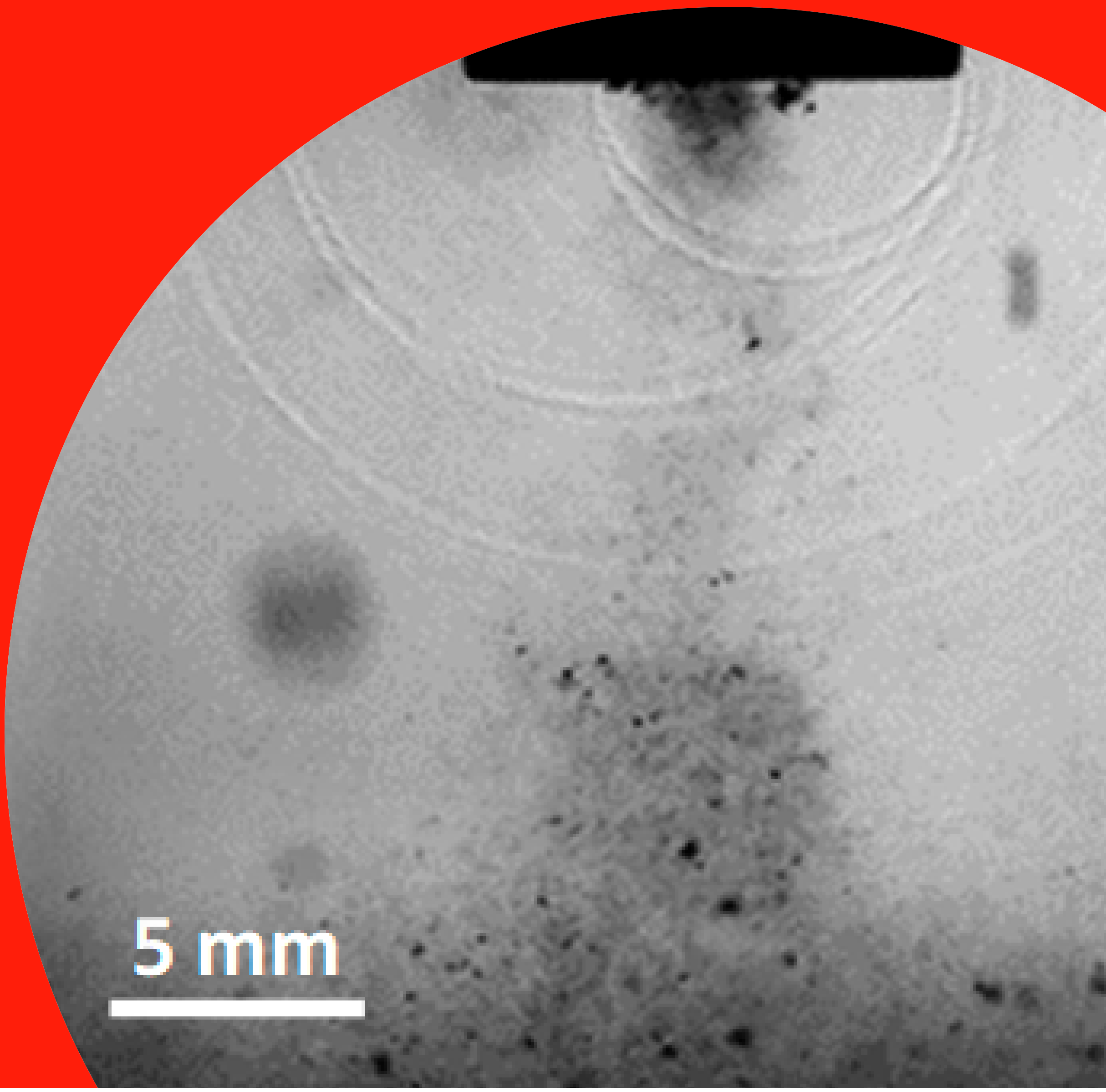
IOP Physical Acoustics Group Tutorial Day:
Acoustics in Action – Sonochemistry

