Published in final edited form as:

IEEE Nanotechnol Mag. 2019 June ; 13(3): 13–28. doi:10.1109/MNANO.2019.2904773.

Candle Soot Carbon Nanoparticles in Photoacoustics: Advantages and Challenges for Laser Ultrasound Transmitters

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Abstract

This manuscript provides a review of candle-soot nanoparticle (CSNP) composite laser ultrasound transmitters (LUT), and compares and contrasts this technology to other carboncomposite designs. Among many carbon-based composite LUTs, a CSNP composite has shown its advantages of maximum energy conversion and fabrication simplicity for developing highly efficient ultrasound transmitters. This review focuses on the advantages and challenges of the CSNP-composite transmitter in the aspects of nanostructure design, fabrication procedure, and promising applications. Included are a brief description of the basic principles of the laser ultrasound transmitter, a review of general properties of CSNPs, as well as details on the fabrication method, photoacoustic performance, and design factors. A comparison of the CSNP-nanocomposite to other carbon-nanocomposites is provided. Lastly, representative applications of carbon-nanocomposite transmitters and future perspectives on CSNP-composite transmitters are presented.

LASER ULTRASOUND TRANSMITTER

The photoacoustic or optoacoustic effect refers to the generation of sound waves by absorbed light in a material [1]. Conventional photoacoustic imaging techniques involve

direct laser exposure of the imaging targets such as tissue and solid samples. In typical photoacoustic imaging, the whole photoacoustic phenomena, i.e. light absorption, thermal expansion, and thermoelastic wave generation, happen in the wave propagation medium [2]. In contrast, a photoacoustic transmitter, also called the laser ultrasound transmitter (LUT), is a device that uses the photoacoustic effect to generate ultrasound waves in place of piezoelectric elements, external to the imaging target. To do so, a layer of material that has higher photoacoustic conversion efficiency than typical acoustic media is added in the transducer stack. Thus, light absorption and wave generation occur in the transducer rather than the propagation medium [3]. The LUT's photoacoustic conversion is improved with high photothermal and thermoelastic properties of the metal layers or synthesized films [4]. Laser ultrasound transmitters have been developed for high power ultrasound ($>1 \text{ W/cm}^2$) applications [5]–[8], as well as photoacoustic imaging applications [9]–[12]. Typical high intensity focused ultrasound (HIFU) transducers have a low operation frequency (less than 5 MHz) with a large concave aperture (diameter larger than 50 mm) [13]. Although the acoustic beam is strongly focused by the low f-number (<1) concave lens, the focal volume is usually larger than micro-scale due to the low frequency and relatively large wavelength used ($>300 \mu\text{m}$). Moreover, most HIFU transducers exhibit a narrow bandwidth (-6 dB fractional bandwidth, $\text{FBW} < 30\%$) and long ringdown time (>5 cycles for -12 dB decaying), since the common HIFU operation condition involves high voltage ($>100 \text{ V}_{\text{rms}}$), long duty cycle ($>5\%$) excitation at the resonance frequency for applying sufficient acoustic energy to the target volume [14], [15]. These characteristics impede precise spatiotemporal control of ultrasound surgery or treatment. Recently, histotripsy, a cavitation-based therapy that uses very short (<10 cycles), high-pressure ($>20 \text{ MPa}$) ultrasound pulses has been used for the improved spatiotemporal control [16]. However, histotripsy still requires a large aperture ($>40 \text{ mm}$), low-frequency ($<3 \text{ MHz}$) focused transducer with high-voltage ($>400 \text{ V}$) operation [17]–[19].

To overcome these limitations, high frequency ($>10 \text{ MHz}$), wideband ($-6 \text{ dB FBW} > 100\%$), focused laser ultrasound transmitters have recently been developed. Since the excitation laser pulse has a very short pulse duration ($<10 \text{ ns}$), high frequency pulsed photoacoustic waves can be generated. These photoacoustic pulses generated from the smaller aperture concave lens ($<15 \text{ mm}$ in diameter) are focused at a tight focal volume ($<500 \mu\text{m}$ laterally) with high pressure amplitude ($>10 \text{ MPa}$) and broad bandwidth ($-6 \text{ dB FBW} > 100\%$) [5]. This high amplitude, short pulse duration is beneficial for precise spatiotemporal control (target volume $<1 \text{ mm}$ in lateral and $<1 \text{ cm}$ in axial) in ultrasound surgeries or treatments. Moreover, the laser ultrasound transducer is immune to electromagnetic interference, and does not involve electrical wiring issues, and thus is readily compatible with magnetic resonance imaging systems. The input laser energy can be controlled over a wide range ($1\text{--}200 \text{ mJ/cm}^2$) without dielectric breakdown caused by high voltage.

Several efficient LUT designs have been developed using carbon-based materials as a light absorption material, such as carbon black powders (CB) [3], carbon nanotubes (CNT) [5], [20], carbon nanofibers (CNF) [21], and candle soot carbon nanoparticles (CSNP) [22]. Transparent elastomers or epoxy materials with a high thermal expansion coefficient ($>200 \times 10^{-6} \text{ K}^{-1}$), such as polydimethylsiloxane (PDMS), are used for a thermoelastic layer. To date, multiwalled carbon nanotube (MWCNT)–PDMS composite is the most widely used

transmitter design in various applications, such as lithotripsy [5], cell-cleaving [23], sonic scalpels [8], microjetting devices [24], and fiber optic imaging [25]. The main advantages of the MWCNT-PDMS composite include a high photoacoustic conversion ratio (1.43×10^{-3} Pa/(W/m²)), fabrication diversity, and robustness against laser-induced ablation [26].

Despite the validated efficacy of the CNT-PDMS design, recent studies have aimed to discover a more efficient structure of carbon-based material with respect to fabrication simplicity, cost efficiency, and light absorption property. To simplify the carbon-coating procedure compared to the CNT growth method, a candle soot-PDMS composite transmitter (FIGURE 1) was developed. We introduced preliminary study results of CSNP-PDMS in several publications [22], [27]–[29], which present a competitive photoacoustic conversion ratio (4.41×10^{-3} Pa/(W/m²)) with a simple and cost-efficient fabrication method [22].

Here we provide a review of the CSNP composite LUT compared to other carbon-composite designs, which may be a useful guideline to design an optimal LUT with both maximum energy conversion and fabrication simplicity. This review focuses on the advantages and challenges of the CSNP-composite transmitter in the aspect of nanostructure design, fabrication procedure, and promising applications. We start by providing a brief description of the basic principle of the laser ultrasound transmitter, since a detailed review on theoretical background is available in previous review articles [2], [30]–[32]. Next, general properties of CSNP are summarized. Then, a detailed review on the fabrication method, photoacoustic performance, and design factors of the CSNP-nanocomposite in comparison with other carbon-nanocomposites is provided. We focus on the review on the CSNP-composite design comparing its features with other composite designs, since the recent articles by Lee *et al.* provides well-organized, comprehensive review on general polymer-nanomaterial composites for photoacoustics [33], [34]. Lastly, representative applications of carbon-nanocomposite transmitters are summarized, and the future perspectives on CSNP-composite transmitters are presented.

