ChemSusChem

Research Article
doi.org/10.1002/cssc.202400802

Chemistry Europe
European Chemical Societies Publishing

www.chemsuschem.org

Surface Cleaning of Oil Contaminants Using Bulk Nanobubbles

Helena H. W. B. Hansen+,[a, b] Lingxi Ouyang+,[a, b] Haotian Cha,[a, b] Jun Zhang,[a, c] Qin Li,[a, c] Beng Hau Tan,[d] Aditya Vashi,[a, b] Nam-Trung Nguyen,*[a] and Hongjie An*[a, b]

The removal of oil from solid surfaces, such as textiles and plates, remains a challenge due to the strong binding affinity of the oil. Conventional methods for surface cleaning often require surfactants and mechanical abrasion to enhance the cleaning process. However, in excess, these can pose adverse effects on the environment and to the material. This study investigated how bulk nanobubble water can clean oil microdroplets deposited on surfaces like glass coverslips and dishes. Microscopy imaging and further image analysis clearly revealed that these microdroplets detached from both hydrophobic and hydrophilic surfaces when washed with bulk nanobubble water within a fluidic microchannel. Oil contaminant cleaning was also conducted in water as mobile phase to mimic the circumstances that occur in a dishwasher and washing machine. Cleaning on a larger scale also proved very successful in the removal of oil from a porcelain bowl. These results indicate that nanobubble water can easily remove oil contaminants from glass and porcelain surfaces without the assistance of surfactants. This is in stark contrast to negligible results obtained with a control solution without nanobubbles. This study indicates that nanobubble technology is an innovative, low-cost, eco-friendly approach for oil removal, demonstrating its potential for broad practical applications.

Introduction

Effective removal of contaminants from surfaces and bulk liquids is notoriously difficult and time consuming due to the inherent affinity of contaminants to surfaces, and their immiscibility with many cleaning solutions. They are traditionally removed by addition of detergents, which reduce the surface tension between the contaminants and bulk liquids, so less energy is required for complete removal. However, excessive use of detergents often poses environmental risks in addition to providing a limited increase in cleaning efficacy.[1] To pursue more sustainable and efficient contaminant removal, emerging nanotechnologies have been requisitioned with great success in forms such as hydrogels and nanobubbles.[2] No extra surfactants or detergents are required, making them more environmentally friendly compared to chemical or microbiological methods, which are also restricted in their uses.[2b] In addition, there is less damage to the materials as no scrubbing or other harsh interactions are necessary.

Nanobubbles are small gaseous bodies that can exist either in bulk or on surfaces that provide unique advantages in surface cleaning applications.[2a,3] Nanobubbles are remarkably stable, persisting for long periods of time, ~a few weeks, contrary to the theories of bubble dissolution.[4] Due to their small size, their movements are based on Brownian motion rather than buoyancy, and are therefore evenly distributed throughout the solution.[5] Generation of nanobubbles can be relatively simple, often requiring minimal or non-invasive methods such as hydrodynamic flow via a venturi tube, or by electrolysis.[6]

Recent studies have shown the promises of electrolysis generated bulk nanobubbles in surface cleaning applications. Electrolysis has emerged as a novel way for generating nanobubbles as gas is generated directly in the desired solution, such as water. No harsh chemicals are required making this a clean method for gas production, with the addition of being cheap. Zhu et al.[2a] applied electrolysis generated bulk nanobubbles for removal of proteins on a surface, demonstrating the ability of bulk nanobubbles to remove biological contaminants as well as preventing re-fouling.

Building upon previous work, our study focuses on exploring the effectiveness of nanobubble solutions in removing lipid and vegetable oil deposits from surfaces. Oils are generally sticky and difficult to detach from dishes and clothes with conventional dishwashers or washing machines. If washing dishes by hand, excessive abrasion is oftentimes necessary. The removal of oils and fats from clothes is especially tricky as many detergents are made to function on liquid oils, so cold washes are not an option. In such cases, enzyme based detergents can be used, but even then some stains can only be properly

[a] H. H. W. B. Hansen,+ L. Ouyang,+ H. Cha, J. Zhang, Q. Li, A. Vashi, N.-T. Nguyen, H. An
Queensland Micro- and Nanotechnology Centre, Griffith University, 170 Kessel Road, Nathan, QLD 4111, Australia
E-mail: nam-trung.nguyen@griffith.edu.au
Hongjie.an@griffith.edu.au

[b] H. H. W. B. Hansen,+ L. Ouyang,+ H. Cha, A. Vashi, H. An
School of Environment Science, Griffith University, 170 Kessel Road, Nathan, QLD 4111, Australia

[c] J. Zhang, Q. Li
School of Engineering and Built Environment, Griffith University, 170 Kessel Road, Nathan, QLD 4111, Australia

[d] B. H. Tan
Shenzhen Institute of Advanced Study, University of Electronic Science and Technology of China, Shenzhen 518110, China

[+] Contributed equally.

removed via abrasion.[7] Enzyme based detergents, however, are not suitable for wool fabrics which, in addition to requiring a cold wash, cannot be subjected to a high level of friction. In such cases specialized techniques and detergents are needed. In addition, detergents may also cause skin irritation. For example, detergents that have an alkaline pH may negatively affect the physiological pH of the skin. Repeated use of detergents has also been shown to result in allergic or otherwise adverse reactions.[8]

Herein, we show that nanobubbles generated during water splitting with a bias can detach pre-deposited vegetable oils from hydrophilic and hydrophobic surfaces in a microchannel setup. We also investigated the efficacy of large-scale oil removal on glass and porcelain using nanobubbles. Lastly, we briefly investigated the interactions between our oil emulsions and our nanobubble solution. We hypothesize that the oil-nanobubble interactions will be preferred by the system as previous studies have shown that hydrophobic interactions are able to stabilize nanobubbles.[9] This preferred interaction will then aid in the removal of oil deposits.

