the cross-sectional area (for simplicity, they are taken equal to unity),
- D_1, D_2 represent the diameter of the attachment surface and working surface,
- α is the angle of inclination of the outline formers with respect to the axis.

Table 2. Formulas for estimating length of sonotrode depending on its shape.

Numerical simulations of the integrated vibration system: a transducer with booster, a sonotrode, and a disc confirmed the initial design assumptions. The maximum deflection values are at the assumed location, i.e. at the surface of the disc, which is the source of the ultrasonic wave.

Figure 7 shows a complete 3D model of the ultrasonic system consisting of a power transducer, a sonotrode, and a working tip focussing the ultrasonic wave. The individual parts of the actual system are connected by screw connections.

3.2 Resonance measurements of the vibrating system

First, the frequency characteristics of the transducer (Figure 8a), the transducer connected to the sonotrode (Figure 8b), and the system used for testing, i.e. the transducer, sonotrode, and disc (Figure 8c), were measured in order to verify the correctness of the calculations made using the FEM. The presented characteristics show that

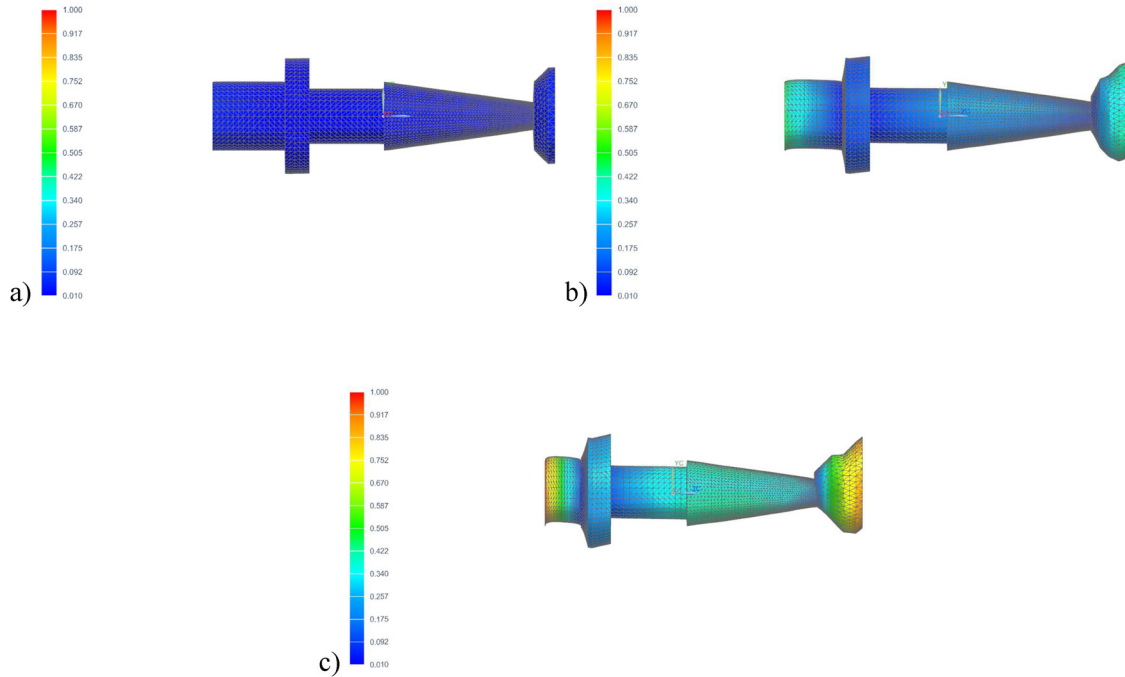


Figure 6. Results of modal analysis of the designed ultrasonic system for 20,000 Hz longitudinal mode. Idle state (a), different phases of tool displacement: +45° (b) and +90° (c).



Figure 7. View of 3D model of the ultrasonic system with the working plate for ultrasonic wave concentration.

the developed ultrasonic system was designed, assembled, and integrated correctly. Its frequency characteristics are distinguished by a narrow resonance at 20,040 Hz, which corresponds to the design assumption of 20,000 Hz. The difference of 40 Hz resulting from calculations and

measurements is negligible and does not affect the operation of the system.

3.3 Analysis of printed traces

The results of the printing process with various modifications are presented in Figure 9. It is clearly visible that all the printed traces are well concentrated in the main axis, with the residues of the spilt ink at the edges. The overspray is visible on either side of each sample. However, intensive spatter occurred in samples printed without bed heating (Figure 9a and c), smudging all around the polymeric substrate. It should be emphasised that the application of ultrasounds influenced the borderline of the traces, making it more irregular (Figure 9c and d). The width of the samples measured with and without spilt ink residues is presented in Table 3. The tracks printed with bed heating showed slightly lower width. However, the application of ultrasounds caused spilling of the deposited ink and, as a result, increased the final width. The comparison