PRINCIPLE OF LASER ULTRASOUND TRANSMITTER

The main mechanism of the laser ultrasound transmitter is thermal expansion induced by the absorbed laser light. This is a series of energy conversion processes: optical to thermal energy, then thermal to mechanical energy [33]. For efficient energy conversion, a specific condition is required, which is called thermal and stress confinement. This means that the laser pulse duration should be shorter than the thermal relaxation time, which confines the heat, then maximizes the temperature in the irradiated volume [32]. In addition, the pulse duration should be shorter than the acoustical relaxation time [35]. Thus, rapid heating is an important prerequisite for efficient photoacoustic wave generation. Based on this prerequisite, the laser ultrasound pressure, P generated from a thin composite film can be simplified as eq. (1) by considering the long-pulse limit ($c\tau \gg 1/\mu_a$), where β , c , C_p , F , μ_a , E_{th} , and τ denote the thermal expansion coefficient, the sound speed, the specific heat capacity, the light fluence in J/cm², the absorption coefficient in cm⁻¹, thermal energy, and the laser pulse duration, respectively [2], [31], [33]. Γ is referred to as the Grüneisen coefficient ($\Gamma = \beta c^2 / C_p$), which is expressed by the selective thermal and acoustic material properties [2].

$$P \cong \left(\frac{\beta c^2}{C_p} \right) \frac{\mu_a F}{c \tau_l} = \Gamma \frac{\mu_a F}{c \tau_l} = \frac{\beta c}{C_p} \frac{E_{th}}{\tau_l} \quad (1)$$

Eq. (1) indicates that the laser-generated ultrasound amplitude is proportional to the specific material properties and irradiation condition. This relation provides some obvious and meaningful points for the laser ultrasound materials. For high pressure amplitude, high thermal expansion, high wave speed, and low heat capacity are required in the composite material of the transmitter. Additionally, it is obvious that high laser fluence with a short pulse duration input condition is required for high pressure amplitude [36]. Hence, for efficient light to thermal energy conversion, it is crucial to have great light absorption by the black-colored carbon particles followed by fast heat transfer to the surrounding thermal expansion medium. It has been highlighted that nano-size particles allow fast heat conversion to the surrounding medium. Thermal diffusion time, τ_{th} in a certain medium can be predicted by the following relationship [37], [38]:

$$\tau_{th} = d^2 / \kappa \alpha \quad (2)$$

where d is the diameter of a particle, α is thermal diffusivity, and κ is a constant varying with particle shape. Eq. (2) indicates that the thermal relaxation time is dependent on the particle size, material type, and particle shape. Firstly, by employing nanoscale particles, heat transfer induced by optical excitation occurs on the order of nanoseconds. This fast thermal diffusion property in nano-size particles allows the photoacoustic transmitter to operate in a relatively high frequency band, i.e. over 10 MHz [4]. Secondly, thermal relaxation time highly depends on material properties [39]. Particularly, carbon-nanomaterials, e.g. CB, CNF, and CNT, have an excellent thermal conductivity [40], [41], which is one of the main reasons why carbon particles are commonly used for photoacoustic transmitters. More details on carbon nanomaterials are described in the following section. Lastly, the thermal relaxation time is affected by the ratio of surface area to volume which is determined by the geometry of the nanoparticles. In other words, the geometric shape of the nanoparticle affects its heat conversion rate. For example, while the value of κ for a cylinder-like shape is 16 [42], [43], a spherical one has κ equal to 24 [44]. Hence, nano-size spherical particles would be more advantageous for efficient heat transfer compared to other shapes [28].

Another dominant parameter for efficient thermal to mechanical energy conversion is the thermal coefficient of volume expansion. Since the carbon material itself has poor thermoelastic property, additional elastomer material has been used as a thermal expansion layer. In virtue of its high thermal expansion coefficient ($\beta=920 \times 10^{-6} \text{ K}^{-1}$) and optical transparency below the infrared band, PDMS has commonly been utilized as an expansion medium. Some designs employed metallic materials or epoxy resins for the interfacial medium [3], [45], yet their thermal expansion properties are not competitive with PDMS.

Considering these optoacoustic properties, we describe the advantages and challenges of the CSNP-composites compared to other composites in the following sections.

CANDLE SOOT AND OTHER CARBON NANOMATERIALS

Burning candles has been an interesting research topic for more than 300 years, due to the complex combustion and soot formation processes [46]. A candle flame with incomplete combustion generates a cloud of black soot which consists of nano-size particles made of carbon (C 91.69%, H 1.75%, N 0.12%, O (calculated) 4.36%) [47]. Since it is easy to produce and collect the soot, candle soot particle is one of the most inexpensive and abundant carbonaceous materials [48]. Although it is not an entirely new type of nanomaterial, the candle soot nanoparticle has received increasing attention recently owing to its cost-effective optical sensors [51], electrocatalysts [50], superamphiphobic coatings [52], electrodes of solar cells [49], optical sensors [51], electrocatalysts [50], superamphiphobic coatings [52], electrodes of solar cells [49], supercapacitors [48], and lithium ion batteries [53]. Note that most of these works have focused on the porous nanostructure of the CSNP, which is a chain-like structure of spherical carbon beads with 20–50 nm diameter [22], [52]. The morphology of CSNP is shown in FIGURE 2 compared to the other carbon structures used in photoacoustics. An interesting finding about the CSNP from the previous studies is that the soot-deposition location (vertical position in the candle flame) changes the electromechanical properties of the collected soot [48]. The soot deposited at the center of the flame contains less carbonaceous material but more wax-like material, thus the coated soot is nonconductive and superhydrophobic [52]. On the other hand, the collected soot from the flame tip exhibits superhydrophilic and conductive properties with sufficiently oxidized carbon particles despite a low soot deposition rate [48], [53]. Although this variable property has not been studied for the photoacoustic application, it might affect candle soot adhesion on the substrate due to the remaining amount of wax-like material.

A nano-size absorber particle has a dominant effect on the fast thermal diffusion to the expansion medium resulting in high photoacoustic efficiency [42]. FIGURE 2 shows that the average size of CNT and CNF particles is nanoscale, whereas the CB powder has a relatively larger particle dimension in the range of nano- to micro-scale. Most of the CB powders used in laser ultrasound transmitters are commercially available, but detailed particle information is not available. Examples include Raven 2500 Ultra (Columbian Chemicals Company, Marietta, GA) [12] and Carbon black 73116 (Acrylicos Vallejo, Barcelona, Catalonia, Spain) [6]. Contrarily, CNF and CSNP are usually prepared by custom processes [21], [22]. The reported uses of CNT in laser ultrasound include both commercial products (724769, 6–9 nm $\times$ 5 μ m, Sigma Aldrich, UK) [9], [54] and custom CNT growth approaches [5]. The detailed fabrication methods of the carbon nanomaterials are described in the next section.