Materials and Methods

Device Design and Fabrication

In this work, we applied a straight channel with one inlet and one outlet. The width and height of the channel were 200 and 50 μm, respectively. The microfluidics channel was fabricated using the standard photolithography and soft-lithography technologies.[10] Briefly, a Polydimethylsiloxane (PDMS) was made using SYLGARD™ 184 (Dow Corning) silicone elastomer base was mixed 10:1 with SYLGARD™ 184 silicone elastomer curing agent. Then, the mixture was placed in a vacuum chamber for 20 mins for degassing before being poured over the microchannel silicon mold. This was then placed in an oven at 70 °C for over 2 hours. After being peeled from the mold, the inlet and outlet holes were punched with 1 mm hole puncher. Finally, the PDMS layer was bonded to a glass substrate using Plasma Cleaner (Harrick Plasma). Tubing was attached to the inlet and outlet of the channel, and the tubing from the inlet was further attached to a syringe placed in a syringe pump.

Contaminated Surface Preparation

An oil emulsion was generated by first dissolving sunflower oil (Sigma–Aldrich) in ethanol at a concentration of 1:3, followed by mixing this solution 1:4 with DI water using mechanical agitation. This resulted in a milky mixture containing oil droplets. The produced solution was pumped into a 200 μm linear PDMS channel. After a rest period of 30 min and 3 hours, for the hydrophobic and hydrophilic channel respectively, oil droplets were deposited onto the glass surfaces. Then the solutions were displaced by clean water to remove all the remaining oil suspensions. The original oil was also directly deposited on glass coverslips and porcelain bowls for dip-washing in a beaker. The contaminated coverslips were dyed with Nile Blue (Sigma-Aldrich), which can be excited by green light and observed using fluorescence microscopy. Cooking oil (Sunflower oil, 1 gram) was deposited on the porcelain plates, and after 30 minutes, the plates were gently rinsed with Milli-Q water to remove excess oil, following by washing and imaging.

Nanobubble Water Generation and Characterization

Nanobubbles were generated via an electrochemical method. To split water, a DC bias from a DC power supply (15 V, LW-K3010D, China) was applied between two platinum mesh electrodes (Ileka Metal Materials Business Department, China) immersed in water in a beaker with a magnetic stirrer.

The concentration versus size distribution of nanobubbles in the generated nanobubble water was characterized by nanoparticle tracking analysis (NTA, Malvern Panalytical). Before use, as well as between each sample, the NanoSight flow cell was cleaned using a specific method. The samples were diluted in accordance with proper NanoSight practice and ten measurements of 15 seconds each were taken for each sample volume. The NanoSight provides the concentration in particles per milliliter and as such the bubbles are referenced as particles in related figures. NTA was also used to assess the interaction of nanobubbles with oil nanodroplet emulsions.

Experimental Setup

The microfluidic chip was placed on the stage of an inverted microscope (Olympus IX73 microscope). A syringe pump (SHENCHEN ISPLab02) infused our solution into the device at a fixed flow rate of 5 μl/min. Videos were captured by an sCMOS camera (PCO Edge 4.2) with a frame rate of 2 frames per second (fps). The open-source software ImageJ Fiji (National Institutes of Health) was used to analyze the recorded videos. Generation of the hydrophobic channel was achieved by pumping Glaco Mirror Coat Zero through the channel for 5 mins. After 10 min, air was gently pumped through to ensure the coating dried properly as per manufacturer instructions. For the hydrophilic channel, no further preparation was done after the plasma treatment as this process results in a hydrophilic surface. The channel was first washed with the control solution, followed by the nanobubble solution at a flow rate of 5 μL/min. A schematic of the setup can be seen in Figure 1a.

Dip cleaning Under Motion

To better simulate a cleaning environment, an oil contaminated glass coverslip and porcelain bowl was submerged in Milli-Q water with or without nanobubbles, Figure 1b. The nanobubbles were generated as described previously. The contaminated glass coverslips were imaged after 0 min (before introduction to the cleaning solution), 1 min, 2 mins, 5 mins, and 10 mins of washing/stirring and the porcelain bowl was imaged after 0 min (before introduction to the cleaning solution), 1 min, 2 mins, 5 mins, 10 mins, 20 mins, and 30 mins of washing/stirring.

Results and Discussion

In this study, we generate nanobubbles using the same methods described by Zhu et al.[2a] Zhu et al. reported that electrolyzed nanobubble water can efficiently remove proteins deposited on silicon surfaces. From this we hypothesize that bubbly water can remove oil contaminants from surfaces as bubbles are hydrophobic, while oils are lipophilic. To demonstrate this, we tested the removal of oil deposits in hydrophobic and hydrophilic microchannels, as well as in a larger system under constant motion to simulate a better washing environment, Figure 1.

