

## Transient ink nucleation: the proof is in the pudding

Craig S. Carlson<sup>1†</sup>, Ryunosuke Matsumoto<sup>2</sup>, Koji Fushino<sup>2</sup>, Miryu Shinzato<sup>2</sup>, Nobuki Kudo<sup>2</sup>, and Michiel Postema<sup>1,3</sup> (<sup>1</sup>School Elec. & Inform. Eng., Univ. Witwatersrand, Johannesburg, South Africa; <sup>2</sup>Fac. Inform. Sci. & Technol., Hokkaido Univ., Sapporo, Japan; <sup>3</sup>BioMediTech, Fac. Med. & Health Technol., Tampere Univ., Finland)

### 1. Introduction

Although ultrasonic technology is most commonly used for medical imaging,<sup>1</sup> it is also regularly applied to manipulate,<sup>2</sup> deform,<sup>3</sup> and disrupt microscopic particles.<sup>4</sup> Ultrasound can disperse nanoparticles, a process which has been associated with inertial cavitation and acoustic streaming.<sup>5</sup> High-speed photography is commonly used to study acoustic cavitation dynamics.<sup>4,6</sup>

Hydrophobic particles can be regarded as particles with a thin gaseous layer surrounding the solid cores.<sup>7</sup> They have been observed to act as cavitation nuclei that lose their gaseous layer during sonication.<sup>8</sup>

Black tattoo ink comprises hydrophobic carbon black nanoparticles. In this study, we hypothesised that black tattoo ink exhibits similar transient inertial cavitation behaviour under sonication.

### 2. Materials and Methods

The first set of experiments was carried out in a Perspex container of 580x235x65 (mm)<sup>3</sup> internal dimensions, which was filled with degassed water.

A C60 5–2 MHz curvilinear probe of a SonoSite<sup>®</sup> M-Turbo<sup>®</sup> sonography device (FUJIFILM SonoSite, Inc., WA, USA) was clamped in the length direction of the container. The sonography device was operating in brightness mode throughout the experiments.

A 160x120x50 (mm)<sup>3</sup> tissue-mimicking phantom with three empty cylindrical wells, manufactured according to the recipe,<sup>9</sup> was positioned in the container such that its 160-mm-wide front face touched the probe surface. The cylindrical wells, with 28-mm diameter and 42-mm height, were located 20-mm from the front face (*cf.* Fig. 1).

Zuper Black pigment dispersion (INTENZE Products, Inc., NJ, USA) was used for black ink. 20 mL each of black ink and SABAX Pour Saline 0.9% (Adcock Ingram Critical Care (Pty) Ltd, Johannesburg, South Africa) was inserted into the centre and right wells respectively.

The ultrasound was set to a penetration depth of 6.6 cm, a mechanical index of 0.6, and a thermal

index of 0.1.

The second set of experiments was carried out in the 0.2-mL observation chamber of a high-speed photography system.<sup>10</sup>

A quantity of 0.5  $\mu$ L Zuper Black pigment dispersion was pipetted into a FALCON<sup>®</sup> 15-mL High-Clarity Polypropylene Conical Tube (Corning Science México S.A. de C.V., Tamaulipas, Mexico), after which 5 mL of 049-16787 Distilled Water (FUJIFILM Wako Pure Chemical Corporation, Osaka, Japan) was added. The tube was manually shaken for 5 minutes. From the tube, 0.2 mL was pipetted into the observation chamber, which was placed under an Eclipse Ti inverted microscope (Nikon Corporation, Tokyo, Japan) with a CFI S Plan Fluor ELWD 20XC objective lens of 20x magnification and 0.45 numerical aperture. Attached to the microscope was an HPV-X2 high-speed camera (Shimadzu, Kyoto, Japan), operating at 10 million frames per second.<sup>11</sup> During camera recording, the materials were subjected to ultrasound pulses.

The pulses consisted of 20 cycles, each at a centre transmitting frequency of 1.0 MHz and a peak-negative pressure of 400 kPa, from a laboratory-assembled single-element transducer.<sup>10,11</sup> The time between subsequent pulses was at least 1 minute. The signal fed into the transducer was generated by an AFG320 arbitrary function

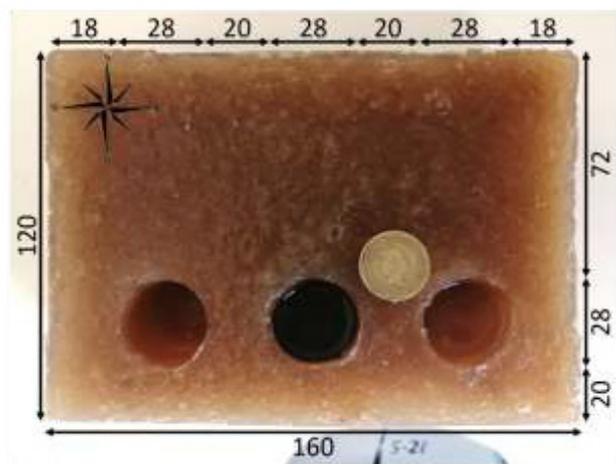


Fig. 1 Top view of the tissue-mimicking phantom with three cylindrical wells. The probe is visible on the South face. A £1 coin is included for scale. Dimensions have been stated in mm.

<sup>†</sup> Correspondence to: Craig.Carlson@wits.ac.za

generator (Sony-Tektronix, Tokyo, Japan) and amplified by a UOD-WB-1000 wide-band power amplifier (TOKIN Corporation, Miyagi, Japan).

