Jean de Bruin Jordaan, Michiel Postema, Craig S. Carlson*, and Ken J. Nixon

Quantification of carbon black tattoo ink hydrophobicity pre- and post-sonication

https://doi.org/10.1515/cdbme-2023-1003

Abstract: Despite growing academic interest for dermal inking in contemporary society, the reason why skin tattoos remain visible for life has predominantly been studied from a biological perspective. In preliminary physics studies of ink it has been presumed that the hydrophobicity of its main constituent prevents further dilution of pigment dispersion and therefore may be a contributing factor to aforementioned life-longevity. According to these early studies, ultrasound might change the hydrophobicity of microparticles. The purpose of this study was to confirm or refute the presence of hydrophobic components in carbon black tattoo ink and to relate sonication to such presence. Cuvettes with *n*-octanol, distilled water, and a droplet of unsonicated or sonicated carbon black ink were shaken, allowed to settle and subsequently photographed. The sonicated ink had been subjected to ultrasound during 5 min at a centre frequency of 1 MHz, a pulse-repetition frequency of 1 kHz, and a 10% duty cycle. The greyscale values in both parts of the cuvettes were averaged. The resulting ratio of light intensities was an indicator for the ink hydrophobicity. The intensity partition coefficient of carbon black ink was measured to be greater than 10^3 before sonication and less than 10^3 after sonication. Carbon black tattoo ink was found to be very hydrophobic. However, sonication was found to make the dispersion less hydrophobic. Influencing the hydrophobicity of tattoo ink might change the permanence of a skin tattoo.

Keywords: Pigment dispersion properties, octanol–water partition coefficient, colourimetry, hydrophobic particles, C65.

1 Introduction

Skin tattoos have become a common form of body decoration [1]. In Europe, an estimated 17% of the adult population has at least one tattoo [2, 3]. Approximately half of the people with tattoos regret at least one tattoo [4, 5]. Regrets may be related to medical complications [6–9]. Recent studies have reported on inflammation of tattooed skin associated with the COVID-19 pandemic [10, 11]. Notwithstanding upcoming EU-wide bans of green and blue tattoo inks [12], many reports on negative side effects concern black tattoo ink [13–15].

Scientific studies on why skin tattoos remain visible for life have concentrated on biology [16–18]. The reason why the alien materials that constitute tattoo ink keep their physical properties has been less studied. A straightforward, yet hypothetical, explanation why alien pigments only gradually fade might be that the pigment particles are protected by gas voids surrounding them, owing to their hydrophobicity. Black ink comprises carbon black pigment dispersed in a carrier medium. Previous studies on hydrophobic carbon black particles have demonstrated that exposure to ultrasound influenced the hydrophobicity of the particles [19, 20]. Preliminary experiments on carbon black pigment dispersion revealed individual particles acting as cavitation nuclei [21–23] and ultrasonic imaging artefacts in inked tissue [24]. Thus, it is known that carbon black ink comprises nuclei of transient inertial cavitation. Yet, its hydrophobicity has not been quantified, neither pre-sonication, nor post-sonication.

The purpose of this study was to confirm or refute the presence of hydrophobic components in carbon black tattoo ink and to relate sonication to such presence.

Hydrophobicity is quantified by the octanol-water partition coefficient [25, 26]. Materials whose partition coefficient is greater than 10^0 are hydrophobic and materials whose partition coefficient is less than 10^0 are hydrophilic. Extremely hydrophobic materials have partition coefficients greater than 10^4 [26]. Determining partition coefficients requires the precise measurement of concentrations. As carbon black pigment is dispersed in minute quantities, the measurement of absolute concentrations is rather challenging. As an alternative to absolute measurements, colourimetry may be reverted to [27], from which a coefficient can be computed that

---

***Corresponding author: Craig S. Carlson,** BioMediTech, Faculty of Medicine and Health Technology, Tampere University, Korkeakoulunkatu 3, 33720 Tampere, Finland and School of Electrical and Information Engineering, University of the Witwatersrand, Johannesburg, 1 Jan Smutslaan, Braamfontein 2050, South Africa, e-mail: craig.carlson@tuni.fi
Jean de Bruin Jordaan, Ken J. Nixon, School of Electrical and Information Engineering, University of the Witwatersrand, Johannesburg, Braamfontein, South Africa
Michiel Postema, BioMediTech, Faculty of Medicine and Health Technology, Tampere University, Tampere, Finland and School of Electrical and Information Engineering, University of the Witwatersrand, Johannesburg, Braamfontein, South Africa

Open Access. © 2023 The Author(s), published by De Gruyter. This work is licensed under the Creative Commons Attribution 4.0 International License.