Research Article
doi.org/10.1002/cssc.202400802

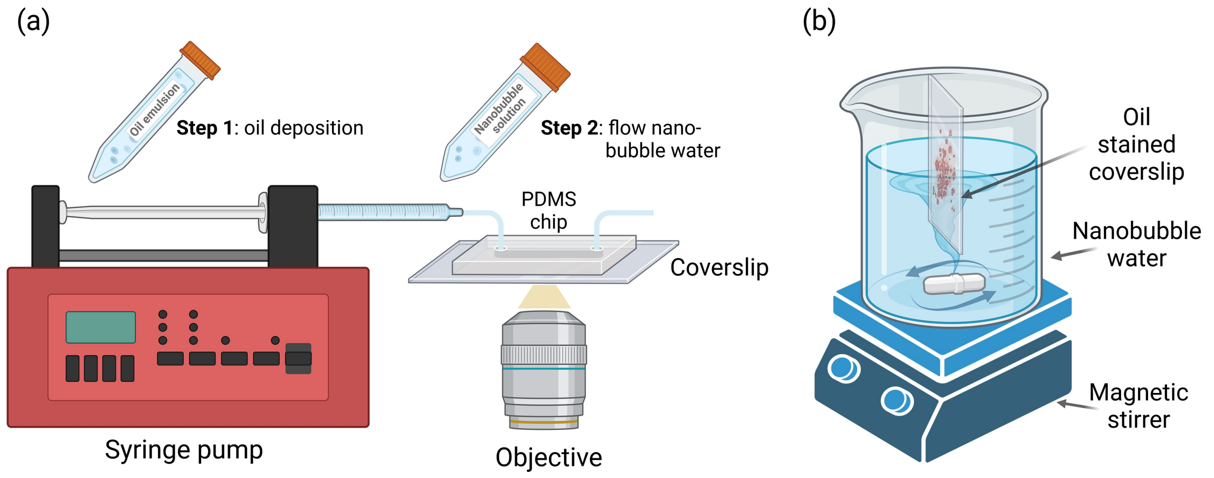


Figure 1. Schematic of oil cleaning processes via (a) a microfluidic channel, and (b) mechanical washing. Oil deposition on a glass coverslip was followed by washing with first a control solution without nanobubbles, and then a solution containing nanobubbles. Made with Biorender.com.

The generated nanobubble water was characterized using NTA. Figure 2 shows the nanobubble concentration versus size distribution, with untreated Milli-Q water as a control. Nanobubble water clearly shows a peak at ~100 nm with a concentration of 10.4×10^6 particles per mL, and a total bubble concentration of 7.78×10^8 particles per mL. In contrast, Milli-Q water has a negligible particle concentration. From this we can confirm that our control samples contain next to no nanobubbles. In addition, this verifies that we had bubble generation in the nano range using electrolysis.

Hydrophilic Surface

To generate a hydrophilic microchannel a glass coverslip was treated with oxygen plasma. Thereafter, the channel was washed with a control solution without nanobubbles (Figure 3a) followed by the nanobubble-containing solution (Figure 3b). From this figure we observe that the oil deposits on the surface are not greatly affected by the control solution. In comparison, using a solution containing nanobubbles resulted in substantial removal of the oil deposits; ~80% in less than one minute (Figure 4). Even at the 45 s mark the large deposit seen on the right in the middle panel is almost gone, and at 90 s there are no large deposits left. In the last two images of Figure 3b the oil deposits start to become unclear. As the focus was constant throughout the measurement, this indicates that these deposits are being worn down by the nanobubble solution.

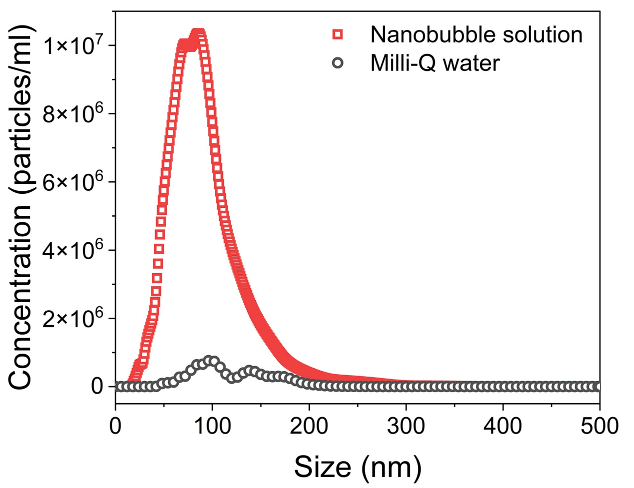


Figure 2. The size distribution and particle concentration of the nanobubble solution (red) and Milli-Q water (black) from NTA.

Oil droplets in aqueous solutions on a hydrophilic surface would form spherical caps due to the dewetting effect, and thus be easily removed by aqueous solutions. The dewetting ability is largely dependent on the hydrophilicity of the surface, which has preferential binding of aqueous material. In contrast, even though some oil deposits may be removed, it was not obvious from the videos until nanobubbles were introduced. Images became blurry once nanobubbles came into the imaging area, indicating a sign of bubble-oil droplet binding. Figure 4 presents the percentage of oil removed on the hydrophilic surface. This figure clearly demonstrates that nanobubbles are necessary for effective oil removal. At 90 s, ~85% of the initial oil deposits had been removed using nanobubbles, whereas very little change was observed for the control; only ~7% was removed. This suggests that even with surface modifications to repel hydrophobic materials such as oil, successful removal is still a challenge using normal water without nanobubbles.

Hydrophobic Surface

Generating the hydrophobic channel was achieved using Glaco and the results of the washing can be seen in Figure 5. As with the hydrophilic channel, washing with the control had little effect on the deposited oil, whereas, once nanobubbles were introduced, oil removal was observed. Compared to the hydrophilic channel, it took almost twice as long before there was any noticeable oil removal. We quantified the oil removal in