Particle sizes of the reported carbon materials for the laser absorber are summarized in TABLE 1. One distinctive feature of CSNP that is advantageous for efficient laser ultrasound generation is its spherical shape (FIGURE 2B). Among various types of carbon-based laser absorbers, the CSNP has the largest surface-to-volume ratio in virtue of its 3-dimensional structure. Hence, a relatively large passageway at the boundary of the light absorber material

and thermal expansion medium is provided for dissipating heat into the surrounding medium. For example, with the same volume ratio of carbon/expansion material, the contact area of CSNP is larger than CNF, which leads to a lower thermal resistance, and thus a higher thermal-to-mechanical energy conversion [22].

FABRICATION OF CSNP-PDMS AND OTHER COMPOSITE TRANSMITTERS

Carbon-based photoacoustic composite transmitters have been fabricated by general thin-film fabrication procedures. The current fabrication methods can be categorized into two different approaches: 1) separate coating processes for the carbon material and elastomer layer (for CNT, CNF, and CSNP composites) and 2) coating of the carbon-elastomer mixture at one time (for CB and CNT composites). TABLE 1 shows a summary of the published fabrication methods and structure of the carbon-nanocomposites to date. Typical coating substrate materials include normal glass, fused silica, and optical fiber. Depending on the coating substrate material and shape, different coating methods have been used, such as spin coating [28], dip coating [9], and electrospinning [25], among others.

For the fabrication of CB-composites, the commercial CB powders have been mixed with elastomer resin, e.g. PDMS, with a certain weight or volume concentration (20–70% of CB powder). This concentration determines the viscosity and laser absorption property [6], [12]. A concentration that is too low may result in low laser absorption ($< 60\%$), but a weight concentration higher than 70% causes poor spin-coating quality due to increased viscosity, which yields an uneven, thick coating of the CB-PDMS mixture [21]. A detailed study on the effects of the CB concentration on the photoacoustic performance has not been reported yet.

CNT-PDMS composites have been fabricated in two different approaches: separate coating (also called bilayer coating) and pre-mixed-solution-coating (integrated coating) methods. In the first use of CNT for photoacoustics by Baac *et al.*, multi-walled CNT were separately grown on a fused silica substrate. The substrate was coated by the catalyst (Fe and Al_2O_3) by sputtering, then the MWCNT were grown in a mixture of $\text{C}_2\text{H}_4/\text{H}_2/\text{He}$ at 775°C . Since the laser extinction by only the CNT layer is 60–70%, a 20 nm-gold layer deposition is then deposited, which results in laser absorption of 85%. After that, the PDMS-thermal expansion layer was spin-coated and cured [5], [42]. The highlighted feature of this method is a high-laser-ablation robustness. In comparison with other composites, the bilayer CNT-PDMS composite film endures the maximum laser energy (up to 477 mJ/cm^2) without laser ablation damage [26]. Most other composites have not been tested for their laser damage thresholds.

For the CNT coating on an optical fiber, Noimark *et al.* used both bilayer coating and integrated coating methods [9], [54]. In both methods, the MWCNT-xylene was used as an initial coating formulation. For the integrated coating, this coating formulation was mixed with xylene and PDMS for dip coating. For the bilayer coating procedure, the initial MWCNT-xylene formulation was additionally treated by sonication with acetone to produce the MWCNT-gel formulation. An optical fiber was inserted into the gel formulation for dip-coating then the coated fiber was dried. Next, the subsequent dip-coating with PDMS was conducted to complete coating of the MWCNT-gel-PDMS layer [9]. For the improved

absorber thickness control technique, an electrospinning method was tested [25], which is categorized into the integrated coating method. The electrospinning of MWCNT-polyvinyl alcohol (PVA) mixture facilitates precise thickness control in the range of 2.3–41.4 μm . Moreover, the PVA nanofiber (~200 nm) generated by this method exhibits a highly porous structure resulting in the fully encapsulated MWCNT by PDMS. Due to the improved contact condition between the absorber (MWCNT) and expander (PDMS), the electrospun-optical fiber exhibits higher photoacoustic performance (5-fold pressure amplitude) than the dip-coating reference sample [25].

For the CNF-PDMS composite transmitter, the CNF thin film is prepared first. Polyacrylonitrile (PAN) is added to N,N-Dimethylformamide (DMF), then stirred at 50°C for 24 h to obtain sufficient viscosity and homogeneity. The electrospinning of the mixture is conducted with a high voltage of 15 kV. After stabilizing at 250°C for 2 h in air, the as-spun fibers are carbonized in nitrogen at 900°C for 2 h with a heating rate of 2°C/min. After this carbonization procedure, the CNFs exhibit a paper-like thin film (~25 μm) structure [21]. The film is attached to a flat glass substrate, and then the PDMS was spin-coated. Although this bilayer coating method provides smooth coating of CNF-PDMS film on a flat glass surface, it is problematic for coating the CNF-composite on a concave surface. Since the paper-like shape of the CNF film cannot be smoothly attached to the concave surface without wrinkles, the unwanted thick and uneven PDMS coating occurs. This may be one reason why a concave-type CNF-transmitter has not been reported.

Motivated by simple, inexpensive, and easy fabrication of porous carbon nanostructures, a CSNP-PDMS composite fabrication method has been developed [22], [27]. The CSNP-PDMS composite has been fabricated by the bilayer coating approach: candle soot (CS) deposition followed by elastomer coating. The initially reported fabrication method involves the transplant of a spin-coated PDMS to the pre-deposited CS layer using two separate glass slides [22], because the uncured PDMS may remove the coated CS during the pouring and spin-coating. Later, it was demonstrated that direct spin-coating of PDMS on the CS-deposited glass does not cause serious damage on the CS layer by reducing the PDMS viscosity (1:100 weight ratio of toluene and PDMS) and gentle pouring [28], [29]. The detailed procedure starts with a flame synthesis using a paraffin wax candle (FIGURE 3A). The CS is collected on a glass slide at about 2 cm above the wick when the flame height is approximately 3 cm. CS thickness is determined by the deposition time showing a linearly proportional relation (FIGURE 3B). Next, the toluene-added PDMS (weight ratio of 1:100) is directly spin-coated on the CS-coated glass slide. After curing the PDMS at high temperature (> 65°C), the complete encapsulation of CSNP by PDMS is observed (FIGURE 3C). In addition to the glass substrate, CSNP have also been coated on an epoxy-type (NOA, UV-curing optical adhesive) microlens for developing optical fiber CSNP-PDMS composite transmitter [55].

The described fabrication procedures for various carbon-based composite transmitters present clear pros and cons. In terms of fabrication, the CSNP-composite transmitter features the following advantages:

1. The fabrication procedure is simpler and more cost effective than any other composite fabrication method; a commercial carbon material is not required, and there is no complex carbonization procedure.
2. Precise control of the laser absorber thickness is easily available; deposition time linearly affects the CS thickness (~1–25 μm thickness in 1–120 s).
3. More than 95% light extinction can be achieved, and an additional absorber coating procedure (e.g. Au-deposition) is not required [29].
4. CS can be deposited on a curved surface of the substrate with a consistent thickness and density.