22 September 2023
Institute of Physics, London, UK



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Programme

9:30am - 9:50am	**Registration and Refreshments**
9:50am - 10:00am	**Welcome**
10:00am - 11:45am	**Invited Speaker Presentations** **10:00am - 10:35am Paul Prentice** Shocking bubbles – on the importance of periodic bubble collapse shock waves in measuring and monitoring acoustic cavitation **10:35am - 11:10am Madeleine Bussemaker** Bubble bubble toil and trouble… **11:10am - 11:45am Iakovos Tzanakis** Ultrasonic Processing of Materials: Fundamentals and Application
11:45am - 12:45pm	**Lunch and Posters**
12:45pm - 1:20pm	**Invited Speaker Presentation** **12:45pm - 1:20pm James Kwan** Emerging sonochemical reactor designs and chemistries
1:20pm - 2:00pm	**Flash Presentations** **1:20pm - 1:30pm Irem Soyler** Optimising ultrasonic parameters for bacterial inactivation **1:30pm - 1:40pm Lillian Usadi** Inertial and Stable Cavitation Noise Characterization of Tetracycline Degradation in a Sonochemical Reactor Utilizing Machine-Learning Techniques **1:40pm - 1:50pm Dong Xia** SonoElectrochemical CO2 Reduction over Cu Catalysts **1:50pm - 2:00pm Cherie CY Wong** Sonocatalytic Conversion of Furfural into High-value Chemicals with Heterogeneous Nanostructured TiO2 Nanocups
2:00pm - 3:30pm	**Posters and Networking**
3:30pm - 3:35pm	**Close of Meeting**

Invited Speakers

Shocking bubbles – on the importance of periodic bubble collapse shock waves in measuring and monitoring acoustic cavitation

Paul Prentice[1]
[1]University of Glasgow, United Kingdom

Invited Speaker Presentations, September 22, 2023, 10:00 - 11:45

In this presentation, I will review research from my lab demonstrating the role of periodic shock waves in measuring and monitoring cavitation activity, for a range of medically and industrially relevant systems/configurations. I will show that periodic shock waves are responsible for all non-linear emissions, including harmonic, subharmonic and broadband signals. Moreover, simple measurements of the shock wave content in the time-domain, can be used to identify the most effective pressure amplitudes, for efficient sono-processing. This will be demonstrated for a range of industrially relevant liquids and solvents.

Bubble bubble toil and trouble…

Madeleine Bussemaker[1]
[1]University of Surrey, United Kingdom

Invited Speaker Presentations, September 22, 2023, 10:00 - 11:45

Sonochemistry relies on cavitation bubbles created through the application of an ultrasonic wave (20 kHz – 2 MHz) to a fluid medium. The micro-sized bubbles oscillate and collapse, creating unique high temperatures and pressures leading to plasma chemistry that have fascinating consequences not seen in other chemical environments. To enable ultrasound application in industry the impacts of flow regimes (overhead stirring, continuous reactors with flow-through) and reactor configuration (horn and plate transducers, applied frequency and power, wave reflection) on the sono-process of interest are researched.

We find that different chemical systems and measurement techniques (dosimetry, sonoluminescence, sonochemiluminescence, pollutant degradation) have inconsistent responses to reactor configuration changes. This is attributed to disparities in energy requirements, reaction locations, and reaction mechanisms. Flow is suggested to impact sonochemical activity through: bubble transience (deviation from sphericity) and collapse shape, which in cases may enable nanodroplet injection or enhanced bubble fragmentation; bubble clustering and subsequent coalescence; wave propagation and hence standing and travelling wave components; and aeration of solution and available nuclei. Results will be discussed in reference to applications such as degradation of per-and poly-fluoroalkyl substances. Variations in results and literature highlight the challenges of generalisation of parametric effects in the chaotic system.

Ultrasonic Processing of Materials: Fundamentals and Applications

Iakovos Tzanakis[1]
[1]Oxford Brookes University, United Kingdom

Invited Speaker Presentations, September 22, 2023, 10:00 - 11:45

Ultra-Sonic Processing (USP) has gained considerable momentum in the last decade due to its versatility as a sustainable, environmentally friendly, and cost-effective technique. By utilizing high-frequency sound waves, USP forms and collapses bubbles within a liquid medium. This phenomenon, called cavitation, generates several mechanical effects, including high-speed liquid jets (more than 100 m/sec), powerful shockwaves (up to 10 MPa) as well as acoustic streaming (up to 1 m/sec) which can be utilised in various technological applications.

Despite its wide range of uses, the practical implementation of USP has largely relied on empirical knowledge rather than a fundamental understanding of the underlying mechanisms. To fully leverage the potential of USP in the synthesis and production of materials, it is essential to gain a deeper insight into the mechanisms driving cavitation.

In this presentation, we aim to provide a comprehensive overview of recent studies that focus on the analysis, optimization, and control of USP for specific applications. We delve into three key areas: grain refinement of aluminum alloys, exfoliation of 2D nanomaterials, and processing of composites.

To investigate the dynamics of cavitation, advanced experimental techniques are employed. High-speed cameras allow us to visualize the formation, expansion, and collapse of cavitation bubbles in real-time. In-situ synchrotron imaging, enhances the liquid/air interfaces, providing valuable insights into the spatial distribution, characteristics and behavior of bubbles under different process conditions. Furthermore, state-of-the-art calibrated cavitometers and hydrophones are utilized to capture and analyze the acoustic waves and shockwave emissions associated with cavitation.