Research Article
doi.org/10.1002/cssc.202400802

Chemistry Europe
European Chemical Societies Publishing

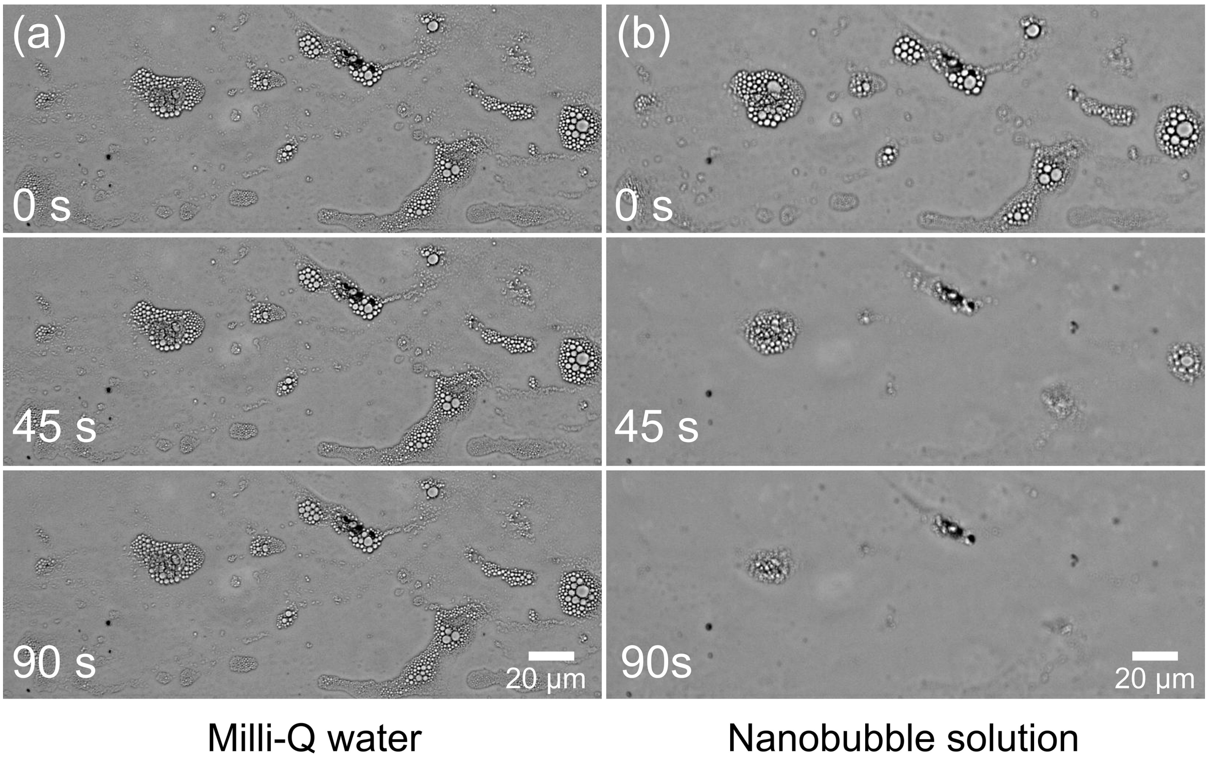


Figure 3. Hydrophilic channel with oil deposits washed with Milli-Q water (a) without nanobubbles (control) and (b) with nanobubbles. Representative images taken at 0 s (before introduction to the solution), 45 s and 90 s. The scale bar is 20 μm.

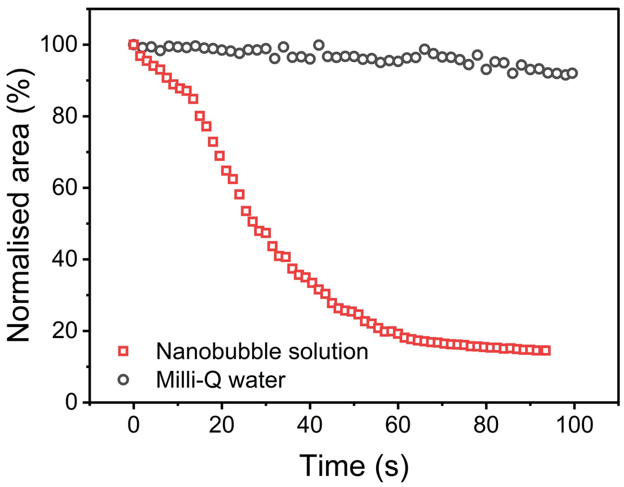


Figure 4. Percentage of oil on the hydrophilic surface over time. Data was normalized with respect to total area of oil at 0 s. Wash with Milli-Q water indicated by black circles and wash with the nanobubble solution indicated by red squares.

Figure 6 and observed that at 90 s, where ~85% of the oil had been removed on the hydrophilic surface, only ~40% had been removed on the hydrophobic surface. After an additional 90 s, the nanobubble solution further removed ~20% of the initial oil. The nanobubble solution was still superior to the control solution as no markable change was observed for this sample over the course of the experiment. In Figure 6, we observe a slight increase in total oil on the sample surface over time which may be due to relocation of oil from downstream. The shear force from the Milli-Q water flow detaches some oil deposits, but without nanobubbles in the solution to interact with, they may re-attach to the sample surface.

This data shows that in a closed system, proper removal of oil deposits cannot be achieved with water alone. In the absence of detergents or other surfactants, it was still possible to clean the surfaces using nanobubbles. Comparing the hydrophilic and the hydrophobic channels, oil removal was easier on the former, as was expected. Oil is hydrophobic and would therefore have a stronger binding affinity to a surface with like characteristics. As mentioned previously, the gas in the nanobubbles is also hydrophobic and would be expected to have a preferential binding to the oil. When simply examining the interactions, one would think that the removal of the oil deposits would be similar no matter the surface characteristics. However, in the case of the hydrophilic surface, the oil would have a weaker interaction with the surface allowing the combination of shear force and nanobubble water to have a synergistic cleaning effect.

In the hydrophobic channel, oil wets the surfaces and stays firmly on the solid surfaces, as indicated by the shape of oil droplets shown in the microscope images. A weak dewetting is expected, and the aqueous solution would be repelled from the channel surface. The increase in cleaning time, as well as the decrease in overall removal, may be explained by the difference in interactions between the hydrophobic and hydrophilic counterparts. As nanobubbles are hydrophobic their interaction with the surface, and by extension the oil deposits, is assumed

