Acoustical Characterisation of Carbon Nanotube-Loaded Polydimethylsiloxane Used for Optical Ultrasound Generation

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***Abstract*—An optical ultrasound generator was used to perform broadband ($2-35$ MHz) acoustical characterisation measurements of a nanocomposite comprising carbon nanotubes (CNT) and polydimethylsiloxane (PDMS), a composite that is commonly used as optical ultrasound generator. Samples consisting of either pure PDMS or CNT-loaded PDMS were characterised to determine the influence of CNTs on the speed of sound and power-law acoustic attenuation parameters. A small weight fraction ($<1.8\%$) of added CNTs was found to yield a prominent increase in the exponent of the power law, resulting in a significant increase in acoustic attenuation at higher frequencies. The speed of sound was found to be nearly identical, however. These results could prove useful in the numerical modelling and design of future optical ultrasound sources based on CNT-loaded PDMS.**

***Index Terms*—Optical ultrasound generator, polydimethylsiloxane, carbon nanotubes, acoustical characterisation, acoustical attenuation**

I. Introduction

Optically generated ultrasound has recently been highlighted as a viable alternative to conventional electronic transducers. Through the photoacoustic effect [1], excitation light is transduced within optically absorbing structures into ultrasound. An absence of metal and electronics results in acoustic sources that are MRI compatible and insensitive to electromagnetic interference, and compared to electronically generated fields, optically generated ultrasound can exhibit similar or better bandwidths and pressures [2]–[4]. Consequently, optical acoustic sources are ideally suited to broadband material characterisation [5] and detector calibration. In addition, optical ultrasound sources have been successfully applied to biomedical imaging [3], [6]–[8] and therapeutic settings [9], [10].

To achieve a high transduction efficiency, composite materials comprising a strong optical absorber and an elastomeric host material are typically used. Recent research efforts have focussed on the use of polydimethylsiloxane (PDMS) as a host material due to its high thermal expansion coefficient and its ease of fabrication and handling [3]. A variety of optical absorbers have been used, ranging from metals [11], [12] to carbon-based materials [3], [13]–[16].

Carbon-PDMS composites have been shown to exhibit strong acoustical attenuation [17], despite the small weight fractions that are typically used. In this work, broadband acoustical characterisation of composites comprising carbon nanotubes (CNT) and PDMS was performed using an optical ultrasound generator fabricated of the same material. Broadband measurements of both pure and CNT-loaded PDMS samples were performed to determine the effect of the presence of a small weight fraction of CNTs on the acoustical properties.

II. Methods

Experimental setup

The output of a pulsed laser (pulse energy: 50 mJ, repetition rate: 20 Hz, wavelength: 1064 nm, pulse duration: 8 ns; Ultra 50, Quantel, France) was expanded to a beam with a diameter of 25 mm, and delivered to a CNT-PDMS coating deposited on a glass slide where ultrasound was generated photoacoustically (Fig. 1). A calibrated needle hydrophone (200 µm, Precision Acoustics, UK) positioned at a distance of 18 mm was used to record the transmitted ultrasound, with samples placed orthogonally to the acoustic propagation path at a distance of 9 mm. Transmission measurements were performed without averaging.

Sample preparation

Samples of either pure PDMS (MED-1000, Nusil, CA, USA) or CNT-loaded PDMS were prepared and acoustically characterised. CNT-loaded samples were prepared using the method described in [3]. Briefly, multi-walled CNTs were first functionalised to allow for uniform dispersion in xylene, and then mixed with PDMS. A weight fraction of less than 1.8% of functionalised CNTs was added to the PDMS. Centimetre-scale samples of various thicknesses were fabricated (Fig. 2); pure PDMS samples measured 160, 260, 530 and 565 µm in thickness, whereas CNT-loaded samples measured 95, 275,

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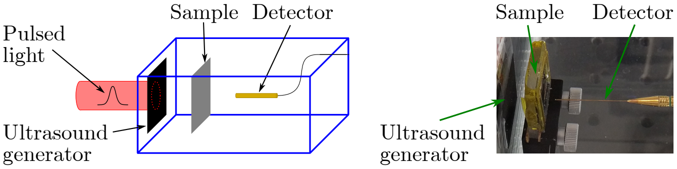

Fig. 1. Schematic (left) and photograph (right) of the acoustical characterisation setup. Pulsed light was absorbed in an optical ultrasound generator, where it was converted into broadband ultrasound through the photoacoustic effect. Transmitted ultrasound was detected using a calibrated needle hydrophone located at a distance of 18 mm from the ultrasound generator. The sample was placed centrally between the generator and detector, and placed orthogonally to the ultrasound propagation path.