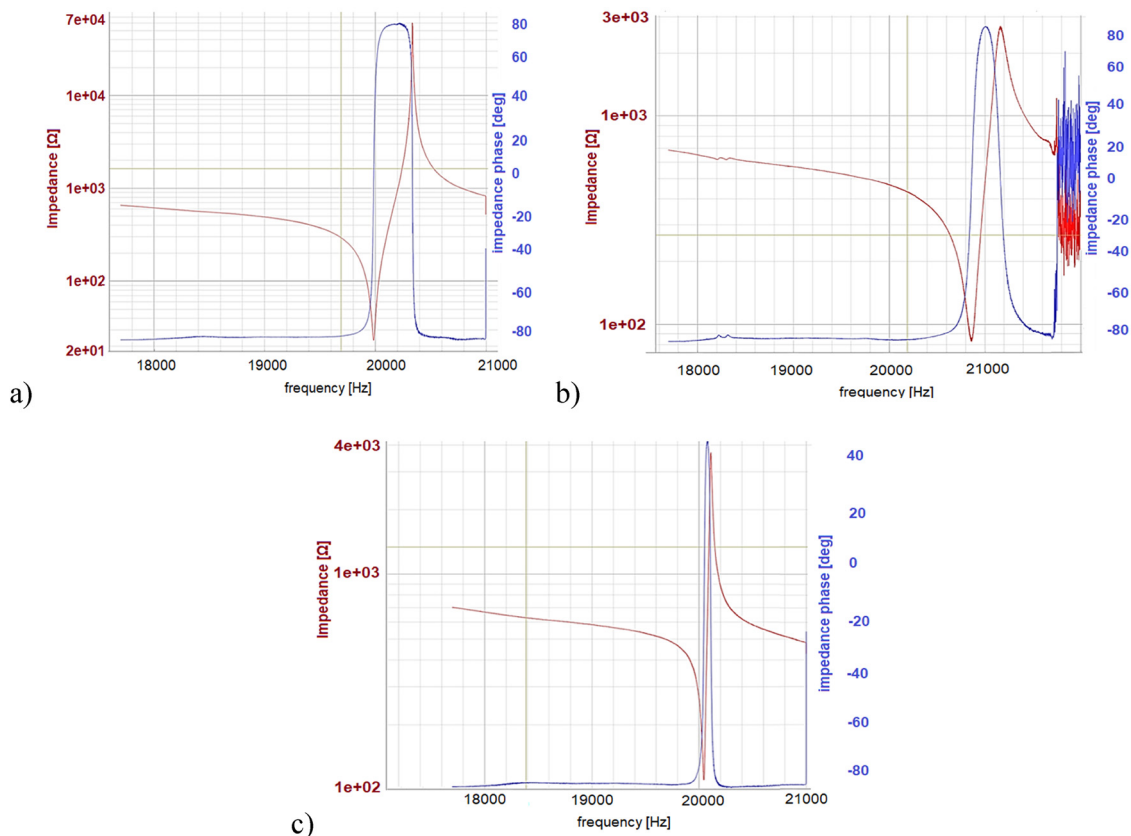


Figure 8. Frequency characteristics of the designed ultrasonic system intended to increase the uniformity of particle distribution of injected paths. Impedance characteristics of transducer (a), transducer coupled with sonotrode (b), and complete ultrasonic system (c).

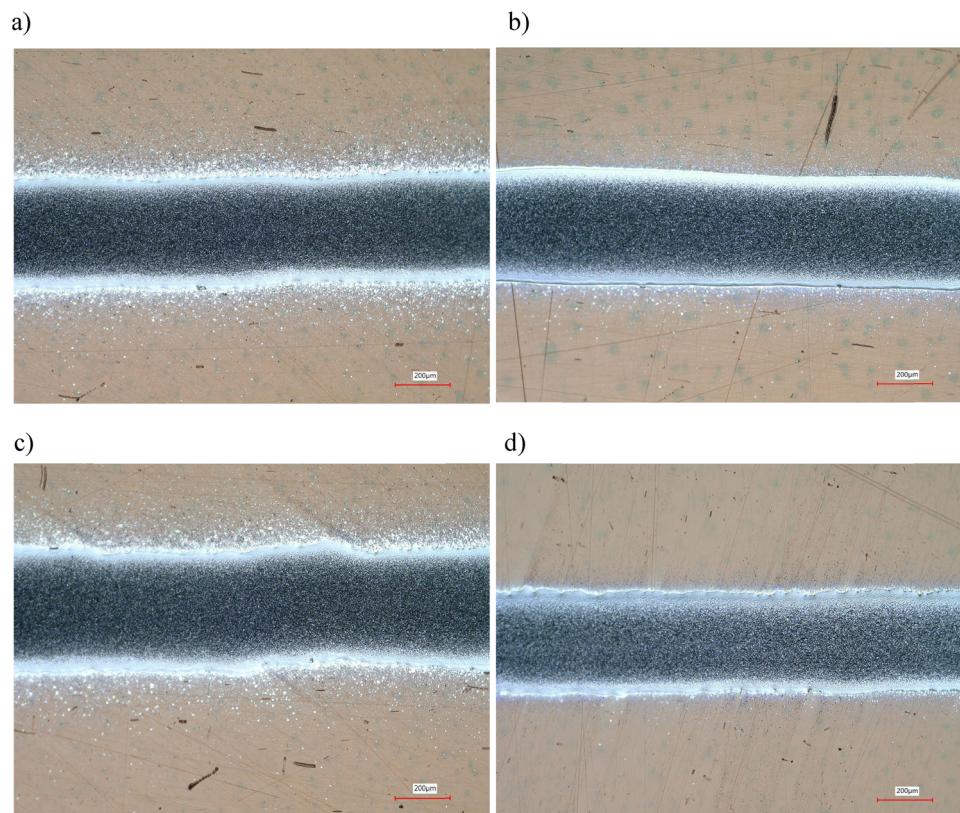


Figure 9. Top view of printed traces: P + S (a), P + H + S (b), P + U + S (c), and P + H + U + S (d).

of samples fabricated without P + S and with ultrasound application (P + U + S) is presented in Figure 10. This phenomenon was stopped by applying substrate heating due to intensive evaporation of dissolvent. Therefore, the width of the sample P + H + U + S was comparable to P + S and P + H + S (Figure 10a and 10b). It is worth noting that the samples produced with ultrasound treatment showed a higher average height value, resulting from the increased mobility of the particles (Table 3).

The sintering process caused total evaporation of the liquid dissolvent and residual polymers. The morphology

of the traces remains unchanged with one exception: the border of the traces is flattened without a noticeable sharp edge (compare Figures 9 and 11). It arises from the total evaporation of the liquid and a decrease in the volume of the trace. Some local porosity of the trace with SOP was found in the spilled region (Figures 12 and 13). The porosity resulted from rapid evaporation of the solvent. Within a short period of time, the drying liquid underwent a phase change to gas, and the resulting pressure facilitated its removal from the deposited material. Crack generation and propagation were observed in the sample P + H +

Sample	Width (µm)		Height (nm)	Roughness		SOP (%)
	Total	Without spilled ink	Average value	Sa (nm)	Sz (nm)	
P + S	406 ± 8	311 ± 11	586 ± 31	123 ± 20	995 ± 33	17.2
P + H + S	383 ± 47	294 ± 29	556 ± 89	91 ± 13	888 ± 30	9.1
P + U + S	526 ± 56	416 ± 35	629 ± 106	148 ± 21	1158 ± 181	5.9
P + H + U + S	415 ± 29	302 ± 33	722 ± 103	92 ± 9	1276 ± 198	2.7

The values presented in the table contain the standard deviation of the obtained results.

Table 3. Geometry and properties of printed traces.

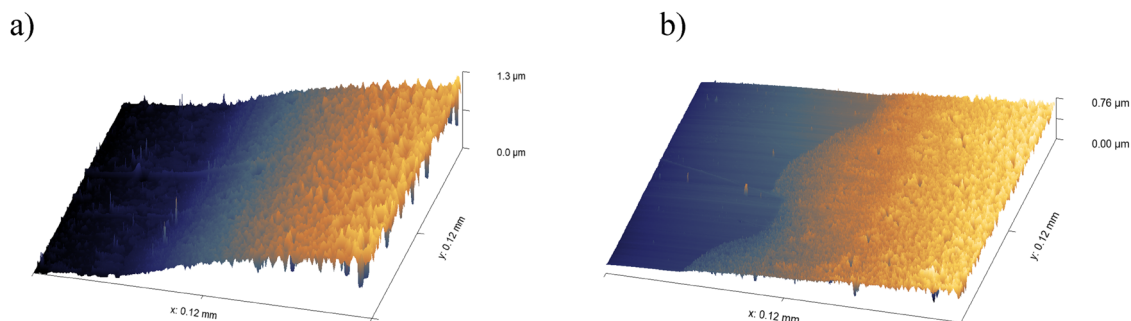


Figure 10. The boundary region of sample P + S (a) and P + U + S (b).