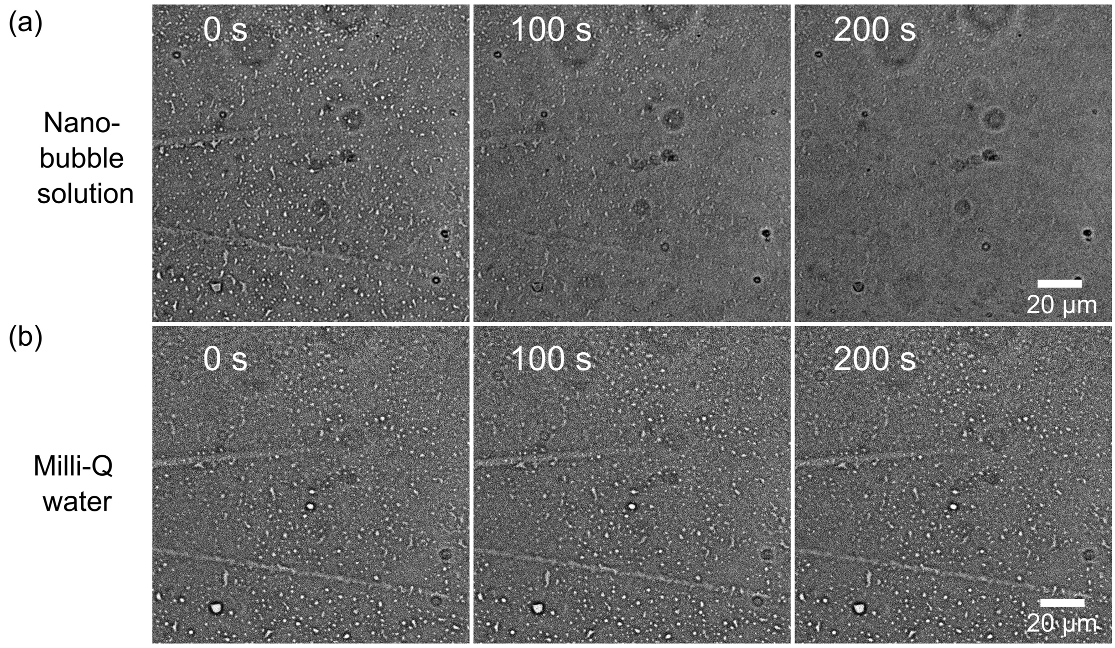


Figure 5. Removal of attached oil in a hydrophilic channel using either (a) a nanobubble solution, or (b) Milli-Q water (control). Images taken from a video, with time stamps chosen at 0 s (before introduction to the solution), 100 s, and 200 s.

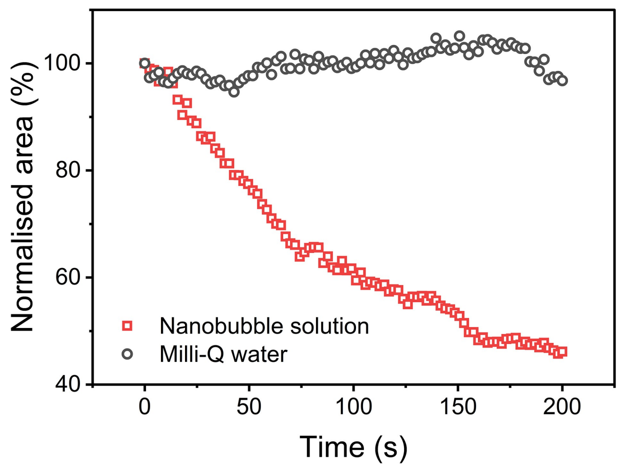


Figure 6. Percentage of attached oil on the hydrophobic surface over time. Data was normalized with respect to total area of oil at 0 s. Wash with Milli-Q water indicated by black circles and wash with the nanobubble solution indicated by red squares.

to be increased. This increased affinity then requires more work to tear the nanobubbles from the surface as they are no longer being repelled by the glass surface at the edges of the oil deposits. In addition, the aqueous solution would have a harder time getting into contact with the nanobubbles.

Dip Cleaning Under Motion

To simulate a washing environment a glass coverslip was coated with oil and placed in Milli-Q water under motion. The coverslips were visualized in the same area for comparison. Using a control solution (Figure 7b) some oil removal was observed, however, in the presence of nanobubbles (Figure 7a) the change in attached oil was more dramatic. The percentage of area covered with fluorescent oil was measured at each timepoint and normalized with respect to the initial amount at 0 min (Figure 8). We observed minor fluctuations in the control solution over the course of ten minutes; ~20% attached oil droplets were removed. In comparison there is a noticeable decrease in the fluorescence intensity over time while using nanobubble solutions. In the span of ten minutes the total area of oil deposits decreased by ~80%.

In this experiment, a larger area could be observed compared to the microchannel experiments in Figures 3 and 5 allowing better representation of the effects of the different solutions. Compared to the microchannel, we observed that the control solution was able to remove oil deposits, albeit at a much slower rate than the nanobubbles. Nile Blue dissolves well in an aqueous solution so it might be suspected that the constant motion of the solution slowly dissolved the dye leaving the oil deposits on the surface. However, as the control samples (Figure 7b) show a minimal decrease in fluorescence over the measurement time, we can conclude that the changes observed in Figure 7a are due to removal of the oil deposits and not simply bleeding of the dye into the solution.

When compared to the tests performed in the microchannels, 80% removal was obtained at around the 1 min mark which is 10 times longer than was observed for the hydrophilic channel. Effective cleaning is expected in a microchannel where the liquid flow is focused and confined to a small cross-section. In dip-cleaning under motion, however, the contaminated surfaces are not subjected to area focused cleaning as the solution can't be directed. This experiment provides a better insight into the large-scale effects of the nanobubble solution

Research Article
doi.org/10.1002/cssc.202400802

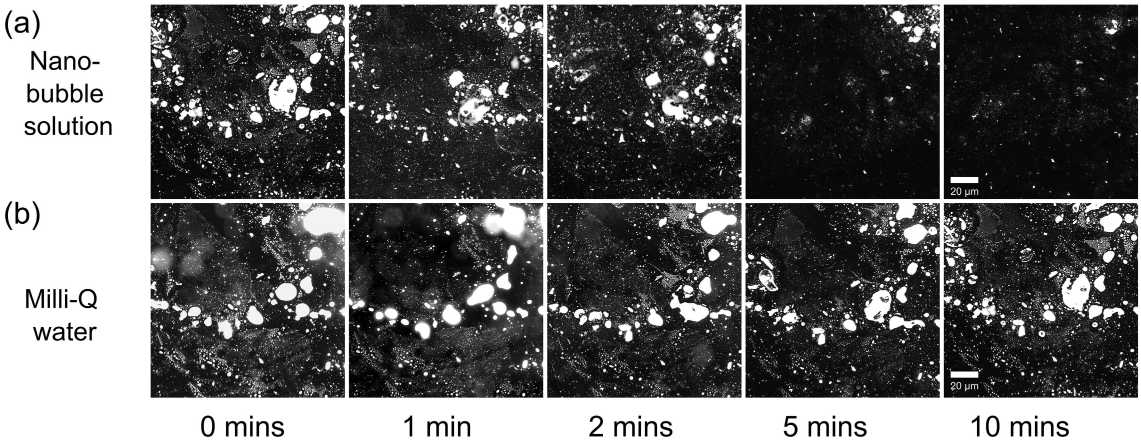


Figure 7. Nile blue dyed oil surface-stained glass coverslip under stirring in a (a) Nanobubble solution after 0 min (start), 1 min, 2 mins, 5 mins, and 10 mins, and a (b) Milli-Q solution for 0 min (start), 1 min, 2 mins, 5 mins, and 10 mins. The scale bar is 100 μm.