There are also some challenges for the fabrication of CSNP-PDMS composites with further optimal designs.

1. Compared to other methods using a pre-mixed solution at room temperature, a coating substrate should be thermostable; during the candle flame synthesis, the substrate is required to endure a high temperature candle flame (approximately 800–1400°C) for 5–120 seconds.
2. Deposited CS on a glass surface typically has a weak adhesion property, which requires extra careful handling during PDMS spin-coating or dip-coating to prevent CS from washing away or forming agglomerations.

PHOTOACOUSTIC PERFORMANCES OF THE CSNP-PDMS AND OTHER CARBON-COMPOSITE TRANSMITTERS

One of distinctive features of carbon-PDMS composite transmitters is an outstanding pressure output (approximately >10 dB at least) compared to other metal-based composite transmitters [42]. Different designs of carbon nanomaterial-PDMS composite transmitters have been introduced showing various photoacoustic performances. The carbon-based laser ultrasound transmitters can be categorized into two designs: 1) flat design and 2) concave design. Typically, an initial prototype with a new carbon material was tested in a flat-aperture shape, and then the concave-type transmitter was developed for actual applications, mostly in therapeutic ultrasound.

The photoacoustic wave produced by the concave-substrate has been called laser-generated focused ultrasound (LGFU) [5]. By virtue of focusing a high frequency ultrasound beam, LGFU provides high pressure amplitude (>20 MPa) with high center frequency (>10 MHz) and tight focal volume (down to ~75 μm in lateral and ~200 μm in axial). The focal gain of the LGFU can be calculated by eq. (3), where f is the center frequency, c_0 is the wave speed, r is the radius of curvature, and f_N is the f-number which is defined as the ratio of the radius of curvature to the lens diameter.

$$G = \frac{2\pi f}{c_0} r \left(1 - \sqrt{1 - \frac{1}{4f_N^2}} \right) \quad (3)$$

With this focal gain, the output pressure amplitude can be significantly amplified at the focal volume. To obtain a high focal gain, it is important to generate the high frequency ultrasound wave using a low f-number transmitter. Various f-number plano-concave lenses have been used as a substrate for many different carbon-composite transmitters. The reported laser ultrasound transmitter specifications are summarized in TABLE 2. The estimated transmission sensitivity of each case is listed based on their pressure amplitude with the given laser fluence. Since the acoustic characterization setups and procedures in each study are different (e.g. pressure measurement distance, laser wavelength, pulse duration, hydrophone, etc.), precise comparative evaluation may not be possible in this review. It can be observed that the CNT-PDMS transmitter with a large diameter (15 mm) and short geometric focus (~9.2 mm) exhibited the maximum transmitting sensitivity of ~30 MPa/mJ among the entire carbon-composite transmitters [58]. This performance is contributed by both the high photoacoustic property of CNT-PDMS composite and high focal gain (220).

The flat design of chromium (Cr)-, CB-, CNF-, and CSNP-PDMS composite transmitters has been evaluated with consistent experimental conditions, such as laser fluence (3.57 mJ/cm²), measurement distance (4.2 mm), and hydrophone used (FIGURE 4) [22]. The comparison result shows the difference of the nanocomposite materials' photoacoustic efficiency regardless of focusing effect. It indicates that the CSNP-PDMS composite exhibits superior photoacoustic efficiency over Cr-, CB-, and CNF-PDMS composites.

Several studies reported photoacoustic energy conversion efficiency as a performance factor of photoacoustic transmitters [5], [21], [22], [27], [38]. The photoacoustic energy conversion efficiency can be determined by eq. (4), where ρ , c , A , and p denote water density, sound speed of water, the acoustic aperture area which can be considered as the laser beam size, and pressure amplitude measured by a hydrophone, respectively [29]. E_a is the acoustic energy, and E_{optical} is the input laser pulse energy.

$$\eta = \frac{E_a}{E_{\text{optical}}} = \frac{1/\rho c A \int_0^\infty p^2(t) dt}{E_{\text{optical}}} \quad (4)$$

It can be noted that the CSNP-PDMS composite exhibits higher photoacoustic conversion efficiency in comparison with other flat composite designs. The unique CSNP property contributes to this performance as briefly aforementioned. The superior performance of the CSNP-PDMS composite can be explained by its spatial configuration in nanoscale. The amplitude of the photoacoustic signal at nanosecond time scales is related to the temporal temperature gradient generated in the PDMS by the heat diffusion from the CNPs [63], [64]. The relatively low interfacial thermal resistance leads to a fast heat transfer and a temperature profile that closely resembles the profile of the laser pulse and shows the stronger photoacoustic signal. In contrast, the higher interfacial thermal resistance increases the nanoparticle temperature but decreases the rate of heat release into the adjacent medium (PDMS). Based on a one-dimensional model, the most effective elastic wave generation occurs when materials in or adjacent to the heated region have large values ($>200 \times 10^{-6} \text{ K}^{-1}$) of $\beta_T(D)^{1/2}$, where β_T is the thermal expansion coefficient, and D is the thermal diffusion

distance. Here, $D=(2\alpha/\omega)^{1/2}$ [65], where α is the thermal diffusion coefficient and ω is the angular frequency.

Among various types of carbon-composites, the absorber in CSNP-PDMS possesses the largest surface to volume ratio due to the three-dimensional nano-size of CSNP; therefore, a relatively larger passageway is available for dissipating heat into PDMS given the same volume ratio of the absorber to the PDMS matrix among various composites. At 10 MHz, the thermal diffusion distance in PDMS is 60 nm while as high as 11 μm in CSNP. The hierarchical and arborized structure of the CSNP allow the thermal energy to transfer to a certain distance within a short amount of time and dissipate into the surrounding PDMS more uniformly and quickly. CNT and CNF layers contain nano-size strands in two dimensions vertical to the fiber growth orientation; thus, energy could be radically transferred into PDMS with high efficiency. However, the contact area with PDMS is smaller than that of CSNP, leading to a higher thermal resistance and, thus, a lower photoacoustic conversion efficiency [22].

