Acoustic Resonance and Atomization for Gas-Liquid Systems in Microreactors

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Background

**Sonochemistry**
is traditionally working in
**batch reactors**

A recent new paradigm:
**Process intensification** via
**sonicated small scale flow reactors**
→ better process control
→ use the raw material efficiently
   and reduce waste
→ improve product quality
→ ease the scale-up
Sonochemical micro-flow reactors

Several possible geometries to sonicate the small channels:
→ submerged tubes in bath (coupling liquid)
→ tubes clamped or glued directly on transducer
→ machined or etched channels mounted on transducer
→ combinations
→ …

*Sarac et.al. Chem. Eng. Process. Process Intensif 150 (2020)*

*John. et.al. Chem. Eng. Process. Process Intensif. 102 (2016)*
How to initiate cavitation?

In any case cavitation in small enviroment:
→ milli- and micro-channels
→ small liquid volumes, clean walls, no air contact
→ cavitation bubble **nucleation** can be a problem!

One approach:
gas phase in the channel
→ "Taylor flow"
   = gas/liquid slug flow
→ "free" boundaries

Gupta et al. 2010, after Triplett et al. 1999
Nucleation at liquid / gas phase boundary

Capillary waves $\rightarrow$ bubble seeding (gas into liquid)
$\rightarrow$ droplet ejection (liquid into gas)

Tandiono et al., Lab Chip 10 (2010)
100 kHz

Sarac et al., Chem. Eng. Process. Process Intensif 150 (2020)
25 kHz
Reminder: Ultrasonic wetting of holes (last Workshop Drübeck 2019)

1 mm blind hole, length : 1.2 mm

Let’s focus on the drops now!

At high amplitude capillary waves inject droplets to gas phase

→ “internal atomization”

M. Kauer et al., Ultrasonics Sonochemistry, 48 (2018)
Microreactor system

→ meandering channel etched in silicon, glued on piezo plate transducer
→ channel dimensions 1.2 mm x 0.6 mm, length 740 mm
→ Liquid/gas **Taylor flow** (slug flow) of water and CO$_2$
Simulated transducer oscillation (411 kHz)

→ High order mode(s) excited: “nodes” and “antinodes”

Comsol Multiphysics
Atomization at phase boundaries

→ Surface waves and droplets

silent

gas

liquid

Silent

47 kHz

411 kHz

601 kHz

47 kHz

411 kHz

601 kHz

500 μm
Atomization at phase boundaries

movie at 411 kHz driving

→ sometimes violent drop ejection, sometimes none!
Since always imaged at the same position:
→ no antinode ("hot spot") effect
Resonant internal atomization

→ conjecture: acoustic resonance in liquid slug amplifies sound field
Resonance confirmed by particles gathering at nodes

→ polystyrene particles in liquid (1 wt%, 10 μm diam.)
Resonance confirmed by particles gathering at nodes

→ particles in resonating slugs are transported by flow (again: no "hot spots")
Simulated slug resonance

→ sound field indeed amplifies in resonant slugs
   (here superimposed by modal oscillation of plate transducer)

Comsol Multiphysics
Simulated slug resonance

- resonance depends on slug length
- also resonances in gas phase possible

Comsol Multiphysics
Resonance in gas volume

→ Droplets show “Kundt’s tube” behavior

\[ \frac{1}{2} \lambda_{g} \]
Conclusion

→ peculiar phenomenon of **massive droplet ejection** (atomization) in micro-channels

→ mechanism relies on **acoustic resonance** in liquid slugs

→ numerical **simulations capture essentials** of this resonance effect

→ considerable **amplification of acoustic pressure** within slugs, also facilitating cavitation

→ acoustic **resonance in gas bubbles** can also partly be observed

→ **atomization mechanism** (at least partly) based on capillary waves and cavitation / entrained bubbles at interface possibly more details to explore…
Outlook

→ further analysis of **physical mechanisms** (e.g. details of atomization, flattening of slug wall: radiation pressure?,...)

→ **application**: increase of gas/liquid interfacial area and **increase of mass transfer** in microreactors

→ resonance within microchannels might **increase efficiency of ultrasound** used for clogging prevention, emulsification or particle size reduction in microreactors

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Thank you for your attention and enjoy the discussions !!!