U + S due to excess of the process energy (Figure 11d). Applied in the printing process, bed heating intensified the formation of silver agglomerates in deposited traces with minimised residues of liquid solvent. Further ultrasonic treatment provided additional energy, which affected the semi-dried material by tearing the agglomerated bonds that formed and leading to crack formation (Figure 12a and b). All of the samples showed an increase in roughness (Sa and Sz) on the surface. However, the mean roughness (Sa)

decreased due to the application of substrate heating in the P + H + S and P + H + U + S samples, while the highest maximum height (Sz) was observed for the ultrasonically treated samples, P + U + S and P + H + U + S (Table 3).

AFM scans in the field of $0.12 \times 0.12 \text{ mm}^2$ showed the presence of porosity in all samples (Figure 13). The highest SOP of 17.2% showed the P + S sample (Figure 14a and Table 3). The application of bed heating or ultrasounds decreased the presence of pores to 9.1 and 5.9%, respectively.

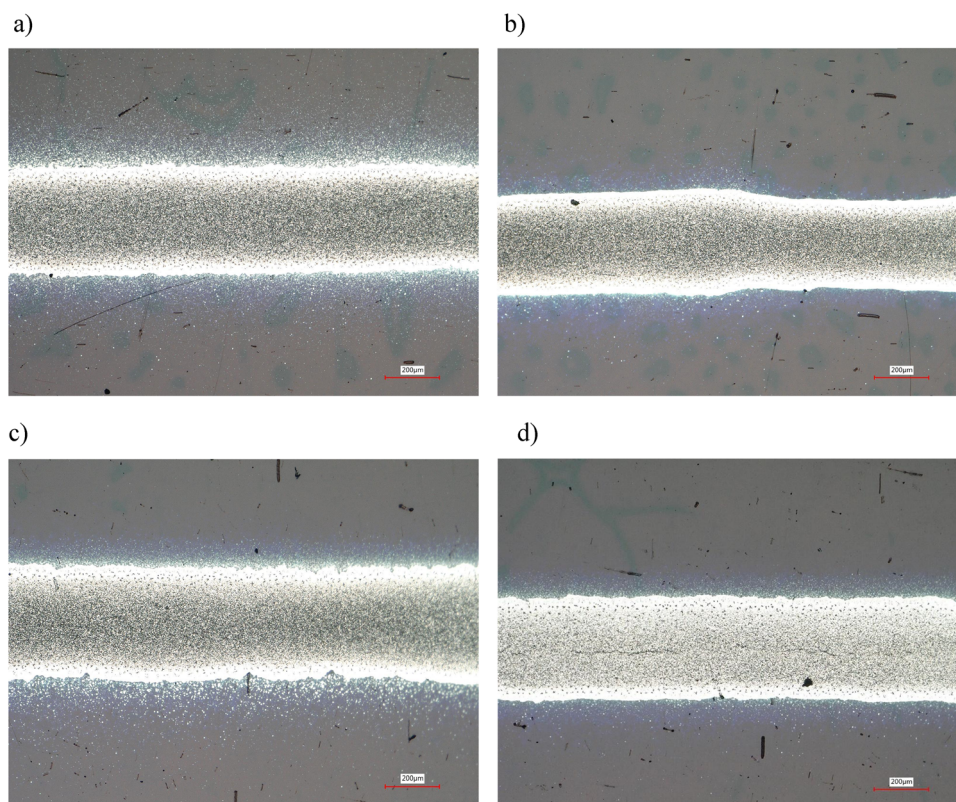


Figure 11. Top view of printed and sintered traces: P + S (a), P + H + S (b), P + U + S (c), and P + H + U + S (d). The dark region in the central part of the trace responds to porosity and high roughness.

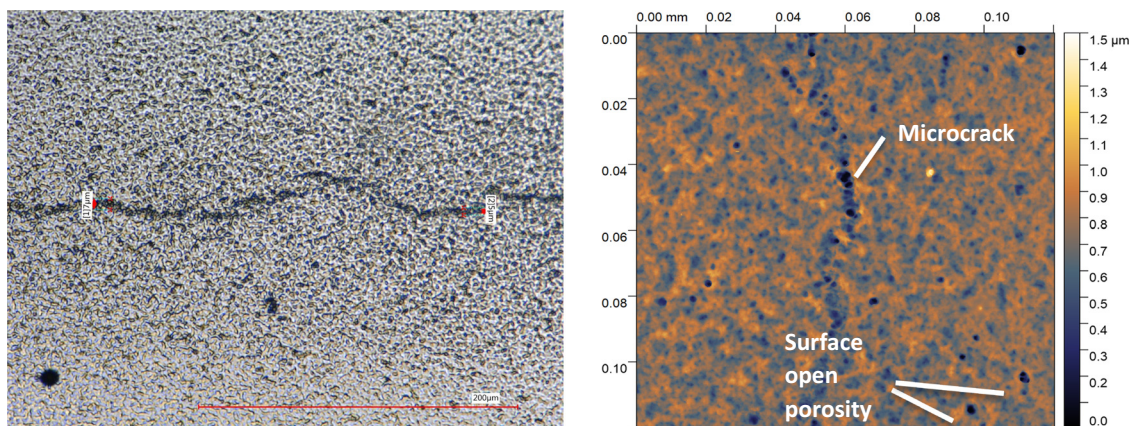


Figure 12. Sample P + H + U + S with a visible crack in the axis: DM (a) and AFM (b).