Our recent study showed that there is an optimal thickness of the CS absorber layer to maximize the photoacoustic pressure amplitude [29]. Since the CS thickness is controlled by the deposition time, the photoacoustic efficiency of the several different deposition cases (5, 10, 20, 30, 60, and 120 seconds) were evaluated. The 5 s-deposition-sample exhibited less than 1 μm -thick, poor CS coating with a low laser absorption property ($< 80\%$) at 532 nm. Once the CS thickness reaches about 2 μm by the longer deposition (10 s), the light absorption improves to 93% resulting in the maximum pressure output (FIGURE 5A). However, over this optimum thickness, the thicker the CS layer, the lower the pressure output observed (FIGURE 5B). As a result, the photoacoustic efficiency reaches a maximum with a 2 μm -thick CS layer, then exponentially decreases with thicker CS thickness (FIGURE 5C). This is because CSNP with 2 μm -thickness possesses complete infiltration of PDMS into the CSNP structure with lowest thermal resistance, which results in the maximum light absorption, heat diffusion, and thermal expansion with minimum attenuation of high-frequency content. Further redundant CSNP-PDMS thickness causes high thermal resistance and significant attenuation of high frequency content. A similar explanation has been provided with the study on photoacoustic effects of CNT density [26], [33]. For CNT growth, the strand length and density is required to be controlled, because too dense CNT forest impedes the complete infiltration of PDMS, which results in the poor heat dissipation and weak adhesion between the composite and substrate [26]. The CSNP-PDMS composite has been used for optical fiber-delivered LGFU, taking advantage of its simple fabrication characteristics [55]. One interesting design point is that the optical bond (NOA) has been used to build a custom small-diameter, transparent, concave lens (D : 2 mm, r : 1.3 mm). Device characterization has demonstrated a peak-to-peak pressure of 16 MPa with a center frequency of 12 MHz, and a -6 dB fractional bandwidth of 130% (FIGURE 5D) [55].

In terms of photoacoustic wave generation performance, the CSNP-PDMS composite has two advantages compared to other carbon-PDMS composites, as summarized below.

1. The CSNP-PDMS composite film generates a shock wave with the highest photoacoustic transduction efficiency (up to $9.02 \times 10^{-3} \text{ Pa}/(\text{W}/\text{m}^2)$) compared to

other carbon-PDMS composites due to the large surface to volume ratio. The efficiency of CSNP-composite is approximately six times higher than 2.6 μm thick-CNT composite and about 50 times higher than 105 μm -thick gold nanoparticles composite [22].

2. The photoacoustic shock wave generated from the flat CSNP-PDMS composite ($\sim 2 \mu\text{m}$ CS layer) has a higher center frequency than CB- and CNF-composites: 10 MHz vs. < 5 MHz. Since the higher frequency provides higher focal gain at a smaller focal volume, significant amplification by LGFU can be expected by using a CSNP-PDMS composite on a concave lens.

Despite the high photoacoustic transduction efficiency, one limitation is its unproven robustness against laser-induced ablation. In previous studies, only small fluence ($< 5 \text{ mJ/cm}^2$) was applied for acoustic characterization. Although the complete infiltration of PDMS into the thin CS-layer ($\sim 2 \mu\text{m}$) was demonstrated, it may be challenging to surpass CNT-PDMS composite's laser damage threshold of 477 mJ/cm^2 [26], due to the possible impurity (wax-like material) at the glass-CS boundary. The recently reported damage threshold for CSNP-PDMS on a planoconcave glass lens is $\sim 81 \text{ mJ/cm}^2$ [60]. Further investigation on the photothermal robustness of CSNP-PDMS and possible modification of CS-deposition procedure (e.g. deposition at the flame tip or using an ultrathin porous filter for particle-sorting [46]) is required.

APPLICATIONS OF LASER ULTRASOUND TRANSMITTERS

The first application of carbon-composite photoacoustic transmitters has been all-optical ultrasound imaging. To overcome a poor photoacoustic response in photoacoustic imaging, a black-PDMS layer was used for a laser-absorber/thermoelastic film [56]. The integrated photoacoustic device consisted of CB-PDMS transmitter (2 mm-diameter) surrounded by a Fabry-Perot (FP) polymer etalon receiver (FIGURE 6A) [66]. The prototype transducer exhibited an attractive pulse-echo response for high resolution imaging (center frequency > 30 MHz and bandwidth > 40 MHz). Recently, this transducer concept has been adopted for designing optical fiber photoacoustic probes for vascular tissue imaging [67]. For improved imaging sensitivity, CNT-PDMS composite was coated on the optical fiber tip (FIGURE 6B). The wideband detector was built on another optical fiber tip using a FP optical cavity. The miniaturized prototype transducer demonstrated a high center-frequency (> 20 MHz) and wideband (-6 dB fractional bandwidth $> 120\%$) pulse-echo response [10]. All-optical, *ex vivo* ultrasound imaging of swine aorta and carotid artery was demonstrated using this miniaturized probe mounted on a motion stage. The demonstrated imaging depth is 3.5 mm. The obtained axial resolution is $64 \mu\text{m}$ and the lateral resolution is $88 \mu\text{m}$ [67]. These studies on imaging applications mainly focused on the high-frequency and short-pulse (wideband) characteristics of carbon-nanocomposites instead of high-pressure amplitudes. On the other hand, LGFU transmitters' capability of generating high amplitude (negative peak $> 30 \text{ MPa}$) with high spatial resolution ($< 100 \mu\text{m}$) has been highlighted in other applications particularly in therapeutic ultrasound.

For therapeutic ultrasound, the high amplitude, high frequency photoacoustic shock wave produced by carbon-nanocomposites has received attention due to its potential for precise

spatiotemporal control of ultrasound treatments. Hence, the LGFU generated from carbon nanomaterial-PDMS composite on the concave glass has been used for lithotripsy [5], cell control [23], [68], drug-delivery [6], thrombolysis [7], and scalpel [8]. The performance of carbon-composite LGFU transmitters in various applications are summarized in TABLE 3. The most remarkable feature of LGFU for therapeutic ultrasound application is its capability of bubble nucleation by a single laser pulse. Since very high peak-negative-pressure ($\text{PNP} > 30 \text{ MPa}$ at free field & $\text{PNP} > 15 \text{ MPa}$ at a hard boundary) is required to nucleate a gas core in degassed water [58], [61], a high-focal-gain (> 100) design was utilized for these applications.

By using a high-focal-gain CNT-PDMS composite transmitter, it was successfully demonstrated that the shear stress induced by cavitation-based effects (microstreaming, microjets, and shock waves) can be precisely applied to a treatment target. For example, the most recent study showed that the CNT-PDMS LGFU transmitter enables precise, cavitation-induced treatment as a sonic scalpel (FIGURE 7A) [8]. The CNT-PDMS composite coated on a low f-number (0.61) concave lens generates PNP higher than 30 MPa at a tight focal volume (lateral dimension $< 100 \mu\text{m}$) which surpasses the free-field-cavitation threshold [58]. After observing the oscillating bubble at the focal spot of the LGFU, the mechanical effect of the cavitation was used to cut a tissuemimicking gel. Since the used LGFU is a short-pulse ($< 100 \text{ ns}$) shock wave with low PRF ($\sim 10 \text{ Hz}$), the cutting process is a non-thermal, solely cavitation-based treatment. As a result, precise gel-cutting with a kerf of $\sim 50 \mu\text{m}$ using the LGFU was demonstrated [8], which is a distinctive feature compared to previous works using similar CNT-PDMS LGFU but inducing cavitation at impedance boundaries with lower cavitation thresholds (PNP of $\sim 15 \text{ MPa}$) [61].