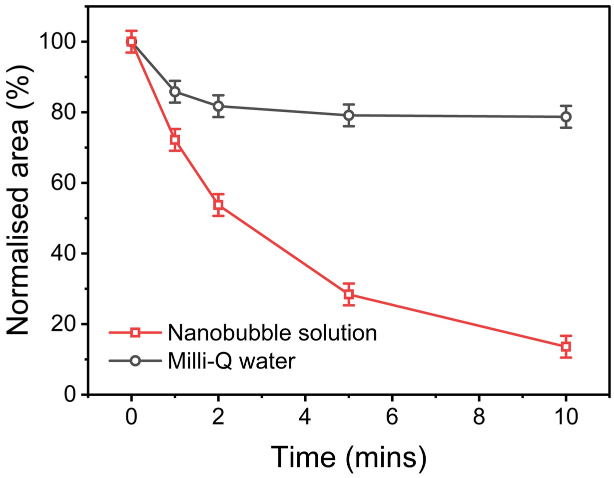


Figure 8. Percentage of oil deposits on the glass surface over time. Data was normalized with respect to total fluorescence at 0 min. Wash with Milli-Q water indicated by the black line, wash with the nanobubble solution indicated by the red line.

and shows that the effects seen on a small scale are transferable to larger scale. Dishwasher programs usually run for at least 30 min, spraying dishes with water and soap, and still do not properly remove oil residue or leftover food. Using nanobubbles we have shown an almost complete removal of oil in under 10 minutes without the need for abrasion. This could provide substantial benefits to dishwashers in the future, supplying cleaner dishes to the user without the long wait.

The interaction of oil and nanobubbles has been extensively studied with respect to oil spills. Previously, microbubbles were applied for many cleaning purposes but in most cases, nanobubbles have proven to be superior due to their large surface area to volume ratio. Nanobubbles are able to bind the biomacromolecules, resulting in light aggregates that rise to the surface of the water preventing refouling.[11] This binding of oil is driven by the interfacial energy minimization as well as electrostatic absorption.[11–12] Even though our oil deposits are attached to a surface instead of free-floating in the aqueous environment we still observed the preferential binding.

Cleaning Porcelain

To simulate the oil removal in a real-world environment oil was deposited into a porcelain bowl. The contaminated bowl was then placed vertically in the beaker. Afterwards the bowl was washed with either Milli-Q water or a nanobubble solution in a beaker under the same circumstances as stated for coverslip cleaning under motion, Figure 9. From this we observe that the nanobubble solution is superior in its ability to remove the oil,

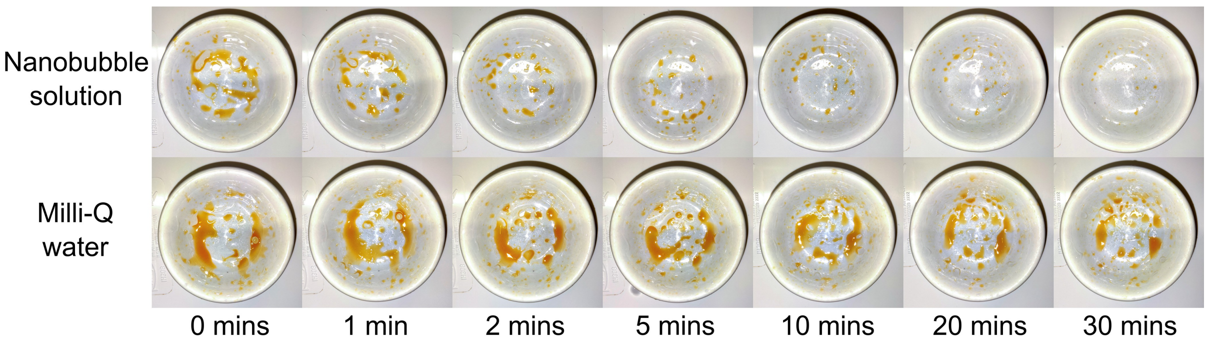


Figure 9. Cleaning oil from a porcelain bowl using a nanobubble solution (top) or Milli-Q water (bottom).

needing only 30 min for near complete removal, Figure 10. In comparison, Milli-Q water only managed partial removal at the 30 min mark, with most of the changes appearing to be from oil redistribution.

The bowl has a concave shape, and the flow of water to the surfaces is slower than to a flat surface. We therefore achieve a marginal cleaning effect compared to the glass coverslip. This can be applied to other dinning ware such as plates, and bowl-shaped or bell-shaped food containers. As discussed above, nanobubbles are better suited in the removal of tough materials compared to normal water, achieving almost full removal in under an hour. Applying nanobubble water in dishwashers would therefore not only reduce the time it takes to run a cleaning cycle, but would also drastically reduce the amount of water needed. Additionally, the amount of required soap would also be reduced, now only requiring what is needed to kill bacteria. Put together, this makes nanobubbles very environmentally friendly.