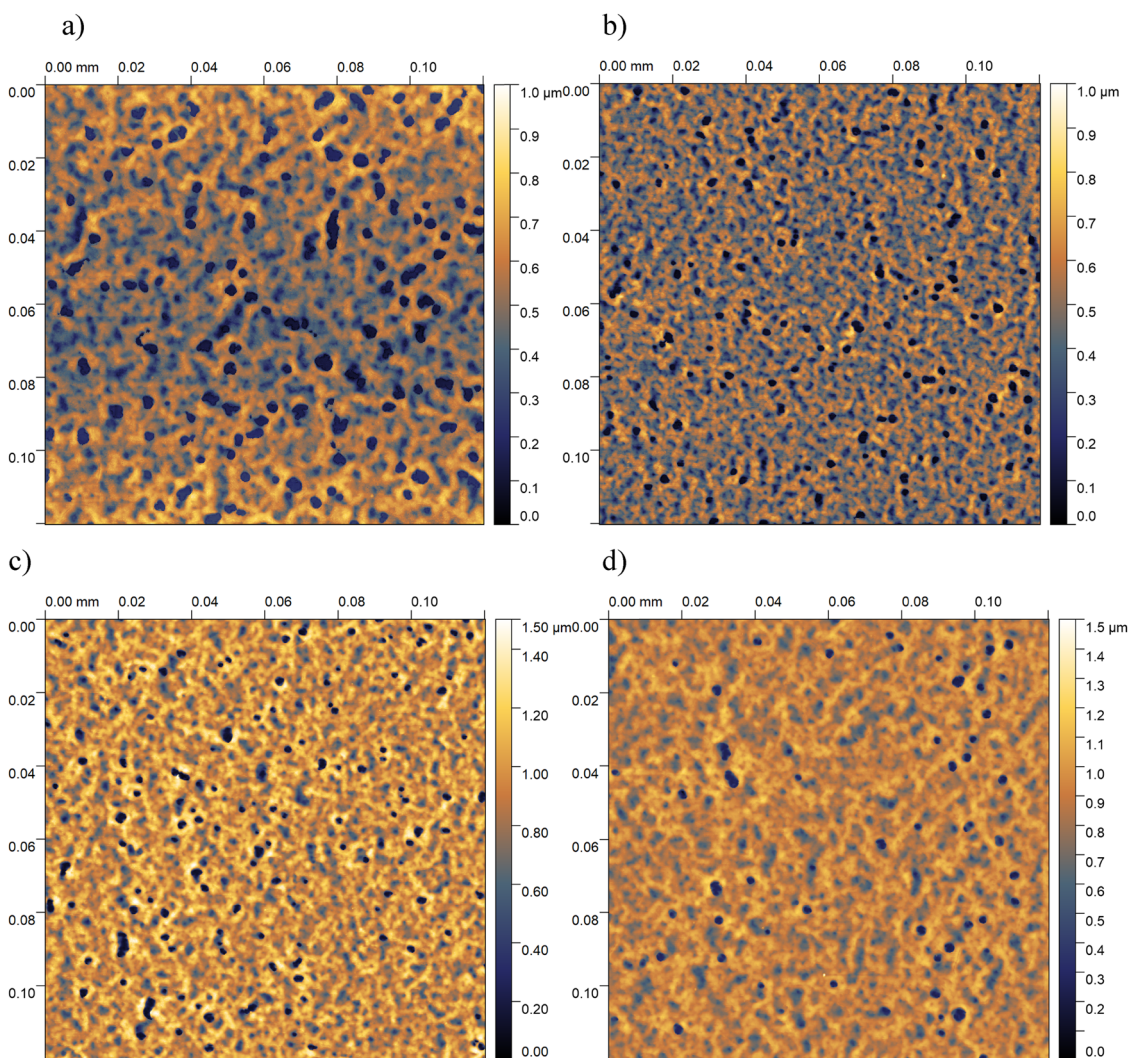


Figure 13. AFM scans of sample surface: P + S (a), P + H + S (b), P + U + S (c), and P + H + U + S (d). Black dots respond to SOP.

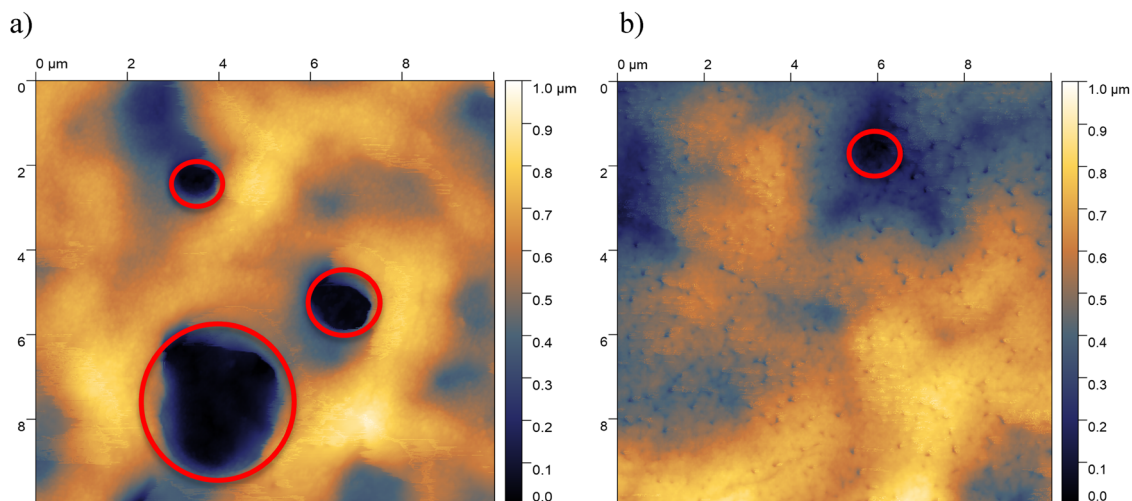


Figure 14. AFM micrographs of P + S (a) and P + U + S (b) samples. Red circles highlight black dots and respond to SOP.