Using a slightly lowered LGFU amplitude (up to 12 MPa PNP) than the bubble-nucleation level, Kim *et al.* demonstrated the feasibility of LGFU for targeting biocompatible microbubbles and microparticles. Since the target bubbles or particles act as cavitation nuclei or impedance boundaries, the lower PNP in the range of 8–12 MPa was considered. Thus, CB-PDMS composite was used, which has a lower photoacoustic efficiency than CNT-PDMS but has the merits of low-cost and simple fabrication [6], [7]. For spatiotemporal drug delivery, LGFU was used as a stimulus for promoted drug release from microgels integrated with drug-loaded polymeric nanoparticles (FIGURE 7B) [6]. In both antitumor and antibacterial studies *in vitro*, the LGFU-treated cases exhibited a significant improvement (up to 270%) in drug release compared to the passive release cases. This result indicates that the LGFU with a PNP of 8–12 MPa can generate cavitation-induced mechanical effect at the alginate microgels boundary and near the poly(lactic-co-glycolic acid) (PLGA) nanoparticle drug carriers.

Moreover, the same design of CB-PDMS LGFU transmitter was used for microbubble-mediated thrombolysis (FIGURE 7C) [7]. The microbubble contrast agent (decafluorobutane core with a stabilizing phospholipid monolayer shell with polyethylene glycol) was utilized as a cavitation nucleus near the blood clot boundary. It was shown that the PNP of 8–12 MPa was sufficient to destroy the microbubbles, since the corresponding mechanical index (MI, defined as PNP in MPa divided by the square root of the operating frequency in MHz) is 2.1 to 3.2, which is much higher than the bubble destruction threshold ($\text{MI} > 0.8$) [69]. After

testing bubble destruction at a tight cavitation zone (down to 500 μm laterally), *in vitro* thrombolysis tests were conducted using bovine blood samples. The inertial cavitation-induced microstreaming, microjets, and shock waves are expected to induce shear stress on the target clot. As a result, a thrombolysis rate up to 1%/min was achieved by microbubble-mediated LGFU treatment [7]. Hence, it was demonstrated that a broadband, short pulsed (<100 ns) excitation can generate cavitation-induced mechanical effects at a small target spot.

In the applications of carbon-nanoparticles thus far, the CSNP-PDMS composite has not been tried for cavitation-based therapy. Although only the flat CSNP-PDMS design has been used, high pressure output (~ 30 MPa-PNP) can be expected by using a large aperture (~ 15 mm), low fnumber (~ 0.6) CSNP-PDMS LGFU transmitter due to high photoacoustic energy conversion efficiency. Thus, CSNP-PDMS LGFU transmitter can be applied for other ultrasound therapies that need high-precision and high efficiency.

SUMMARY AND FUTURE PERSPECTIVES

In this paper, we reviewed carbon-nanocomposite laser ultrasound transmitters, focusing on the candle soot carbon nanoparticles. The CSNP exhibits its merits in the aspects of nanostructure, fabrication, and photoacoustic performance. Compared to CB, CNT, and CNF, CSNP surrounded by PDMS has higher photoacoustic energy conversion efficiency due to its large surface-to-volume ratio. Moreover, CSNP is an inexpensive carbon particle, easy to produce, and can be deposited on a glass surface with a short deposition time (less than a minute). Since it is a relatively new carbon material in photoacoustics, several performance limits remain to be explored, such as maximum PNP and focal volume size achievable with a concave lens. It will also be required to confirm the photothermal robustness of the CSNP-PDMS composite to estimate the maximum pressure output by high fluence (>200 mJ/cm^2). Considering current advantages and challenges of the CSNP-nanocomposite, future perspective can be summarized as below.

1. The future CSNP composite can be designed with an optimal thickness of CS for the highest photoacoustic energy conversion efficiency (~ 2 μm CS by 10 s deposition). The optimal thickness-CS-PDMS composite can be used with a plano-concave glass substrate for high amplitude LGFU at a tight focal volume.
2. For high-fluence applications (>100 mJ/cm^2), the photothermal robustness of CSNP composites should be improved. To enhance adhesion of CS with a glass surface, additional primer material can be coated on the substrate, e.g. thin layer of wax or PDMS [71]. Furthermore, other type of a transparent substrate can be used instead of glass slides, such as optical bond (NOA), epoxy bond, and PDMS lens. As a substrate material and surface condition have a critical effect on the photothermal robustness, it is worth discovering an appropriate lens material.
3. To improve the photoacoustic efficiency, optimum CS deposition technique can be investigated, because property of carbon particles (size, structure and by-product composition) has a crucial effect on the photoacoustic efficiency. For example, CS can be collected at a different height of candle flame to deposit

CSNP with a different property. Moreover, a nano-sized porous filter can be used to collect smaller size CSNP [46],

4. In a recent photoacoustic study, an interesting technique to produce a user-defined wavefield at the specific imaging plane (e.g. shape of numeral '7') was introduced [72]. A tailored surface profile on a 3D-printed lens with a light absorber generates this customized wavefield. By adopting this technique and applying CSNP-composite coating on the tailored aperture profile, improved wavefield design with high SNR can be available due to the high amplitude, high frequency characteristics of CSNP-nanocomposites.

ACKNOWLEDGMENT

The authors would like to thank Isabel G. Newsome and Thomas M. Kierski for reviewing the manuscript. The lead author would like to thank Jaguar Lee for his contribution on a graphical abstract. This work is supported by NIH under Grants No. R01EB015508 and No. R01HL141967, and DOE under Grant No. DE-NE0008708.

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FIGURE 1.

Schematic of a candle soot carbon nanoparticle (CSNP)-elastomer photoacoustic transmitter. The absorbed heat from the pulsed laser through the glass substrate induces a thermal expansion of surrounding elastomer. The rapid expansion of the radiation surface generates a pulsed shock wave.

FIGURE 2.

Candle soot carbon nanoparticles (CSNP) and other carbon nanoparticles for laser ultrasound composite transmitters; (A) Photograph of CSNP preparation on a glass substrate, (B) Scanning electron microscopy (SEM) images of the coated CSNP on the glass in various resolutions, (C) Carbon black powder-PDMS thin film (SEM image, reused with permission of AIP Publishing [21]), (D) Transmission electron microscopy (TEM) image of carbon nanotubes (reused under a Creative Commons Attribution 3.0 Unported License [54]), (E) Carbon nanofiber-PDMS thin film (reused with permission of AIP Publishing [21]).

FIGURE 3.

Fabrication of candle soot carbon nanoparticle (CSNP) composite transmitter. (A) fabrication procedure (the schematic is not drawn to scale): CSNP deposition by flame synthesis, PDMS pouring, and PDMS spin-coating; (B) Relation between deposition time and CSNP thickness (reused with permission of IEEE [28]); (C) The coated results of CSNP (30 s-deposition) on a glass slide (left), and the cross-sectional view of the PDMS-coated CSNP-composite (right). This figure is reused with permission of IEEE [29].