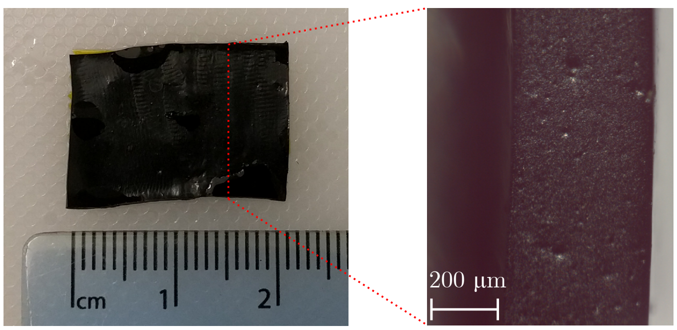

Fig. 2. Photograph (left) of a typical CNT-PDMS nanocomposite sample. Microscopic imaging (right) of the edge of a section revealed this sample had smooth surfaces and a thickness of 440 µm.

440 and 780 µm. The same composite material was used as optical ultrasound source, resulting in broadband (2 − 35 MHz) ultrasound generation.

Signal processing

The speed of sound was obtained from the time-of-flight difference between measurements through water and through a sample. The time-of-flight was determined using an automated picker [18], and the speed of sound was computed from

$$\frac{1}{c_w} + \frac{1}{c_s} = \frac{t_w - t_s}{d}, \quad (1)$$

where c_s is the speed of sound (m/s) in a sample with thickness d (m), $c_w = 1480$ m/s is the speed of sound in water at room temperature, and t_w and t_s (s) are the times-of-flight of transmitted ultrasound through water and the sample, respectively. For both pure and CNT-loaded samples, all four samples were measured and analysed.

The acoustical attenuation was determined by comparing the power spectra of the transmitted ultrasound through two samples of different thicknesses. By comparing different thicknesses, unknown reflection losses due to acoustical impedance mismatches were cancelled out. The attenuation parameter α was obtained by fitting a power law, $\alpha(f) = a \cdot f^b$ (dB/cm), to the power spectrum difference Δ,

$$\Delta(f) = \frac{S_1(f) - S_2(f)}{100\ (\mathrm{cm/m})\ \cdot (d_2 - d_1)}, \quad (2)$$

where S_1 and S_2 are the power spectra (dB) of the ultrasound transmitted through the thinner and thicker sample, respectively, d_1 and d_2 (m) are the thicknesses of the thin and thick samples, and f is the frequency (MHz). The scale factor a and exponent b were determined using a least squares fit across a $5 - 30$ MHz bandwidth. This analysis was repeated for both pure and CNT-loaded sample sets, and for all six possible thickness combinations.

III. Results

The presence of a small weight fraction of CNTs did not have a measurable effect on the speed of sound (Fig. 3). Across four samples of different thicknesses, the speeds of sound were found to be

$$c = 956 \pm 22 \text{ m/s} \quad \text{for pure samples, and}$$
$$c = 968 \pm 31 \text{ m/s} \quad \text{for CNT-loaded samples.}$$

As both pure and CNT-loaded samples exhibited the same speed of sound, and nearly the same density, nearly identical values for their acoustic impedances are expected.

Using a CNT-PDMS composite as optical ultrasound generator, acoustic characterisation measurements were performed across a noise-limited bandwidth of $2 - 35$ MHz, with power laws fitted between $5 - 30$ MHz. The addition of CNTs was found to have a prominent effect on the acoustical attenuation. Analyses performed on all six sample thickness pairs yielded the following attenuation parameters;

$$\begin{aligned} \alpha &= (1.60 \pm 0.07) \cdot f^{1.47 \pm 0.03} \text{ dB/cm (pure)} \\ \alpha &= (1.17 \pm 0.05) \cdot f^{1.61 \pm 0.09} \text{ dB/cm (CNT).} \end{aligned} \quad (3)$$

The seemingly small difference in the exponent can result in significant additional attenuation (Fig. 4): for a 500 µm thick coating an additional attenuation of 13 dB is expected at a frequency of 50 MHz.

IV. Discussion and Conclusion

To the authors knowledge, this was the first study in which broadband acoustical characterisation of a CNT-PDMS nanocomposite material was performed. This material is widely used in biomedical all-optical ultrasound imaging and therapeutic applications, as its performance is favourable for many applications. Indeed, in this study, precisely this material was utilised to generate the broadband acoustic pulses ($2 - 35$ MHz) used to perform the acoustic transmission measurements, at sufficiently high amplitudes to obviate signal averaging.