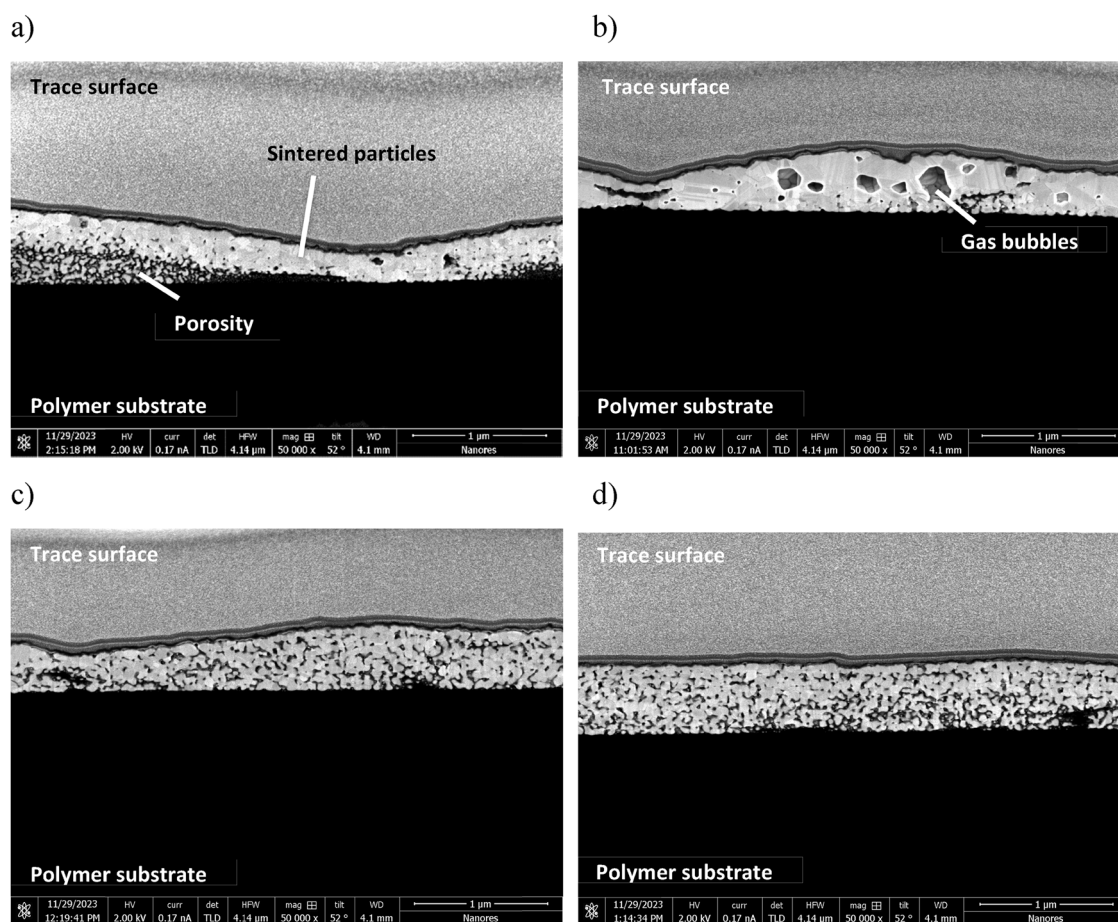


Figure 15. SEM micrographs presenting cross-section of printed traces: P + S (a), P + H + S (b), P + U + S (c), and P + H + U + S (d).

The lowest SOP of 2.7% showed a sample P + H + U + S. It should be emphasised that large pores are present in the P + S sample response for trace open porosity. When comparing sample P + S to P + U + S, it is clearly observed that the size, shape, and depth of the pores are tremendous (Figure 14). The quality of the sintering process was evaluated by traces cross-section (Figure 15). The multi-sectional region with porosity, nonuniform particle agglomerate distribution, and irregular sintered structure showed samples P + S. The top layer is fully sintered, forming a shell, while the middle region is characterised by weakly bonded particles. The posttreatment quality presented by the P + H + S sample is characterised by a uniformly sintered structure of bulk material showing the grain boundary (Figure 15b). However, fine pores joined in large gas bubbles because of the rapid solidification process blocking degassing. Both ultrasonically treated samples, i.e. P + U + S and P + H + U + S, presented well-bonded regular structures with uniform particle distribution and the presence of just some local nanoporosity.

The results of resistance measurements and the following resistivity calculations are presented in Figure 16. The highest resistance of 24.49 Ω was measured for the P + S sample and decreased due to process modifications to 14.01 Ω received for P + H + U + S. The width and height of all the traces were comparable, except P + U + S and P + H + U + S, which were 30% wider and 20% higher, respectively (Table 3). Higher width and height of the traces result in higher cross-sectional area and consequently decreased resistance. Nevertheless, the geometry of the traces arose from an appropriate post-treatment process. The manufacturing process influences the quality of the material. Therefore, the resistivity of the samples differs.

The highest resistivity of 6.85 $\mu\Omega \text{ cm}$ was calculated for the untreated P + S sample, defined by highest porosity, while the lowest resistivity of 4.57 $\mu\Omega \text{ cm}$ achieved P + H + U + S sample. Evidently, the sample P + S showed the lowest quality, while P + H + S and P + H + U + S were comparable.

4. Discussion

A sonotrode is part of an ultrasonic system that transfers high-frequency vibrations to the material to be properly treated. The geometry and dimensions of the sonotrode affect the amplitude, frequency, and mode shape of the vibrations, which in turn influence the quality and efficiency of the ultrasonic process [46,47]. According to Li *et al.* [48], the optimal design of a sonotrode depends on several factors, such as the material properties, the operating frequency, the desired amplitude, the stress distribution, the temperature distribution, and the coupling with the transducer. Finite-element analysis was used to model and optimise the sonotrode geometry and dimensions for various applications. The measured characteristics showed that the developed ultrasonic system was designed, assembled, and integrated correctly and prepared to work with the AJP printer. The dimensions of the sonotrode were carefully tuned and matched to the ultrasonic transducer and the substrate material. Proper tuning ensured maximum efficiency of the system, minimising energy losses. It is worth emphasising that ultrasonic vibrations generate heat; therefore, sonotrode material considers thermal effects. Excess heat can affect the performance of the sonotrode and the materials being processed [49]. However, the cooling systems

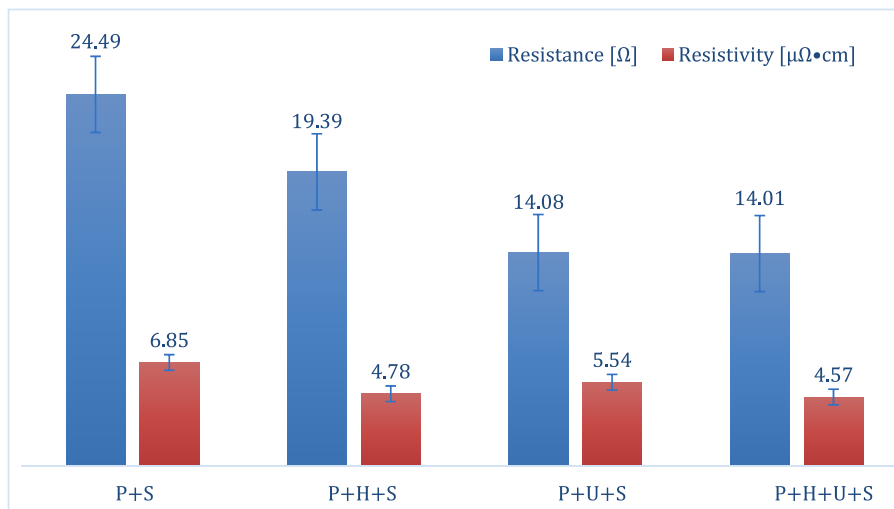


Figure 16. Results of resistance measurements and resistivity calculations.

were not incorporated due to the short working cycles of the ultrasonic generator.