Figure 4.

Acoustic characterization of flat Cr-, CB-, CNF-, and CSNP-PDMS composite transmitters.

(A) Experimental setup for measuring photoacoustic waves, (B) Measured photoacoustic pulses in time-domain, (C) Measured signals in frequency-domain. (reused with permission of AIP Publishing [22]).

FIGURE 5.

Photoacoustic performance of the CSNP-PDMS composite transmitters. (A) Measured waveform of the sample with 120 s-CS-deposition (corresponding CS thickness of $\sim 25 \mu\text{m}$, $1 \text{ mJ}/\text{cm}^2$ fluence) [29], (B) the sample with 10 s-CS-deposition (corresponding CS thickness of $\sim 2 \mu\text{m}$, $1 \text{ mJ}/\text{cm}^2$ fluence) [29], (C) The relationship between the photoacoustic transduction efficiency and the thickness of CSNP/PDMS composites [29], (D) the pulsed wave generated from the optical-fiber LGFU transducer (fluence of $1.5 \text{ mJ}/\text{cm}^2$) [55]. The figures are reused under permission of IEEE [29], [55].

FIGURE 6. Carbon-composite transducers for biomedical imaging application. (A) CB-PDMS composite transducer [66], (B) Optical fiber CNT-PDMS composite transducers [10], [67].

FIGURE 7.

Carbon-nanocomposite LGFU transmitters for therapeutic applications. (A) CNT/PDMS composite for invisible sonic scalpel (reused under permission of John Wiley and Sons [8]), (B) CB-PDMS composite for ultrasound-triggered drug delivery (reused under permission of Elsevier [6]), (C) CB-PDMS composite for microbubble-mediated sonothrombolysis [7].

TABLE 1.

Summary of carbon-composite transmitter fabrication methods and structure.

Design	Type	Fabrication method	Particle dimension	Composite layer thickness	Features
CB-PDMS [6], [7], [21], [56], [57]	Pre-mixed solution	<ul style="list-style-type: none"> Thermal decomposition or partial combustion method using hydrocarbons Commercial CB powder is mixed with PDMS Spin coating of CB-PDMS mixture (40–60% CB weight concentration, RPM: 2000–3000) and curing 	~25 nm–10 μ m diameter	25–30 μ m	<ul style="list-style-type: none"> Low cost carbon material Simple process Difficult to control the particle size Relatively low conversion efficiency
CNT-PDMS (CNT-growth) [5], [9], [42], [58]	Separated coating	<ul style="list-style-type: none"> Sputtering catalyst layers of Fe (~1 nm) & Al₂O₃ (~3 nm) Multi-walled CNT growth at high temperature (~770°C) chemical vapor deposition (CVD) Au layer deposition (~20 nm) Spin coating of PDMS (RPM: 2000) and curing 	~25 nm (tube diameter)	~16 μ m	<ul style="list-style-type: none"> High cost carbon material Complicated growth process Consistent strand diameter size Relatively high conversion efficiency Light extinction > 85%
CNT-PDMS (solution) [9], [54]	Pre-mixed solution	<ul style="list-style-type: none"> Functionalization of commercial CNT powder for dissolution in xylene Mixture of CNT-xylene and PDMS Dip-coating of the optical fiber (withdrawal speed of 2.8 mm/s) Room temperature curing (facing down) 	6–9 nm $\times$ 5 μ m	< 20 μ m	<ul style="list-style-type: none"> High cost carbon material Simpler process than CNT growth Relatively high conversion efficiency Light extinction > 97%
CNF-PDMS [21], [59]	Separated coating	<ul style="list-style-type: none"> Electrospinning of the polyacrylonitrile (PAN)-N,N-Dimethylformamide (DMF) solution High temperature (900°C) carbonization under a nitrogen atmosphere CNF thin film attached on the glass Spin coating of PDMS (RPM: 3000) and curing 	~133 nm (averaged diameter)	~58 μ m (~24 μ m of CNF film)	<ul style="list-style-type: none"> Complicated process Consistent fiber size Relatively high conversion efficiency Difficult to make a flat surface Difficult to attach on the concave lens: crumpled surface Light extinction > 90%
CSNP-PDMS [22], [27], [29], [55], [60]	Separated coating	<ul style="list-style-type: none"> Flame synthesis using a paraffin wax candle (20–120 s) 	~45 nm (averaged diameter)	16–25 μ m	<ul style="list-style-type: none"> Low cost carbon material

Design	Type	Fabrication method	Particle dimension	Composite layer thickness	Features
		<ul style="list-style-type: none">• Spin coating of PDMS (RPM: 2000–3000)• Dip coating of the miniaturized concave lens			<ul style="list-style-type: none">• Simple process• Consistent particle size• Relatively high conversion efficiency• Light extinction >90%

TABLE 2.

Specifications of the laser ultrasound transmitters hitherto reported. The laser wavelength and pulse duration are marked as λ and τ , respectively. The transmitting sensitivity is defined as ‘peak-to-peak pressure at the peak frequency/fluence.’ D and t denote the diameter and composite thickness, respectively. NOA is an UV-curing optical adhesive.

Design	Lens	D (mm)	Focal length (mm)	* Focal gain (G_{eff})	t	Max. fluence/pulse (mJ/cm ²) τ @ λ	Est. transmitting sensitivity (MPa/mJ) @ f_c	Photoacoustic conversion efficiency (Pa/(W/m ²))
CB-PDMS (Flat) [21], [22]	Glass	~12	N/A	N/A	30 μm	3.71 6 ns 532 nm	0.22–0.57 ~3 MHz	1.5–3.4 ($\times 10^{-4}$)
CB-PDMS (concave) [6], [7]	Glass	12–12.5	12.4	~90–99	~25 μm	17.4 6 ns 532 nm	1.5 ~14 MHz	
CNF-PDMS (Flat) [21], [22]	Glass	~12	N/A	N/A	~58 μm (~24 μm CNF film)	~3.5 6 ns 532 nm	0.7–3.27 ~3.5 MHz	0.4–1.66 ($\times 10^{-3}$)
CNT-Au-PDMS (CNT-growth, concave) [5], [24], [61]	Fused silica	6	5.5	~54	~16 μm	>177 6 ns 532 nm	~2.67 ~15 MHz	1.4 ($\times 10^{-3}$)
CNT-Au-PDMS (CNT-growth, concave) [5], [23], [62]	Fused silica	12	11.46	~100	~16 μm	>60 6 ns 532 nm	>1.4 ~15 MHz	
CNT-Au-PDMS (CNT-growth, concave) [8], [58]	Fused silica	15	9.2	220	~16 μm	~70 6 ns 532 nm	~30 ~15 MHz	
CNT-PDMS (solution) [54]	Optical fiber	0.2	N/A	N/A	~20 μm	~36.3 2 ns 1064 nm	~0.025 18 MHz	
CNT-PDMS (solution) [25]	Optical fiber	0.2	N/A	N/A	13.7 μm	35 2 ns 1064 nm	~0.05 31 MHz	
CNT-PDMS (solution) [9]	Optical fiber	0.2	N/A	N/A	~20 μm	~33.1 2 ns 1064 nm	>0.04 19.4 MHz	
CB paint (Concave) [10]	Glass	2	3.4	9.2	~20 μm	1.34 2 ns 1064 nm	~0.65 ~20 MHz	
CSNP-PDMS (Flat) [22]	Glass	~12	N/A	N/A	25 μm (6 μm CS & 19 μm PDMS)	~28 6 ns 532 nm	~28	4.41 ($\times 10^{-3}$)
CSNP-PDMS (Flat) [29]	Glass	~12	N/A	N/A	18 μm (2 μm CS & 16 μm PDMS)	~1 6 ns 532 nm	~2.5 ~10 MHz	9.02 ($\times 10^{-3}$)
CSNP-PDMS (Concave) [55]	NOA	~0.6	~1.3	~2.2	N/A **	~25 6 ns 532 nm	~1.07 ~13 MHz	