The results of these studies demonstrate the effectiveness of optimized USP conditions in enhancing grain refinement in aluminum alloys. For instance, in direct-chill (DC) casting processes, cavitation-induced shockwaves play a significant role in fragmenting intermetallic crystals and dendrites, leading to refined grain structures and improved mechanical properties.

Moreover, the exfoliation of 2D nanomaterials like graphene is driven primarily by the energy imparted by shockwaves during cavitation events. By understanding and controlling the cavitation parameters, it becomes possible to achieve precise and efficient exfoliation, enabling the production of high-quality graphene with unique properties.

In addition to grain refinement and exfoliation, USP also offers advantages in processing composites. The cavitation activity enhances the dispersion of reinforcing fibers within highly viscous polymers, resulting in improved matrix stability and strength. This aspect has significant implications for the development of advanced composite materials with enhanced mechanical properties and tailored functionalities.

Overall, the combination of advanced experimental techniques and modelling allows us to gain valuable insights into the fundamental mechanisms driving USP. By understanding and optimizing these processes, we can unlock the full potential of USP for a wide range of applications, leading to significant advancements in materials science and engineering.

Emerging sonochemical reactor designs and chemistries

James Kwan[1]
[1]University of Oxford, United Kingdom

Invited Speaker Presentation, September 22, 2023, 12:45 - 13:20

Sonochemistry is a green method for performing unique chemistries under ambient conditions. Currently, much of sonochemistry involves off-the-shelf sonochemical reactor designs. Here we will discuss a novel sonochemical reactor design that reflects a cylindrically spreading acoustic wave 180°. The reflected wave converges to create a "hot zone" positioned along the axis of the reaction vessel. Using a chemical probe, we determined the SE of our sonochemical reactor at various operation conditions and compared it to other sonochemical reactors. We will also show that by including certain catalysts promotes the conversion of carbon dioxide to hydrocarbons such as methane and ethane. Indeed, both the design of the reactor and the catalyst may be new avenues to explore for sonochemistry.

Flash Presentations

Inertial and Stable Cavitation Noise Characterization of Tetracycline Degradation in a Sonochemical Reactor Utilizing Machine-Learning Techniques

Lillian Usadi[1]
[1]University Of Oxford, United Kingdom

Flash Presentations, September 22, 2023, 13:20 - 14:00

The ability to detect, characterize, and predict inertial and stable cavitation has tremendous promise in the fields of biomedicine and sonochemistry. Two notable application fields are biomedicine and chemistry in which the ability to control cavitation can be used to enhance the breakdown of kidney stones and facilitate chemical reactions critical for hydrogen production and wastewater treatment. Typically, spectral features of cavitation noise signals obtained through Fourier analysis are analyzed resulting in a qualitative categorization of the signal. This presentation will utilize machine-learning techniques in order to correlate cavitation noise with the degradation of tetracycline, a common antibiotic in wastewater, using a bespoke sonochemical reactor capable of noise monitoring and cavitation inception. Further research will be done in optimizing acoustic parameters such as pulse repetition frequency (PRF), pulse length (PL), and duty cycle (DC) in order to achieve optimal efficiency for other sonochemical reactions.

Optimising ultrasonic parameters for bacterial inactivation

Ms Irem Soyler[1], Dr Katie Costello, Dr Jorge Gutierrez-Merino, Dr Madeleine Bussemaker
[1]University of Surrey, Guildford, United Kingdom

Flash Presentations, September 22, 2023, 13:20 - 14:00

Ultrasound is a non-thermal technology with diverse applications in the food industry. Recently, its antimicrobial efficiency has gained attention, especially for providing microbiological safety for heat-sensitive food products. However, the antimicrobial mechanism of ultrasound remains unclear.

Characterized by the formation and implosion of bubbles, ultrasound-induced cavitation facilitates various chemical reactions. Sonochemistry can be considered one such mechanism, as it involves the impact of free radicals and reactive oxygen species on bacterial cells. Previous studies have demonstrated the significant impact of varying conditions on the sonochemical efficiency of ultrasound, with the highest activity observed within the frequency range of 300–500 kHz. However, comprehensive comparisons of experimental conditions across different frequencies and power levels have been lacking. Notably, active bubble distribution in carrier liquids varies at different frequencies, necessitating a careful assessment of experimental conditions to optimise ultrasonic impact.

In this work, SL, SCL and dosimetry were used to elucidate the optimal position of the sample vial for subsequent treatment of bacteria. The results will underpin a mechanistic understanding of ultrasound treatment of bacteria with respect to various bubble dynamics.