The AJP process contains printing and sintering; however, various modifications are possible to increase the quality of the prints. A commonly applied extension is bed heating [50,51] that provides fast evaporation of the liquid phase prior to sintering. Nevertheless, still the most problematic issue is the formation of porosity. Heated liquids transform into gas and can lead to the formation of gas bubbles, especially while rapid sintering by electromagnetic radiation, e.g. IR. Radiation is absorbed by the top layer of the printed material, and due to the accumulation of energy, nano-sized particles are sintered [52]. Afterwards, heat is conducted to the lower part of the print, and sintering embraces the residual part of the material. This phenomenon occurred in the P + S sample and is clearly visible in Figure 15a. A sintered top layer forms a shell-like structure and blocks degassing of the lower part. Consequently, the pressure of the evaporating gas increases together with the increase in the temperature of the material and forms gas bubbles (Figure 15b).

To solve the problem, an ultrasonic treatment was proposed to remove the gas when the printing was done. The waves propagating through the material shattered the pores and ensured degassing of the material in a semi-dry state [53,54]. As a result, SOP and surface roughness were significantly minimised (Table 3 and Figure 14), while the bubbles present in the cross-section view of the traces were totally eliminated (Figure 16). The appropriate ultrasonic energy was focused on the printed material due to the dedicated shape of the sonotrode. The side effect of ultrasonics is the alteration of surface tension between ink and polymer substrate [55,56]. The ink flows on the boundary region, increasing the width of the trace, hindering the miniaturisation of the prints. However, the problem was solved by combining printing with bed heating and ultrasound treatment. The semi-dried material by bed heating on the boundary region blocked the spread of the ink in ultrasound treatment. The improved quality resulted in satisfactory electrical resistivity of the prints. In addition, it is stated that ultrasonically modified structures will increase the mechanical strength and plasticity of the prints. However, this thesis should be verified by further research.

5. Conclusions

In this article, the UA-AJP process is presented from a technical point of view. The dedicated, self-made device consisted of an ultrasonic generator, a booster, and a conical sonotrode combined with a spherical focussing cup.

The system was designed and analysed using the 3D FEM. After synchronisation, the UH was placed in a tandem position behind an AJP printing nozzle. The UH was fixed vertically at a calculated distance to focus ultrasound waves at the small point in the printed trace. Four types of tests were performed: (i) printing and sintering by IR lamp; (ii) printing with bed heating and sintering by IR lamp; (iii) printing, ultrasonic treatment, and sintered by IR lamp; and (iv) printing with bed heating, ultrasonic treatment and sintering by IR lamp.

The quality of the traces printed with and without the assistance of ultrasounds differed significantly. The printed and sintered sample was full of defects, such as porosity, open pores, non-uniform particle distribution, high roughness, and nonregular sintered structure. Application of additional bed heating solved problems with nonregular sintered structure. Nevertheless, the heated substrate increased gas fabrication and led to the formation of gas bubbles inside the prints. The ultrasonic treatment favoured degassing, as well as increased uniform of particle distribution and, as a result, structure regularity. As a result, the resistivity of the UA-AJP prints was decreased.

Printing with bed heating provides two main advantages. First of all, it limits the spatter and formation of the overspray, and additionally, it blocks the spread of the ink in ultrasonic treatment due to the presence of semi-dried material in the boundary region. As a result, traces printed with bed heating showed slightly lower width. Combining substrate heating and ultrasonic treatment showed one disadvantage. Applied in the printing process, bed heating intensified the agglomeration of silver nanoparticles, while further ultrasonic treatment caused tearing to form agglomerated bonds and led to crack formation. To solve the problem, ultrasonic power should be decreased, while process time increased.

Summarising, the UA-AJP process represents a significant advancement in additive manufacturing. By harnessing the power of ultrasonic waves, this technique offers improved material compatibility, increased deposition rates, and enhanced print quality. The potential applications are vast, spanning across electronics, sensors, and biomedical fields. As research progresses, the integration of ultrasonic assistance is expected to unlock new possibilities in additive manufacturing, paving the way for the creation of innovative and tailored functional structures.

Acknowledgements

This work was funded by The National Centre for Research and Development (LIDER/42/0142/L-11/19/NCBR/2020)

(project title: Sonic Jet – precise printer for manufacturing elastic electronics).

Funding information

The project number has been funded by The National Centre for Research and Development (LIDER/42/0142/L-11/19/NCBR/2020).

Author contributions

Marcin Korzeniowski: developed the theoretical framework, designed the model and the computational framework, performed the numerical calculations for the suggested experiment, performing experiment; Marcin Winnicki: developed the theoretical framework, conceived and planned the experiments, the main conceptual ideas and proof outline, performing experiment; Bartosz Swiadkowski: samples and results characterization, interpreting the results; Wojciech Lapa: contributed to sample preparation, assisted with measurements.

Conflict of interest statement

The author has no conflicts of interest to declare.