The small weight fraction of CNTs required for high optical absorption was observed to have a negligible effect on the speed of sound of CNT-loaded PDMS samples. However, a prominent increase in acoustical attenuation was observed. While the mechanism behind this increased attenuation is unknown, there are important implications for CNT-PDMS based sources. As the increased attenuation inherently limits the achievable bandwidth, the coating thickness could be reduced to broaden the generated bandwidth. Alternatively, two-part coatings comprising a thin layer of CNT-loaded

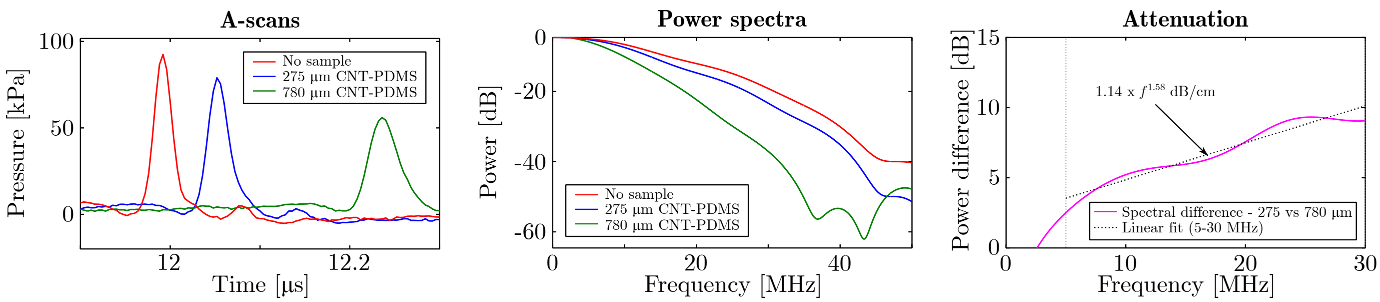

Fig. 3. Acoustical transmission measurements through water ("No sample") or samples of CNT-PDMS with thicknesses of 275 and 780 µm. The arrival times of the acoustic pulses (left) were used to determine the speed of sound in the samples, whereas power spectra of the acoustic pulse transmitted through the samples (middle) revealed the occurrence of acoustical attenuation. This attenuation was quantified (right) by fitting a power law to the power difference between transmission measurements through two CNT-PDMS samples of different thicknesses (shown on a logarithmic vertical scale).

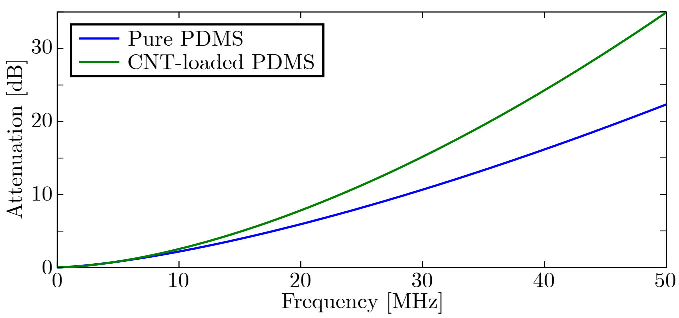

Fig. 4. Acoustical attenuation computed from the power laws of Eq. 3 for a coating thickness of 500 µm. Especially at high frequencies, the addition of carbon nanotubes (CNT) caused strong additional attenuation.

composite covered with a second layer of pure PDMS (which have previously been shown to generate wider bandwidths [3]) could be used to reduce the propagation distance through the more attenuative CNT-PDMS composite layer.

The small standard deviations of the measured speed of sound and attenuation parameters confirm the repeatability of the manufacturing process previously reported in [3]. The use of functionalised CNTs allowed for the fabrication of a large range of sample thicknesses. The measured acoustical properties did not depend on the measurement location across the sample (data not shown), which was indicative of sample homogeneity and surface smoothness. As the CNT-PDMS material used here is compatible with spin coating [19] and other fabrication methods such as moulding, it can be applied to a wide range of surfaces including those with geometries that are impractical to achieve with conventional electronic transducer materials.

Using the optical acoustic source presented here, a single ultrasound pulse per sample was sufficient to perform acoustic transmission measurements across a bandwidth of $2-35$ MHz. In contrast, with conventional electronic transducers several ultrasound sources would have typically been required to cover this bandwidth, resulting in multiple measurements and transducer manipulation. As such, a broadband optical acoustic source can greatly reduce the experimental duration and complexity.

This work demonstrated how the presence of CNTs in PDMS can have a prominent effect on its acoustical attenuation. The results found in this study can be used in numerical models to optimise the bandwidth and efficiency of future optical ultrasound sources.

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