Sonocatalytic Conversion of Furfural into High-value Chemicals with Heterogeneous Nanostructured TiO2 Nanocups

Cherie CY Wong[1], Professor James Kwan[1]
[1]University of Oxford, Oxford, United Kingdom

Flash Presentations, September 22, 2023, 13:20 - 14:00

Furfural is a promising biomass platform chemical and has recently drawn great attention. Its highly functionalised molecular structure allows diverse reaction pathways, yielding an array of value-added products. However, prevailing furfural conversion techniques suffer from poor selectivity and conversion rates, leading to extensive energy consumption for product purification in the downstream processes. Here, we introduce a novel approach employing a sonochemical reactor that concentrates acoustic waves to activate inertial cavitation-mediated oxidation of furfural for the first time. We believe that the hydroxyl radicals that generated from cavitation trigger a ring-opening reaction, subsequently forming various chemicals. Through the incorporation of heterogeneous nanostructured TiO_2 nanocups, the conversion rate of furfural is significantly increased. This is attributed to the preferential generation of cavitation bubbles on the TiO_2 surface via heterogeneous nucleation. Furthermore, as these bubbles nucleate and collapse near the TiO_2 surface, the formation of radicals is in proximity with furfural molecules. Remarkably, besides its role as a cavitation agent, TiO_2 might function as a catalyst, converting furfural along specific chemical pathways; this acts as a starting point for selective oxidation. We hope the innovative application of sonochemistry in furfural conversion will facilitate the development of more economical approaches for biomass valorisation.

SonoElectrochemical CO2 Reduction over Cu Catalysts

Dr Dong Xia[1], Miss Yi Qin, Dr Yagya Regmi, Professor Laurie King, Professor James Kwan
[1]University of Oxford, Oxford, United Kingdom, [2]Manchester Metropolitan University, Manchester, United Kingdom

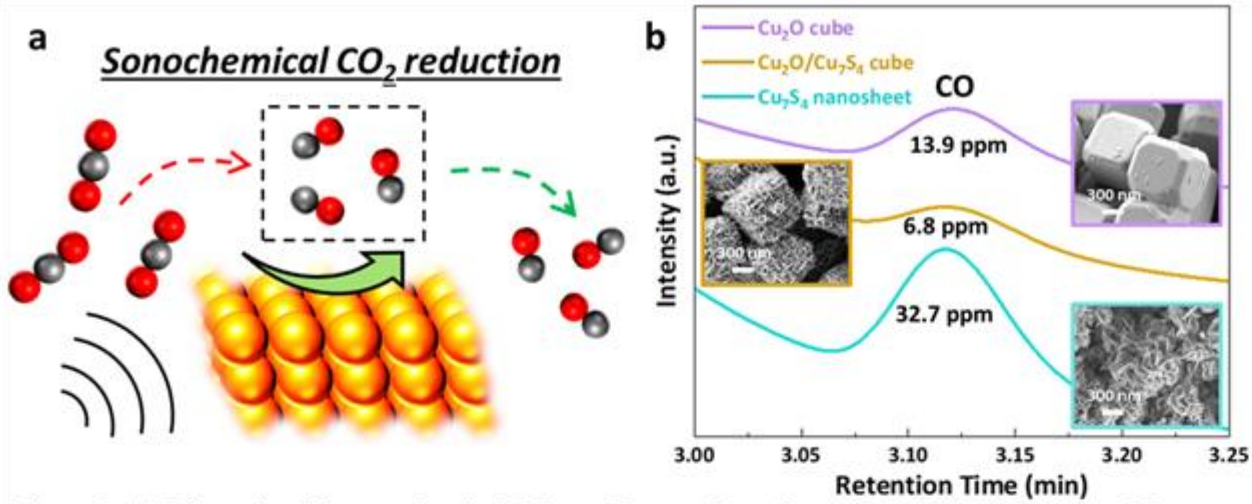
Flash Presentations, September 22, 2023, 13:20 - 14:00

Sonochemisty contributes considerably to enhancing chemical processes in the field of energy storage and conversion.1,2 Currently, sonochemisty is rarely touted as a highly-plausible methodology for converting CO2 to value-added chemicals/fuels. To verify plausibility, Cu-based catalysts (e.g. Cu2O cube, Cu2O/Cu7S4 cube and Cu7S4 nanosheet) with various morphologies are synthesized for sonochemical CO2 reduction (CO2R), results show that all the catalysts exhibit strong cavitation phenomena and remarkable CO2-to-CO conversion capability (Figure 1). Specifically, Cu7S4 nanosheet is the most optimal with a maximum CO production.