References

- [1] Wu, W., Inorganic nanomaterials for printed electronics: a review, *Nanoscale*, 2017, 9: 7342–7372, doi: 10.1039/C7NR01604B
- [2] Suganuma, K., Introduction to printed electronics, New York, NY: Springer; 2014, doi: 10.1007/978-1-4614-9625-0
- [3] Shaik, R.A., Rufus, E., Recent trends and role of large area flexible electronics in shape sensing application – a review, *Ind. Robot. Int. J. Robot Res. Appl.*, 2021, 48: 745–762, doi: 10.1108/IR-10-2020-0234
- [4] Choi, H.W., Zhou, T., Singh, M., Jabbour, G.E., Recent developments and directions in printed nanomaterials, *Nanoscale*, 2015, 7: 3338–3355, doi: 10.1039/C4NR03915G
- [5] Mosa, MdA, Jo, J.Y., Kwon, K.-S., Fast on-off jet control of aerosol jet printing (AJP) using internal rotary valve, *Addit. Manuf.*, 2023, 67: 103466, doi: 10.1016/j.addma.2023.103466
- [6] Seiti, M., Degryse, O., Ferraris, E., Aerosol Jet® printing 3D capabilities for metal and polymeric inks, *Mater. Today Proc.*, 2022, 70: 38–44, doi: 10.1016/j.matpr.2022.08.488
- [7] Tursunniyaz, M., Meredith, A., Andrews, J., Aerosol jet printed resistive temperature sensors with high sensitivity, *Sens. Actuators A Phys.*, 2023, 364: 114777, doi: 10.1016/j.sna.2023.114777
- [8] Ramesh, S., Mahajan, C., Gerdes, S., Gaikwad, A., Rao, P., Cormier, D.R., et al., Numerical and experimental investigation of aerosol jet printing, *Addit. Manuf.*, 2022, 59: 103090, doi: 10.1016/j.addma.2022.103090
- [9] Hines, D.R., Gu, Y., Martin, A.A., Li, P., Fleischer, J., Clough-Paez, A., et al., Considerations of aerosol-jet printing for the fabrication of printed hybrid electronic circuits, *Addit. Manuf.*, 2021, 47: 102325, doi: 10.1016/j.addma.2021.102325
- [10] Zhu, Y., Yu, L., Wu, D., Lv, W., Wang, L., A high-sensitivity graphene ammonia sensor via aerosol jet printing, *Sens. Actuators A Phys.*, 2021, 318: 112434, doi: 10.1016/j.sna.2020.112434
- [11] Ako, H., O'Mahony, J., Hughes, H., McLoughlin, P., O'Reilly, N.J., A novel approach to the manufacture of dissolving microneedles arrays using aerosol jet printing, *Appl. Mater. Today*, 2023, 35: 101958, doi: 10.1016/j.apmt.2023.101958
- [12] Gamba, L., Lajoie, J.A., Sippel, T.R., Secor, E.B., Multi-material aerosol jet printing of Al/Cu nanothermites for versatile fabrication of energetic antennas, *Adv. Funct. Mater.*, 33: 2304060, doi: 10.1002/adfm.202304060
- [13] Fisher, C., Skolrood, L.N., Li, K., Joshi, P.C., Aytug, T., Aerosol-jet printed sensors for environmental, safety, and health monitoring: a review, *Adv. Mater. Technol.*, 2023, 8: 2300030, doi: 10.1002/admt.202300030
- [14] Degryse, O., Bloemen, V., Ferraris, E., Collagen composite inks for Aerosol Jet® printing in bone tissue engineering applications, *Procedia CIRP*, 2022, 110: 180–185, doi: 10.1016/j.procir.2022.06.033
- [15] Wilkinson, N.J., Smith, M.A.A., Kay, R.W., Harris, R.A., A review of aerosol jet printing—a non-traditional hybrid process for micro-manufacturing, *Int. J. Adv. Manuf. Technol.*, 2019, 105: 4599–4619, doi: 10.1007/s00170-019-03438-2
- [16] Skarżyński, K., Krzemiński, J., Jakubowska, M., Słoma, M., Highly conductive electronics circuits from aerosol jet printed silver inks, *Sci. Rep.*, 2021, 11: 1–9, doi: 10.1038/s41598-021-97312-5
- [17] Taccola, S., da Veiga, T., Chandler, J.H., Cespedes, O., Valdastri, P., Harris, R.A., Micro-scale aerosol jet

- printing of superparamagnetic Fe_3O_4 nanoparticle patterns, *Sci. Rep.*, 2022, 12: 17931, doi: 10.1038/s41598-022-22312-y
- [18] Safaee, S., Schock, M., Joyee, E.B., Pan, Y., Chen, R.K., Field-assisted additive manufacturing of polymeric composites, *Addit. Manuf.*, 2022, 51: 102642, doi: 10.1016/j.addma.2022.102642
- [19] Moon, S.K., Ng, N.P.H., Chen, L., Ahn, D.-G., A novel quality inspection method for aerosol jet printed sensors through infrared imaging and machine learning, *CIRP Ann.*, 2023, 72: 165–168, doi: 10.1016/j.cirp.2023.03.029
- [20] Dunst, P., Hemsel, T., Bornmann, P., Littmann, W., Sextro, W., Optimization of ultrasonic acoustic standing wave systems, *Actuators*, 2020, 9: 9, doi: 10.3390/act9010009
- [21] Ostasevicius, V., Jurenas, V., Golinka, I., Gaidys, R., Aleksa, A., Separation of microparticles from suspension utilizing ultrasonic standing waves in a piezoelectric cylinder actuator, *Actuators*, 2018, 7: 14, doi: 10.3390/act7020014
- [22] Haake, A., Dual, J., Micro-manipulation of small particles by node position control of an ultrasonic standing wave, *Ultrasonics*, 2002, 40: 317–322, doi: 10.1016/S0041-624X(02)00114-2
- [23] Ikram, A., Mehmood, F., Sheridan, R.S., Awais, M., Walton, A., Eldosouky, A., et al., Particle size dependent sinterability and magnetic properties of recycled HDDR Nd–Fe–B powders consolidated with spark plasma sintering, *J. Rare Earths*, 2020, 38: 90–99, doi: 10.1016/j.jre.2019.02.010
- [24] Cravotto, G., Gaudino, E.C., Cintas, P., On the mechanochemical activation by ultrasound, *Chem. Soc. Rev.*, 2013, 42: 7521–7534, doi: 10.1039/C2CS35456J
- [25] Harris, N.R., Hill, M., Particle manipulation using ultrasonic fields, In: Li, D., (Ed.). *Encyclopedia of microfluidics and nanofluidics*, US, Boston, MA: Springer; 2008, p. 1597–1602, doi: 10.1007/978-0-387-48998-8_1186
- [26] Courtney, C.R.P., Ong, C.-K., Drinkwater, B.W., Bernassau, A.L., Wilcox, P.D., Cumming, D.R.S., Manipulation of particles in two dimensions using phase controllable ultrasonic standing waves, *Proc. R. Soc. A Math. Phys. Eng. Sci.*, 2011, 468: 337–360, doi: 10.1098/rspa.2011.0269
- [27] Hill, M., Harris, N.R., Ultrasonic particle manipulation, In: Hardt S., Schönfeld F., (Eds.). *Microfluidic technologies for miniaturized analysis systems*, US, Boston, MA: Springer; 2007, p. 357–392, doi: 10.1007/978-0-387-68424-6_9
- [28] Łapa, W., Winnicki, M., Orłowska, K., Investigation of aerosol droplets diameter generated in aerosol jet printing, *Mater. Sci. Poland*, 2022, 40: 78–90, doi: 10.2478/msp-2022-0046
- [29] Sreenilayam, S.P., McCarthy, É., McKeon, L., Ronan, O., McCann, R., Fleischer, K., et al., Additive-free silver nanoparticle ink development using flow-based laser ablation synthesis in solution and aerosol jet printing, *Chem. Eng. J.*, 2022, 449: 137817, doi: 10.1016/j.cej.2022.137817
- [30] Williams, B.A., Trejo, N.D., Wu, A., Holgate, C.S., Francis, L.F., Aydil, E.S., Copper–zinc–tin–sulfide thin films via annealing of ultrasonic spray deposited nanocrystal coatings, *ACS Appl. Mater. Interfaces*, 2017, 9: 18865–18871, doi: 10.1021/acsami.7b04414
- [31] Binder, S., Glatthaar, M., Rädlein, E., Analytical investigation of aerosol jet printing, *Aerosol Sci. Technol.*, 2014, 48: 924–929, doi: 10.1080/02786826.2014.940439
- [32] Secor, E.B., Principles of aerosol jet printing, *Flex. Print. Electron.*, 2018, 3: 035002, doi: 10.1088/2058-8585/aace28
- [33] Alhendi, M., Sivasubramony, R.S., Lombardi, J., Weerawarne, D.L., Borgesen, P., Poliks, M.D., et al., Laser sintering of aerosol jet printed conductive interconnects on paper substrate, 2019 IEEE 69th Electronic Components and Technology Conference (ECTC), 2019, p. 1581–1587, doi: 10.1109/ECTC.2019.00243
- [34] Gramlich, G., Huber, R., Häslich, F., Bhutani, A., Lemmer, U., Zwick, T., Process considerations for Aerosol-Jet printing of ultra fine features, *Flex. Print. Electron.*, 2023, 8: 035002, doi: 10.1088/2058-8585/ace3d8
- [35] Sui, Y., Tsui, L., Thibodeaux, A.J., Lavin, J.M., An aerosol jet printed resistance temperature detector-micro hotplate with temperature coefficient of resistance stabilized by electrical sintering, *Adv. Mater. Technol.*, 2023, 8: 2202053, doi: 10.1002/admt.202202053
- [36] Pandhi, T., Kreit, E., Aga, R., Fujimoto, K., Sharbati, M.T., Khademi, S., et al., Electrical transport and power dissipation in aerosol-jet-printed graphene interconnects, *Sci. Rep.*, 2018, 8: 10842, doi: 10.1038/s41598-018-29195-y
- [37] Zhang, Y., Chen, X., Particle separation in microfluidics using different modal ultrasonic standing waves, *Ultrason. Sonochem.*, 2021, 75: 105603, doi: 10.1016/j.ultsonch.2021.105603
- [38] Silva, G.T., Lopes, J.H., Leão-Neto, J.P., Nichols, M.K., Drinkwater, B.W., Particle patterning by ultrasonic standing waves in a rectangular cavity, *Phys. Rev. Appl.*, 2019, 11: 054044, doi: 10.1103/PhysRevApplied.11.054044