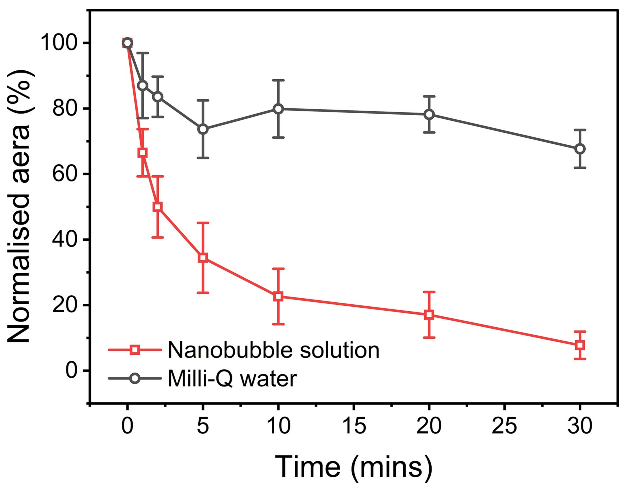


Figure 10. Percentage of remaining oil over time. Data normalized with respect to total area of oil at 0 min. The black line represents oil removal using Milli-Q water, and the red line represents oil removal using a nanobubble solution.

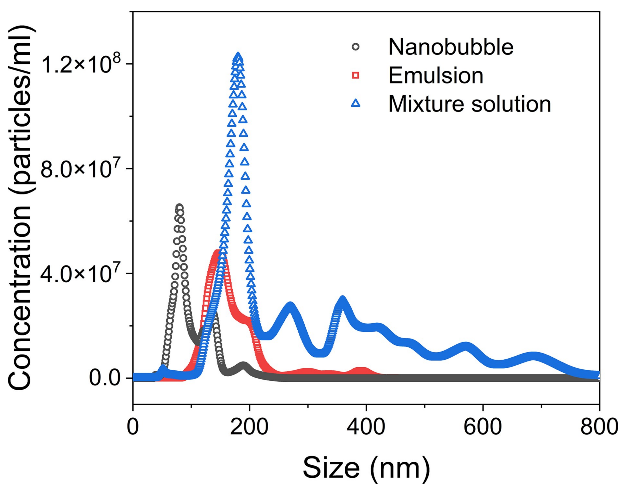


Figure 11. Size and concentration distribution measured by NTA of the oil emulsion in Milli-Q water (black circles), the nanobubble solution (red squares), and the oil emulsion/nanobubble mix (blue triangles).

Oil Nanodroplet and Nanobubble Interactions

When washing with nanobubbles, it is hypothesized that the nanobubbles interact with and bind to the oil. In this way the nanobubbles help the detachment of the oil from the surface and prevent re-fouling. To show this interaction, we performed initial experiments by mixing an oil emulsion with a solution containing nanobubbles. The mixture was allowed to rest for 30 min with occasional gentle mixing. The mean sizes of our three solutions were measured using NTA built-in software, and were as follows: 104.0 $\pm$ 2.7 nm for the nanobubble solution, 176.7 $\pm$ 3.8 nm for the oil emulsion, and 332.2 $\pm$ 7.7 nm for the mixed solution. The size distribution and particle concentration can be seen in Figure 11.

The near doubling in mean size after mixing indicates some manner of interaction between the two solutions. In a previous study where we characterized the stability of oil emulsions, we saw that there was no remarkable change in the mean sizes of the oil emulsions over time.[13] In combination with the knowledge that nanobubbles are incredibly stable, this heavily suggests that the increase in mean size is not due to Oswald ripening of either the nanobubbles or the oil emulsion. Rather, we can say that the change is due to oil droplets interacting with the nanobubbles.

In addition, Figure 11 shows us that there is not one single particle size; they vary up to about 800 nm indicating that there are larger aggregates of oil/nanobubbles. The drastic increase in size between the original solutions and the mixture using NTA is mainly due to the dynamic changes in the sample. Mixing bubble water and oil emulsion is a complex process, and there are several things that can happen.

In a study by Rico et al.[14] they observed the effects of bringing attached droplets and bubbles in proximity to each other, and subsequently separating them. As the distance between the bubble and the oil droplet decreases, the initial interaction will be repulsive, and deformation of the two will occur. However, as the interfacial tensions aren't the same their deformations won't be equal. This repulsive force continued until they started to retract the bubble, in which case they observed a suction effect where the film between the bubble and the oil started to thin. This thinning creates an attraction and may lead to coalescence if allowed to continue.

In a different study by Yan et al.[15] a microbubble was allowed to finish its interaction with an oil droplet, which resulted in the film between the two rupturing, and the oil droplet completely covered the bubble. The drainage and rupture of the film were found to be driven mainly by the Laplace pressure.

In the first study, the oil and gas were not allowed to fully interact, whereas in the second study, it was shown that an interaction was possible to form a co-shell particle. A combination of the two may help explain the phenomena observed in this paper. When mixing the oil emulsion and the bubble solution, gentle mixing would force the two entities to oscillate near each other. Upon collision, the oil droplets from the emulsion may coat the gas bubble, thereby increasing the size of the particle. Continuous interaction with oil droplets may thicken the oil shell, potentially explaining the broad spectrum of sizes we observed with NTA (Figure 11).

In the case of our oil removal discussed in previous sections, it is hypothesized that the nanobubbles interact with the oil deposits due to their shared hydrophobic properties. Upon interaction, the oil may adhere and coat the bubble, thereby reducing the Laplace pressure. In a different case, the nanobubble may interact, but not completely bind to the oil due to the repulsive forces discussed by Rico et al.[14] But, as the flow forces the nanobubble along the channel, the attractive force would take hold and the oil would be pulled away from the surface. Once in solution as oil droplets, they can then bump into nanobubbles, generating this co-shell particle which would help prevent re-fouling. Complete removal of surface-attached nanobubbles can be achieved via bulk oversaturation.[16] It would therefore be interesting in future experiments to test the effect of different nanobubble concentrations to observe if oversaturation would increase oil removal, and by extension, decrease the required cleaning time.