Figure 1. (a) Schematics of the sonochemical CO2-to-CO over Cu surface. (b) Sonochemical CO2-to-CO performances of as-synthesized catalysts.

The objectives of using Cu-based catalysts are their electrochemical selectivity towards multicarbon (C2+) are unsatisfactory.3 While inertial activation benefits yielding key *CO intermediates (as evidenced in Figure 1b) which are essential for *CO dimerization (or C-C coupling, Figure 2b) to form multicarbons during the electrochemical CO2R process.4 Therefore, we purpose using the sonoelectrochemical CO2R method (Figure 2a) to boost C2+ formation aiming to expedite the process of carbon neutrality.

Figure 2. (a) Schematic of a sonoelectrochemical reactor for CO2-to-C2+ with high faradaic efficiency (FE). (b) Mechanistic schematic of the sonoelectrochemical CO2-to-C2+ conversion over Cu surface accompanying with cavitation-boosted C-C coupling.



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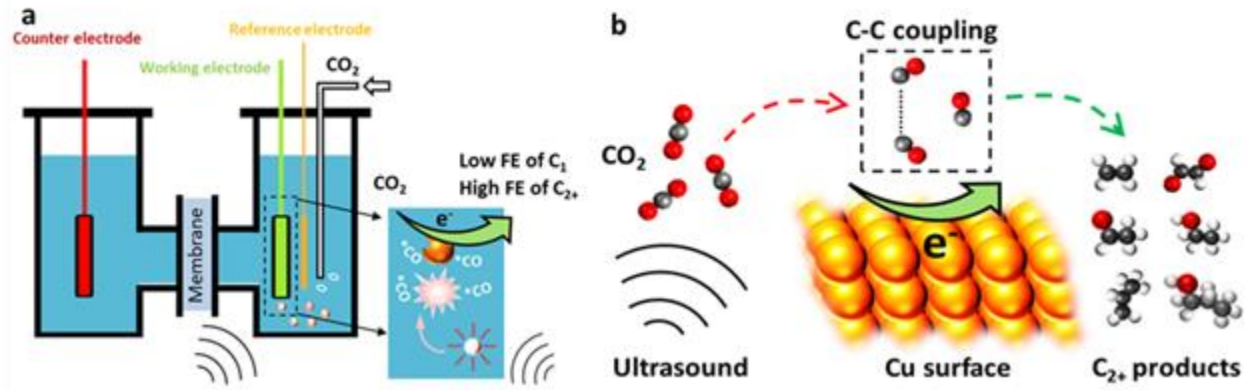

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Poster Presentations List

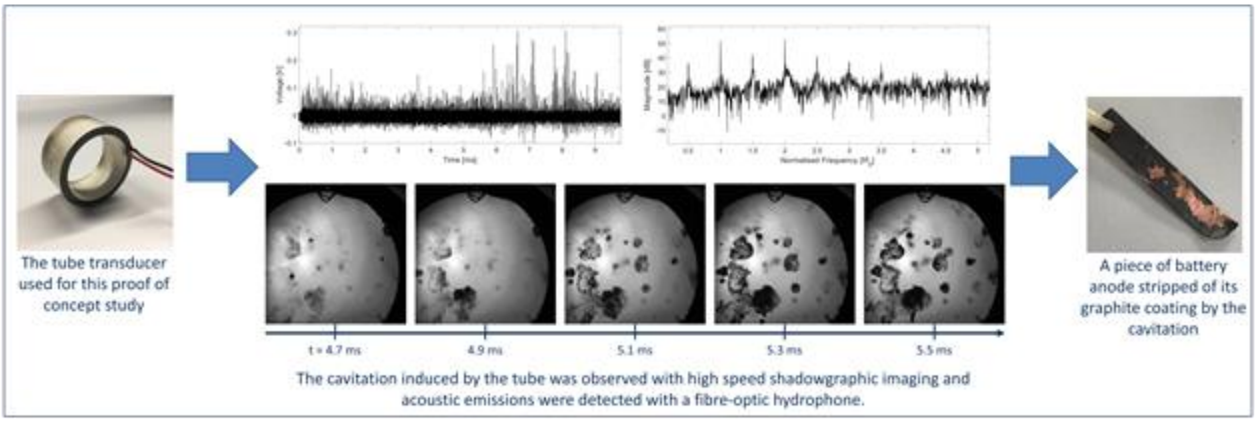
Paul Prentice	Tube transducer configuration for flow-based ultrasonically enhanced technology critical metal recovery from electronic waste
Paul Prentice	3.4 MHz resonance in fibre-optic hydrophone detection of cavitation shockwaves
Yi Qin	Catalytic Sonochemistry of Cu2O Particles for CO2-to-CO Conversion
Irem Soyler	Optimising ultrasonic parameters for bacterial inactivation
Lillian Usadi	Inertial and Stable Cavitation Noise Characterization of Tetracycline Degradation in a Sonochemical Reactor Utilizing Machine-Learning Techniques
Muhammad Auwal Yunusa	Per- and Poly-fluoroalkyl Substances (PFAS) Remediation Using Cold Atmospheric Plasma and Ultrasound
Yucheng Zhu	Implementation of sonoluminescence and sonochemiluminescence -based select of ultrasonic chemical reaction parameters