is related to the light intensity fraction:

$$P_\mathrm{I} = \frac{I_{\mathrm{H_2O}}}{I_{\mathrm{C_8H_{18}O}}} \propto \frac{[c_{\mathrm{C_8H_{18}O}}]}{[c_{\mathrm{H_2O}}]}\,, \tag{1}$$

where $[c_{\mathrm{C_8H_{18}O}}]$ is the concentration of pigment dispersion in octanol, $[c_{\mathrm{H_2O}}]$ is the concentration of pigment dispersion in water, $I_{\mathrm{C_8H_{18}O}}$ is the intensity of light rays travelling through pigment dispersed in octanol, $I_{\mathrm{H_2O}}$ is the intensity of light rays travelling through pigment dispersed in water, and P_I is the intensity partition coefficient, which is typically converted to a logarithmic scale.

2 Materials and methods

For every experiment, eleven disposable plastic cuvettes (Hughes & Hughes Ltd., Romford, Essex, UK), each with a width of 9.9 mm and with an internal volume of 4 cm^3, were prepared to each contain quantities of 375 nl Zuper Black pigment dispersion (INTENZE Products, Inc., Rochelle Park, NJ, USA), 1.5 ml *n*-octanol (Hopkin & Williams Ltd, Chadwell Heath, Essex, UK), and 1.5 ml reverse osmosis distilled and degassed water (CJ Distribution, Midrand, South Africa).

The pigment dispersion in ten cuvettes had been subjected to five minutes of sonication at a centre frequency of 1 MHz, a pulse-repetition frequency of 1 kHz, and a 10% duty cycle in 0.25‰ dilution, using a custom-manufactured single-element very broadband transducer (Neoety AS, Kløfta, Norway) that had been calibrated for acoustic amplitudes up to 90 kPa.

A control cuvette was prepared according to the same procedure but without pigment dispersion.

All twelve cuvettes, i.e., ten with sonicated pigment dispersion, one with unsonicated pigment dispersion, and one without ink, were mixed using a PSS 200 AC Orbital Sander (Robert Bosch GmbH, Gerlingen-Schillerhöhe, Germany).

The cuvettes were placed inside custom-printed housing of polylactic acid (Ultimaker BV, Utrecht, Netherlands) in front of a white non-creasing paper background (Mondi, Bedfordview, South Africa) which was illuminated by an FCL-22CW″D Fluorescent Circular Tube (Galaxy Lighting & Brass Ltd., Richmond, BC, Canada).

Photos were captured 7′30″ after cuvette placement, using a LifeCam HD-3000 webcam (Microsoft Corporation, Redmond, WA, USA) that had been positioned 40 cm in front of the cuvette housing and whose complementary metal-oxide semiconductor had dimensions of 1280×720 pixels. The webcam was connected to a laptop computer for camera control and offline image processing.

A schematic line drawing of the experimental setup is shown in Figure 1.

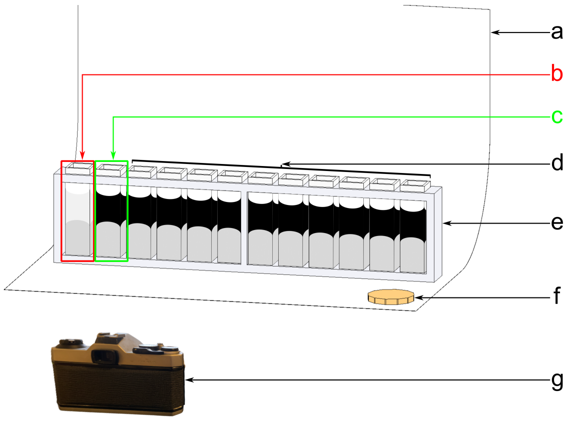

Fig. 1: Line drawing of the photography part of the experimental setup, composed of a non-creasing paper background (a), a control cuvette without ink (b), a cuvette with unsonicated ink (c), ten cuvettes with sonicated ink (d), a cuvette holder (e), a 1-SHP coin for scale (f), and a webcam (g).

The image processing part was done with an `onan` routine in MATLAB® (The MathWorks, Inc., Natick, MA, USA). Photographs were stored offline in bitmap format. Each photograph was converted to greyscale and cropped to exclude all areas outside the cuvettes. The greyscale values of all pixels in the *n*-octanol part and in the aqueous part of a cuvette were added separately, representing the average light intensity. For the average intensities inside each cuvette, the intensity partition coefficient was computed using (1), compensating for cuvette presence by dividing through by the control intensities. Forty eight intensity partition coefficients were measured, whose means and errors were calculated [28].