- [39] Winnicki, M., Łapa, W., Świadkowski, B., A novel approach to improve reliability of aerosol jet printing process, *EiN – Maintenance and Reliability*, 2024, 26(2), doi: 10.17531/ein/180012
- [40] Kinart, A.E., Moscicki, A.J., Nano-inks for printing electric circuits for microelectronics technology, 2014, <https://science24.com/paper/31307#gsc.tab=0> (access 16 July 2024).
- [41] Gallego-Juárez, J.A., Graff, K.F., 1 - Introduction to power ultrasonics, In: Gallego-Juárez J. A., Graff K. F., (Eds.). *Power ultrasonics*, Oxford: Woodhead Publishing; 2015, p. 1–6, doi: 10.1016/B978-1-78242-028-6.00001-6
- [42] Seah, K.H.W., Wong, Y.S., Lee, L.C., Design of tool holders for ultrasonic machining using FEM, *J. Mater. Process. Technol.*, 1993, 37, 801–816, doi: 10.1016/0924-0136(93)90138-V
- [43] Dipal, A., Analysis of different shaped sonotrodes used for plastic welding. Conference paper, Institute of Technology, Nirma University; 2011
- [44] Yassin, M.M., Design of ultrasonic processing device for aluminum surfaces, Master thesis, Waterloo, Ontario, Canada: Univ. of Waterloo; 2018
- [45] Nad, M., Ultrasonic horn design for ultrasonic machining technologies, *Appl. Comput. Mech.*, 2010, 4, 79–88
- [46] Liesegang, M., Yu, Y., Beck, T., Balle, F., Sonotrodes for ultrasonic welding of titanium/CFRP-joints—materials selection and design, *J. Manuf. Mater. Process.*, 2021, 5, 61, doi: 10.3390/jmmp5020061
- [47] Wang, Y., Chen, Z., Yu, Q., Cheng, F., Modeling of sonotrode system of ultrasonic consolidation with transfer matrix method, *Front. Mater.*, 2021, 8, 642896, doi: 10.3389/fmats.2021.642896 (accessed January 2, 2024)
- [48] Li, H., Cao, B., Liu, J., Yang, J., Modeling of high-power ultrasonic welding of Cu/Al joint, *Int. J. Adv. Manuf. Technol.*, 2018, 97, 833–844, doi: 10.1007/s00170-018-2002-1
- [49] Li, H., Cao, B., Yang, J.W., Liu, J., Modeling of resistance heat assisted ultrasonic welding of Cu-Al joint, *J. Mater. Process. Technol.*, 2018, 256, 121–130, doi: 10.1016/j.jmatprotec.2018.02.008
- [50] Rurup, J.D., Secor, E.B., A real-time process diagnostic to support reliability, control, and fundamental understanding in aerosol jet printing, *Adv. Eng. Mater.*, 26, 2301348, doi: 10.1002/adem.202301348
- [51] Li, L., Zhang, K., Cheng, H., Ma, T., Niu, Y., Li, A., et al., Experimental and simulation investigations on the morphology of aerosol jet printed polymer traces under in-situ UV and thermal curing conditions, *Addit. Manuf.*, 2023, 69, 103515, doi: 10.1016/j.addma.2023.103515
- [52] Vaithilingam, J., Simonelli, M., Saleh, E., Senin, N., Wildman, R.D., Hague, R.J.M., et al., Combined inkjet printing and infrared sintering of silver nanoparticles using a swathe-by-swathe and layer-by-layer approach for 3-dimensional structures, *ACS Appl. Mater. Interfaces*, 2017, 9, 6560–6570, doi: 10.1021/acsami.6b14787
- [53] Abramov, V.O., Abramova, A.V., Bayazitov, V.M., Nikonov, R.V., Cravotto, G., Pores-free aluminium alloy by efficient degassing ultrasonic treatments, *Appl. Acoust.*, 2021, 184, 108343, doi: 10.1016/j.apacoust.2021.108343
- [54] Białas, K., Buchacz, A., Active reduction of vibration of mechatronic systems, *EiN*, 2015, 17, 528–534, doi: 10.17531/ein.2015.4.7
- [55] Wu, H., Zheng, H., Li, Y., Ohl, C.-D., Yu, H., Li, D., Effects of surface tension on the dynamics of a single micro bubble near a rigid wall in an ultrasonic field, *Ultrason. Sonochem.*, 2021, 78, 105735, doi: 10.1016/j.ultsonch.2021.105735
- [56] Sarasua, J.A., Rubio, L.R., Aranzabe, E., Vilela, J.L.V., Energetic study of ultrasonic wettability enhancement, *Ultrason. Sonochem.*, 2021, 79, 105768, doi: 10.1016/j.ultsonch.2021.105768