Poster Presentations

Tube transducer configuration for flow-based ultrasonically enhanced technology critical metal recovery from electronic waste

Hilde Metzger[1], Paul Daly[1], Andrew Feeney[1], <u>Paul Prentice</u>[1]
[1]University of Glasgow, Glasgow, United Kingdom

With the growing demand for technology critical metals (TCMs), as a finite resource on which the UK is 100% import reliant, it is crucial to develop sustainable approaches for recycling from electronic waste (E-waste), contributing to a circular economy.
TCMs have been delaminated from complex E-waste structures (e.g. printed circuit boards and aged photovoltaics) by immersion in deep eutectic solvents (DESs); catalytic etchants that are economical and more environmentally friendly than traditionally used mineral acids. The application of power ultrasound is postulated to significantly accelerate TCM delamination efficiency.
This work presents initial work on the design of a flow-based sonoprocessing system. Crushed E-waste suspended in DES, flows through a pipe with embedded piezoelectric tube transducers, generating cavitation within the stream. The TCMs are then delaminated for recovery, downstream.
Transducer fabrication was informed by finite element modelling software package COMSOL Multiphysics at all stages, and good agreement with electrical impedance measurements found. High-speed shadowgraphic imaging with simultaneous acoustical detection was used to characterise the cavitation produced across a range of input powers, to determine the optimal parameters for efficient sonoprocessing, for upscaling to industrially relevant levels.

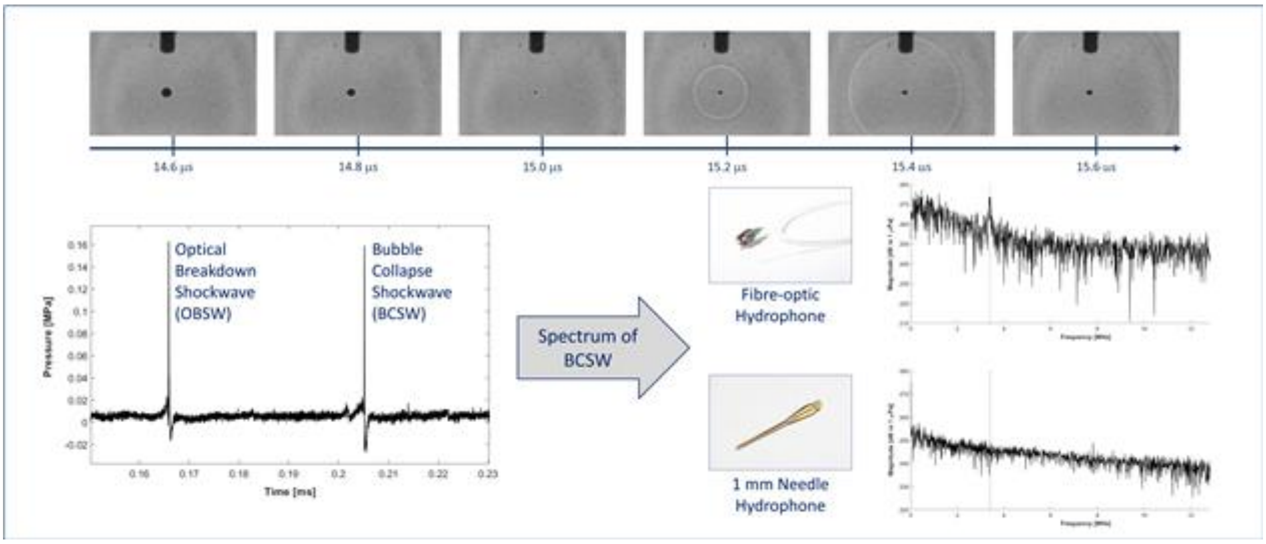


The cavitation induced by the tube was observed with high speed shadowgraphic imaging and acoustic emissions were detected with a fibre-optic hydrophone.