A total number of 5800 images and 87 high-speed video sequences (each sequence consisted of 256 frames) were recorded during seven imaging sessions.

### 3. Results and Discussion

Brightness-mode scans of the phantom wells are shown in **Fig. 2**. Transient scattering was observed from individual spots inside the ink-filled well only (Fig. 2a, left). After 9 minutes, no more scattering was observed (Fig. 2b, left). This scattering can be attributed to the transient hydrophobic behaviour of carbon black, similar to the transient behaviour in an earlier study.<sup>8</sup> No scattering was observed in the saline-filled well. To observe this transient hydrophobicity at higher frame resolution, high-speed photography experiments were performed.

**Fig. 3** shows three frames of a representative example from the high-speed video recordings of black ink. The frames show multiple ink particles before, during, and after 1.0-MHz sonication. During sonication, inertial cavities were seen to nucleate on the ink particles, after which cavity

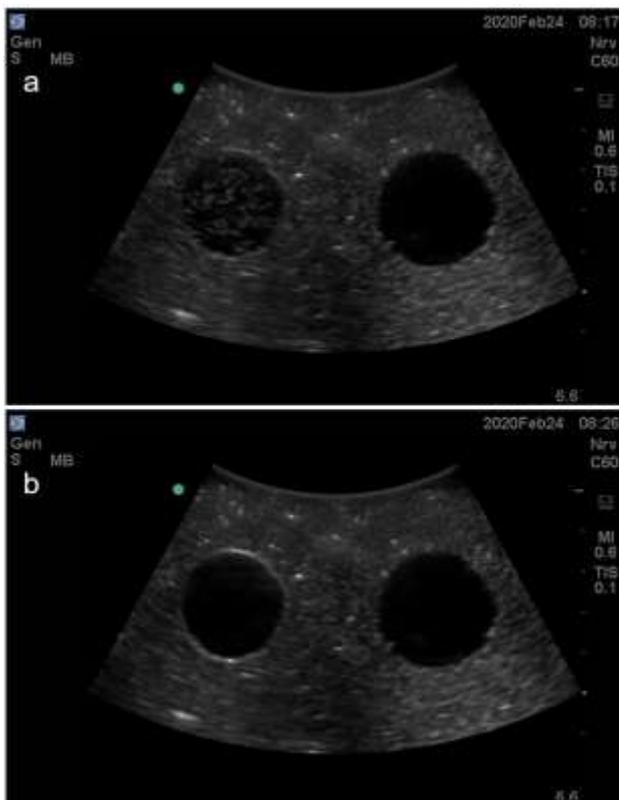


Fig. 2 Phantom wells filled with black ink (left) and saline (right), directly after filling (a) and 9 minutes later (b). The green dot corresponds to the left-hand side of the probe. Each tick corresponds to 1.1 cm.

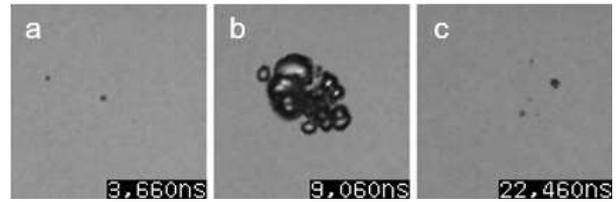


Fig. 3 Zuper Black ink particles before (a), during (b), and after (c) sonication. Each frame corresponds to a  $36 \times 36 \text{ } (\mu\text{m})^2$  area. Time stamps, in nanoseconds, are in the lower right corners.

clusters were generated from interaction with other nucleations. During subsequent ultrasound pulses, the ink particles that had been involved in this nucleation cycle remained unaffected (not shown). This supports the observation from the brightness-mode scans.

### 4. Conclusions

Using brightness-mode sonography and high-speed photography, black tattoo ink was observed to transiently nucleate. Subsequent ultrasound bursts did not reactivate the ink. It is concluded that ultrasound changes the hydrophobicity of carbon black ink particles.

### Acknowledgement

This work was supported by JSPS KAKENHI Grant Numbers 17H00864, 20H04542.

### References

1. S. Ishikura, M. Yoshizawa, N. Tagawa, and T. Irie, *Jpn. J. Appl. Phys.* **57** (2018) 07LF20.
2. T. Kambayashi, T. Saeki, and I. Buchanan, *Jpn. J. Appl. Phys.* **56** (2017) 07JE04.
3. J. Kim, J. Kim, J. Yeom, K. Ha, and M. Kim, *Jpn. J. Appl. Phys.* **56** (2017) 07JD04.
4. S. Nishitaka, D. Mashiko, S. Yoshizawa, and S. Umemura, *Jpn. J. Appl. Phys.* **57** (2018) 07LF25.
5. M. Kim and J. Kim, *Jpn. J. Appl. Phys.* **57** (2018) 07LE03.
6. K. Suzuki, R. Iwasaki, R. Takagi, S. Yoshizawa, and S. Umemura, *Jpn. J. Appl. Phys.* **56** (2017) 07JF27.
7. P. Attard, *Adv. Colloid Interface Sci.* **104** (2003) 75.
8. M. Postema, R. Matsumoto, R. Shimizu, A.T. Poortinga, and N. Kudo, *Jpn. J. Appl. Phys.* **59** (2020) SKKD07.
9. R.O. Bude and R.S. Adler, *J. Clin. Ultrasound* **23** (1995) 271.
10. N. Kudo, *IEEE Trans. Ultrason. Ferroelect. Freq. Control* **64** (2017) 273.
11. S. Imai and N. Kudo, *IEEE Int. Ultrason. Symp.* (2018) 184.