The position of the webcam was computed to influence the path lengths of the light through the suspensions by less than 2%. Therefore, the measured intensities were not compensated for these differences in light path length.

3 Results and discussion

Figure 2 shows a photograph of a typical experiment. The difference in intensities in the octanol-comprising parts and the aqueous parts was clearly visible. However, differences between cuvettes containing unsonicated or sonicated ink required precise measurements.

Figure 3 shows an overview of the intensity partition coefficients averaged over all experiments as a function of sonication amplitude. The logarithmic intensity partition coefficient of unsonicated carbon black dispersions was measured to be $\log_{10} P_\mathrm{I} = 3.3 \pm 0.2$. This value had dropped to $\log_{10} P_\mathrm{I} = 2.7 \pm 0.3$ for dispersions that had been sonicated for five minutes at a pressure amplitude of 10 kPa. The minimum

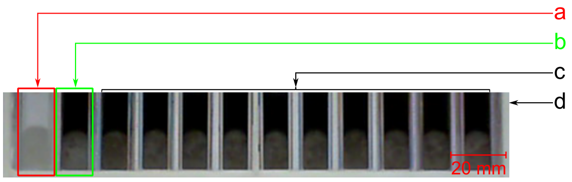

Fig. 2: Cropped frontal photograph of a representative experiment, showing a control cuvette without ink (a), a cuvette with unsonicated ink (b), ten cuvettes with sonicated ink (c), and a cuvette housing (d). The top of each cuvette comprised n-octanol, whereas the bottom comprised distilled water.

value was found to be $\log_{10} P_\mathrm{I} = 2.1 \pm 0.2$ for dispersions that had been sonicated for five minutes at a pressure amplitude of 50 kPa.

With the settings used in this study, the transmitted light intensity was clearly measured to drop with increased sonication amplitude. However, at sonication amplitudes greater than 50 kPa, no difference in intensity partition coefficients was found in these preliminary experiments. This counter-intuitive finding requires further study under different acoustic regimes and dispersion concentrations.

As the intensity partition coefficient P_I of carbon black ink was determined to be greater than 10^3 before sonication and less than 10^3 after sonication, the carbon black dispersion used in this study qualified as very hydrophobic, close to extremely hydrophobic.

It has not been established what the influence is of the carrier medium on the hydrophobicity of the ink. However, we do note that several organic compounds have partition coefficients similar to the intensity partition coefficients measured in our study [25]. It should be noted, however, that the intensity partition coefficient may differ from the actual partition coefficient.

4 Conclusions

Carbon black tattoo ink was found to be very hydrophobic. However, sonication was found to make the dispersion less hydrophobic. Influencing the hydrophobicity of tattoo ink might change the permanence of a skin tattoo.

Author Statement

Research funding: This work was supported by the Academy of Finland, Grant Number 340026, and by the National Research Foundation of South Africa, Grant Number 127102. Conflict of interest: Authors state no conflict of interest. Informed consent: Authors state that informed consent is not applicable. Ethical approval: Authors state that no ethical approval was required for this research as no human or animal samples or data were used. This manuscript was written according to recipe [29], without the aid of artificial intelligence.

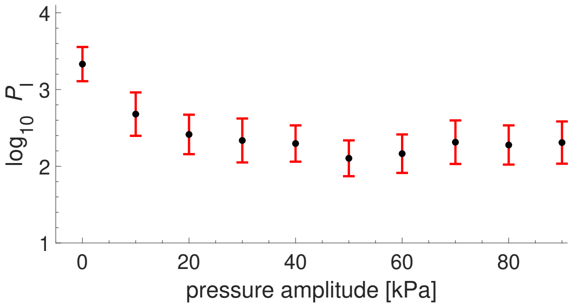

Fig. 3: Intensity partition coefficients of carbon black tattoo ink as a function of pre-applied ultrasound pressure amplitude. Coefficients have been represented on a logarithmic scale.