3.4 MHz resonance in fibre-optic hydrophone detection of cavitation shockwaves

Hilde Metzger[1], Paul Morris[2], Andrew Hurrell[2], <u>Paul Prentice</u>[1]
[1]University of Glasgow, Glasgow, United Kingdom, [2]Precision Acoustics Ltd, Dorchester, United Kingdom

The role of periodic bubble collapse shockwaves (BCSWs) in the nonlinear emissions generated by acoustic cavitation [1], and the requirement to use hydrophones calibrated over a sufficiently large bandwidth for quantitative measurements, are generally well accepted. Existing techniques for calibration, however, do not always involve impulsive pressure transients, which therefore may not fully characterise a hydrophone for the purpose of cavitation detection.
In this work, BCSWs generated by a pulsed laser-induced cavity and a 20 kHz sonotrode, are simultaneously detected with a commercially available fibre-optic (FOH) and a needle hydrophone (NH), both with broadband calibration. The results suggest that the FOH has an impulse response that includes a resonance at circa 3.4 MHz, which is not detected by the NH, or the FOH calibration. We believe that this feature has been recently misinterpreted as inherent to the power spectra of BCSWs generated during sonotrode application [2].
Although the source of the FOH resonance remains the subject of an ongoing investigation, we have determined that the prominence of the feature varies probe-to-probe, which may implicate structural variations during manufacture. The findings also suggest that characterising the impulse response of a detector is necessary for accurately detecting and monitoring acoustic cavitation.



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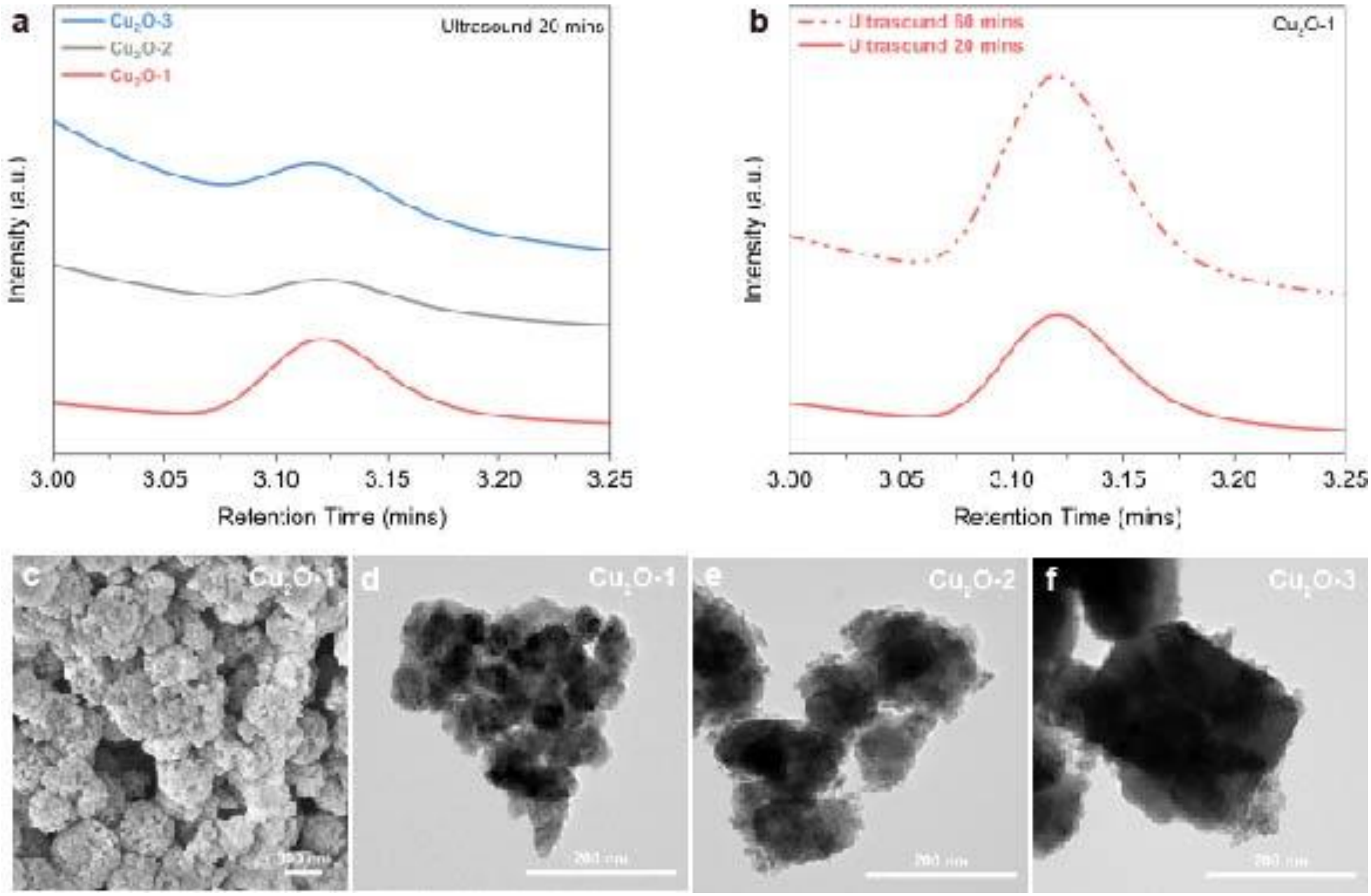
Catalytic Sonochemistry of Cu2O Particles for CO2-to-CO Conversion.