Design	Lens	D (mm)	Focal length (mm)	* Focal gain (G_{eff})	t	Max. fluence/pulse (mJ/cm ²) τ @ λ	Est. transmitting sensitivity (MPa/mJ) @ f_c	Photoacoustic conversion efficiency (Pa/(W/m ²))
CSNP-PDMS (Concave) [60]	Glass	6	4.71	~62	~24.5 μm (12.1 μm CS & 12.4 μm PDMS)	~25 6 ns 532 nm	~1.11 ~14 MHz	

* The effective focal gain, G_{eff} , is an approximated value considering the attenuation of water [5].

** The film thickness was not measured in the work [55].

TABLE 3.

Summary on the representative applications of nanocomposite laser ultrasound transmitter. Composite materials are presented in abbreviation: carbon black (CB), carbon nanotube (CNT), polydimethylsiloxane (PDMS). t , D and r denote composite layer thickness, diameter of aperture, and the radius of curvature, respectively.

Application	Design	Specification	Performance	Features
Lithotripsy [5]	CNT-Au-PDMS t : ~16 μ m plano-concave glass lens, D : 12 mm r : 11.46 mm	Input: 532 nm, 6 ns pulse, 20 Hz PRF, >40 mJ, >1000 pulses, Output: ~50 MPa (P_p), >13 MPa (P_N), f_c of ~15 MHz	Generated a 300–400 μ m hole on the surface of the artificial kidney-stone; destroyed line width of ~150 μ m on the moved kidney-stone sample	<i>In vitro</i> study; Medium: water; Cavitation effect at the boundary of the kidney stone sample; Precise treatment in microscale
Cell fractionation [23]	CNT-Au-PDMS t : ~16 μ m, plano-concave glass lens, D : 12 mm r : 11.46 mm	Input: 532 nm, 6 ns pulse, 20 Hz PRF, >50 mJ, ~32 s-treatment, Output: >50 MPa (P_p), >20 MPa (P_N), f_c of 15 MHz	Cleaved a densely packed cell cluster with ultrasonic sharpness of 100 μ m, Outward pushing effect	<i>In vitro</i> study; Medium: EDTA solution; Cavitation-induced liquid jet along the wall; non-contact, non-thermal modality
Drug delivery [6]	CB-PDMS t : ~25 μ m, plano-concave glass lens, D : 12 mm r : 12.4 mm	Input: 532 nm, 6 ns pulse, 10 Hz PRF, 18 mJ, 30 s-treatment, Output: ~14.5 MPa (P_p), ~8 MPa (P_N), f_c of 14 MHz	2-fold improved drug release compared to the control group (passive release); 3-fold improved by the 3-min treatment	<i>In vitro</i> study; Medium: degassed water, Eppendorf tube, PBS; Cavitation at the surface of alginate drug-carriers; Spatiotemporal remote control of drug release
Thrombolysis [7]	CB-PDMS t : ~25 μ m, plano-concave glass lens, D : 12.5 mm r : 12.4 mm	Input: 532 nm, 6 ns pulse, 10 Hz PRF, 20 mJ, 30 min-treatment, Output: ~18 MPa (P_p), ~12 MPa (P_N), f_c of 14 MHz	Approximately 30% mass reduction by the microbubble (10^8 bubbles/ml, 100 μ l/min)-mediated treatment.	<i>In vitro</i> study; Medium: degassed water, Tygon tube, PBS with microbubbles; Cavitation of microbubbles at the focal volume
Microjetting [24]	CNT-PDMS Transparent concave lens, r : 5.5 mm	Input: 532 nm, 6 ns pulse, 50 mJ/pulse Output: >20 MPa (P_N), Focal spot size of ~100 μ m	Primary jet speed: up to 50 m/s, Secondary jet speed: up to 200 m/s; Maximum jet height of ~500 μ m	Medium: water; Cavitation at the boundary between air and water; Nozzle free jetting system; Liquid printing
Ultrasonic scalpel [8]	CNT-PDMS Plano-concave glass lens, D : 15 mm r : 9.2 mm	Input: 532 nm, 6 ns pulse, 16 mJ, Output: >30 MPa (P_N), f_c of 15 MHz	Cutting of a tissue-mimicking phantom with a kerf width of ~45 μ m; Clear ablation volume at the organoid and pig eyeball samples	<i>In vitro</i> study; Medium: water; Microcavitation (<100 μ m) at the sample surface; Non-thermal, cavitation-based ablation;
All-optical ultrasound imaging	CB-PDMS [66] t : ~11 μ m; Flat glass; D : 2 mm; r : 18 and 12 mm	Input: 1064 nm, 5 ns pulse, 200 nJ/pulse, 5 kHz PRF Pulse-echo response @ 1 mm distance: f_c of 33 MHz –6 dB bandwidth of 41–51 MHz	Imaged three 50- μ m-diameter metal wires; All the 3 wires visible with the dynamic range of 10 dB	Synthetic aperture approach; All optical theta-array system (1D array transmitter & detection array along a circle (2.5 mm diameter));
	CNT-PDMS [9], [10] t : ~20 μ m, Flat optical fiber tip, CB paint, D : 200 μ m; Plano-concave glass lens, D : 2 mm, r : 3.4 mm	Input: 1064 nm, 2 ns pulse, 500 Hz PRF, 1.34 mJ/cm ² (for the concave lens); 100 Hz PRF, 96.8 mJ/cm ² (for the no-lens design); Output: 0.35–1 MPa, f_c >20 MHz	2D B-mode imaging by mechanical scanning; <i>ex vivo</i> aortic tissue imaging at depth of 2–9 mm, Clear boundary image (no quantification for the image contrast and resolution)	<i>Ex vivo</i> imaging; Optical fiber-directed miniaturized imaging probe; Non-coaxial alignment of the laser transmitter & optical detector; Useful element for a 2D optical fiber array for 3D endoscopic imaging [70]