References

[1] Cherep MA. Barbie can get a tattoo, why can't I?: first amendment protection of tattooing in a Barbie world. Wake Forest L Rev 2011;46:331–357.
[2] Piccinini P, Pakalin S, Contor L, Bianchi I, Senaldi C. Safety of tattoos and permanent make-up: final report. EUR 27947 EN 2016.
[3] Battistini B, Petrucci F, De Angelis I, Failla CM, Bocca B. Quantitative analysis of metals and metal-based nano- and submicron-particles in tattoo inks. Chemosphere 2020;245:125667.
[4] Khosla V, Joseph V, Gordon H. Tattoos: what is their significance? Adv Psychiatr Treat 2010;16:281–287.
[5] Aslam A, Owen CM. Fashions change but tattoos are forever: time to regret. Br J Dermatol 2013;169:1364–1366.
[6] Klügl I, Hiller K-A, Landthaler M, Bäumler W. Incidence of health problems associated with tattooed skin: a nation-wide survey in German-speaking countries. Dermatology 2010;221:43–50.
[7] Khunger N, Molpariya A, Khunger A. Complications of tattoos and tattoo removal: stop and think before you ink. J Cutan Aesthet Surg 2015;8:30–36.
[8] Rahimi IA, Eberhard I, Kasten E. Tattoos: what do people really know about the medical risks of body ink? J Clin Aesthet Dermatol 2018;11:30–35.
[9] Maarouf M, Saberian C, Segal RJ, Shi VY. A new era for tattoos, with new potential complications. J Clin Aesthet Dermatol 2019;12:37–38.
[10] Wollina U. Inflammatory reaction to recently applied red tattoo ink after COVID-19. J Cosmet Dermatol 2022;21:7237–7239.
[11] Ramesh M, Hope L, Cockerell C, Hope R. Late onset pseudolymphomatous reaction to blue tattoo pigment precipitated by Covid vaccination. SKIN 2023;7:594–597.
[12] Serup J. Chaotic tattoo ink market and no improved costumer safety after new EU regulation. Dermatology 2023;239:1–4.

[13] Høgsberg T, Hutton Carlsen K, Serup J. High prevalence of minor symptoms in tattoos among a young population tattooed with carbon black and organic pigments. J Eur Acad Dermatol Venereol 2013;27:846–852.

[14] Hutton Carlsen K, Serup J. Photosensitivity and photodynamic events in black, red and blue tattoos are common: a 'beach study'. J Eur Acad Dermatol Venereol 2014;28:231–237.

[15] Mathis AK, Johnson BA. An impressive finding of sarcoid-like reaction to tattooing. Cureus 2023;15:e35401.

[16] König K. Multiphoton tomography of intratissue tattoo nanoparticles. Proc SPIE 2012;8207:82070S.

[17] Sepehri M, Sejersen T, Qvortrup K, Lerche CM, Serup J. Tattoo pigments are observed in the Kupffer cells of the liver indicating blood-borne distribution of tattoo ink. Dermatology 2017;233:86–93.

[18] Baranska A, Shawket A, Jouve M, Baratin M, Malosse C, Voluzan O, Vu Manh T-P, Fiore F, Bajénoff M, Benaroch P, Dalod M, Malissen M, Henri S, Malissen B. Unveiling skin macrophage dynamics explains both tattoo persistence and strenuous removal. J Exp Med 2018;215:1115–1133.

[19] Postema M, Matsumoto R, Shimizu R-i, Poortinga AT, Kudo N. Ultrasound homogenises suspensions of hydrophobic particles. Proc Symp Ultrason Electron 2019;40:2E2–4.

[20] Postema M, Matsumoto R, Shimizu R-i, Poortinga AT, Kudo N. High-speed footage shows transient ultrasonic nucleation of different hydrophobic particles in suspension. Jpn J Appl Phys 2020;59:SKKD07.

[21] Carlson CS, Matsumoto R, Fushino K, Shinzato M, Kudo N, Postema M. Transient ink nucleation: the proof is in the pudding. Proc Symp Ultrason Electron 2020;41:2E5–1.

[22] Carlson CS, Matsumoto R, Fushino K, Shinzato M, Kudo N, Postema M. Nucleation threshold of carbon black ultrasound contrast agent. Jpn J Appl Phys 2021;60:SDDA06.

[23] Carlson CS, Deroubaix A, Penny C, Postema M. On the attenuation of pure black tattoo ink. SAIEE Afr Res J 2021;112:24–31.

[24] Carlson CS, Postema M. Deep impact of superficial skin inking: acoustic analysis of underlying tissue. BIO Integration 2021;2:109–120.

[25] Wasik SP, Tewari YB, Miller MM, Martire DE. Octanol/water partition coefficients and aqueous solubilities of organic compounds. NBSIR-81-2406 1981.

[26] Jonker MTO. Determining octanol–water partition coefficients for extremely hydrophobic chemicals by combining "slow stirring" and solid-phase microextraction. Environ Toxicol Chem 2016;35:1371–1377.

[27] Kohl SK, Landmark JD, Stickle DF. Demonstration of absorbance using digital color image analysis and colored solutions. J Chem Educ 2006;83:644–646.

[28] Squires GL. Practical Physics. 4th ed. Cambridge: Cambridge University Press 2001.

[29] Carlson CS, Postema M. A pocket-sized recipe for cooking up a scientific manuscript. Tampere: Tampere University 2023.