Yi Qin[1], Dong Xia[1], Yagya Regmi[2], Laurie King[2], James Kwan[1]

[1]University of Oxford, Oxford, United Kingdom, [2]Manchester Metropolitan University, Manchester, United Kingdom

The sustainable conversion of abundant CO2 into value-added chemicals and fuels is a critical challenge. Among various techniques for CO2 reduction reaction (CO2RR), sonocatalysis, which employs ultrasound as a stimulus, offers advantages like operational safety and ambient condition suitability. Here, we introduce Cu2O materials as bifunctional agents that act both as cavitation nuclei and catalysts, thereby enhancing CO2 activation. We demonstrate that aggregated Cu2O particles, featuring multiple gas pockets, facilitate a strong cavitation effect that triggers efficient CO2-to-CO conversion. Notably, a size effect is observed: 20 nm Cu2O particles exhibit optimal CO2RR performance. This study provides a facile sonochemical approach to address CO2 challenges and holds promise for advancing future sonoelectrocatalysis applications.



Implementation of sonoluminescence and sonochemiluminescence -based select of ultrasonic chemical reaction parameters

Miss Yucheng Zhu, Mr. Xueliang Zhu, Prof. Xuhai Pan, Dr. Lianxiang Liu, Dr, Madeleine Bussemaker
[1]University of Surrey, Guildford, United Kingdom, [2]Nanjing Tech University, Nanjing, China

When a liquid is irradiated with ultrasound, acoustic cavitation (AC) is produced that results in sonochemical processes. The ultrasonic-driven bubbles, go through bubble generation, growth, and collapse processes which can effectively accelerate chemical processes. However, directly monitoring the AC location and intensity in a reactor is difficult. The accompanying light emission from the formed bubbles allows AC to be visualised. The light can be formed directly (SL: sonoluminescence) and indirectly (SCL: sonochemiluminescence). This work aims to explore the bubble behaviours in the SL and SCL regimes under different bubble stability-related parameters, such as ultrasound frequency, reactor configuration, and solvent properties. The spatial distribution of bubbles in the sonic field was characterised by an ANDOR iXon3 EMCCD camera. A MATLAB programme was written and applied to analyse the bubble behaviours in the captured images. Results show three typical frequency ranges for bubble behaviours: $f<200$ kHz, $200 \text{ kHz}<f<1000 \text{ kHz}$, and $1000 \text{ kHz}<f<2000 \text{ kHz}$. The effect of pressure amplitude is more pronounced when $f>200$ kHz. Furthermore, dimensionless and multiple regression analyses were applied to further quantify AC. An empirical correlation between the bubble behaviours and key driving factors was developed. These results can provide significant guidance in designing chemical processes and reactors.

Per- and Poly-fluoroalkyl Substances (PFAS) Remediation Using Cold Atmospheric Plasma and Ultrasound

<u>Mr Muhammad Auwal Yunusa</u>[1], Dr Madeleine Bussemaker[1], Dr Patrick Sears[1]
[1]University of Surrey, Guildford, United Kingdom

Per- and poly-fluoroalkyl substances (PFAS) are a set of synthetic chemicals containing carbon-fluorine bonds, one of the strongest bonds in organic chemistry. The widespread presence of PFAS in human and animal blood, drinking water, and the environment has raised concerns due to their potential for adverse health impacts.
This study explores the synergistic effect of ultrasound and plasma-activated water (PAW) to address the persistence of PFAS pollutants. Ultrasound induces degradation through the collapse of cavitating bubbles, generating plasma conditions, radical species and extreme temperatures and pressures, leading to the breakdown of PFAS compounds. Cold plasma, characterized by reactive oxygen and nitrogen species production, is used to obtain PAW.
High-frequency ultrasound (HFUS) has shown notable efficacy for PFAS treatment. To address the limitations of current treatment methods, this work proposes a novel approach; the integration of HFUS and PAW. Cold plasma gas (generated using air at 5 L/min) is bubbled for 30 min into PFAS-contaminated water. The treatment water is then sonicated at 400 kHz for 30 min. PFAS mineralization is measured via the percentage of defluorination. This combined approach aims to achieve enhanced degradation efficiency, toward PFAS remediation.

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