Conclusions

In conclusion, our study successfully demonstrated the effective removal of oil deposits from both hydrophilic and hydrophobic surfaces using a solution containing nanobubbles generated with electrolysis. When compared to a control solution consisting solely of Milli-Q water, our nanobubbles exhibited superior performance in dislodging oil attached to glass and porcelain bowls. This phenomenon was consistently observed in both microchannel experiments and on a larger scale. Furthermore, our preliminary data suggest potential interactions between oil emulsions and bulk nanobubbles, warranting further investigation.

Notably, all experiments were conducted at room temperature. As highlighted in the introduction, traditional detergents are primarily effective on liquid oils or lipids and may not be ideal for materials requiring washing at or near room temperature. Moreover, materials such as wool are sensitive to friction during washing, necessitating the use of specialized detergents. The use of nanobubbles offers a promising solution, as they can effectively remove oils at temperatures below their melting point without requiring abrasive action. This suggests that nanobubbles have the potential to simplify the cleaning process for delicate materials, offering a gentler and more efficient alternative to traditional cleaning methods.

Acknowledgements

H.A. acknowledges the support from the Australian Research Council Future Fellowship (FT180100361) and Discovery Project (DP230100556). B.H.T. is supported by the Excellent Young Scientists (Overseas) fund of the National Natural Science Foundation of China. H.A. also thanks proof reading and editing by Grace An.

Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

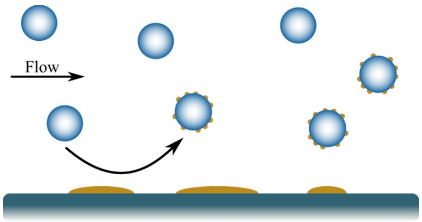
Keywords: Bulk nanobubbles · Cleaning · Oil contaminants · Microfluidic channels

[1] S. A. Mousavi, F. Khodadoost, *Environ. Sci. Pollut. Res. Int.* **2019**, *26*, 26439–26448.
[2] a) J. Zhu, H. An, M. Alheshibri, L. Liu, P. M. Terpstra, G. Liu, V. S. Craig, *Langmuir* **2016**, *32*, 11203–11211; b) N. Jin, F. Zhang, Y. Cui, L. Sun, H. Gao, Z. Pu, W. Yang, *Particuology* **2022**, *66*, 1–9.
[3] Z. Wu, H. Chen, Y. Dong, H. Mao, J. Sun, S. Chen, V. S. Craig, J. Hu, *J. Colloid Interface Sci.* **2008**, *328*, 10–14.
[4] a) H. H. Hansen, H. Cha, L. Ouyang, J. Zhang, B. Jin, H. Stratton, N.-T. Nguyen, H. An, *Biotechnol. Adv.* **2023**, *63*, 108091; b) B. H. Tan, H. An, C.-D. Ohl, *Curr. Opin. Colloid Interface Sci.* **2021**, *53*, 101428; c) B. H. Tan, H. An, C.-D. Ohl, *Phys. Rev. Lett.* **2020**, *124*, 134503.
[5] J. Jin, R. Wang, J. Tang, L. Yang, Z. Feng, C. Xu, F. Yang, N. Gu, *Colloids Surf. A* **2020**, *589*, 124430.
[6] a) Y. Qiu, H. Ren, M. A. Edwards, R. Gao, K. Barman, H. S. White, *Langmuir* **2020**, *36*, 6073–6078; b) E. P. Favvas, G. Z. Kyzas, E. K. Efthimiadou, A. C. Mitropoulos, *Curr. Opin. Colloid Interface Sci.* **2021**, *54*, 101455; c) A. J. Jadhav, M. Barigou, *Ind. Eng. Chem. Res.* **2021**, *60*, 17999–18006.
[7] R. Mercadé-Prieto, S. Bakalis, *Text. Res. J.* **2014**, *Text. Res. J.*, 52–65.
[8] L. Landeck, L. A. Baden, S. M. John, *Kanerva's Occupational Dermatology* **2020**, 1131–1143.
[9] B. H. Tan, H. An, C.-D. Ohl, *Phys. Rev. Lett.* **2018**, *120*, 164502.
[10] a) H. Cha, H. Fallahi, Y. Dai, S. Yadav, S. Hettiarachchi, A. McNamee, H. An, N. Xiang, N.-T. Nguyen, J. Zhang, *Lab Chip* **2022**, *22*, 2789–2800; b) G. M. Whitesides, E. Ostuni, S. Takayama, X. Jiang, D. E. Ingber, *Annu. Rev. Biomed. Eng.* **2001**, *3*, 335–373.
[11] Z.-H. Zhang, S. Wang, L. Cheng, H. Ma, X. Gao, C. S. Brennan, J.-K. Yan, *Food Rev. Int.* **2023**, *39*, 4213–4235.
[12] K. Zhou, V. Maugard, W. Zhang, J. Zhou, X. Zhang, *Nanomaterials* **2023**, *13*, 1496.
[13] H. H. Hansen, G. Kijanka, L. Ouyang, N.-T. Nguyen, H. An, *J. Mol. Liq.* **2024**, 124323.
[14] R. F. Tabor, C. Wu, H. Lockie, R. Manica, D. Y. Chan, F. Grieser, R. R. Dagastine, *Soft Matter* **2011**, *7*, 8977–8983.
[15] S. Yan, X. Yang, Z. Bai, X. Xu, H. Wang, *Chem. Eng. Sci.* **2020**, *223*, 115740.
[16] L. Ouyang, Q. Zeng, N.-T. Nguyen, B. H. Tan, H. An, *Colloids Surf. A* **2024**, *689*, 133665.

Manuscript received: April 15, 2024
Revised manuscript received: May 19, 2024
Version of record online: ■■, ■■

RESEARCH ARTICLE

Nanobubbles in clean water effectively clean oil from surfaces like glass and dishes without using detergents. This eco-friendly method shows great promise in dishwashers and washing machines, providing a low-cost, sustainable cleaning solution.



H. H. W. B. Hansen, L. Ouyang, H. Cha, J. Zhang, Q. Li, B. H. Tan, A. Vashi, N.-T. Nguyen, H. An**

1 – 9

Surface Cleaning of Oil Contaminants Using Bulk Nanobubbles