Sonoluminescence emission spectra of a 3.6 MHz HIFU in sweep mode, applied to control magnesium dissolution

RESEM Project VOBUSURF

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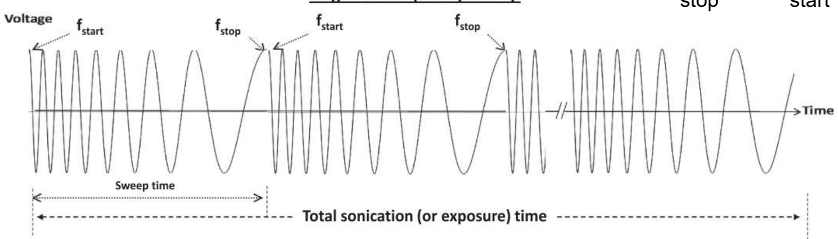
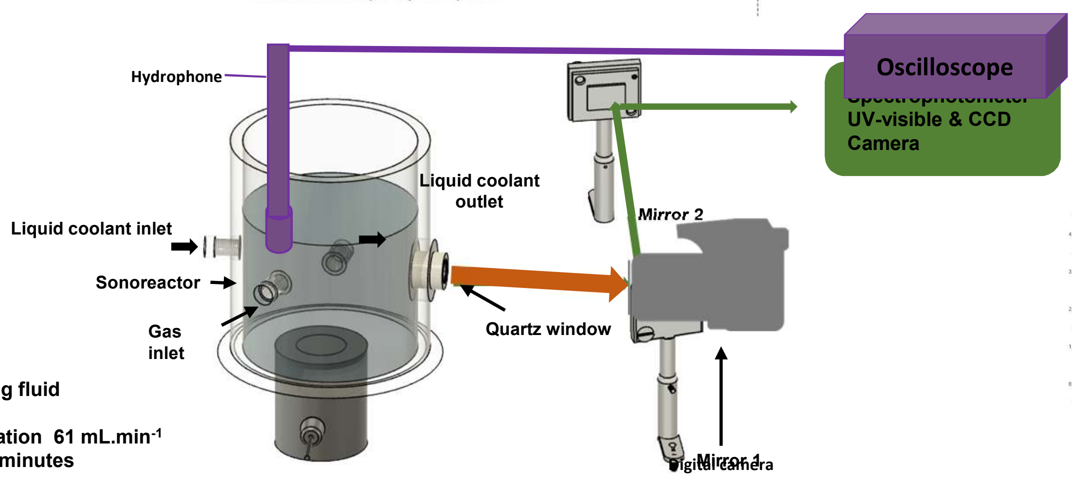
Negative Frequency Sweep $\Delta F < 0 : F_{stop} < F_{start}$



$$r_{sweep} = \frac{\Delta F}{T_{sweep}} \text{ (MHz/s)}$$

$$\Delta F = F_{stop} - F_{start} \text{ (MHz)}$$

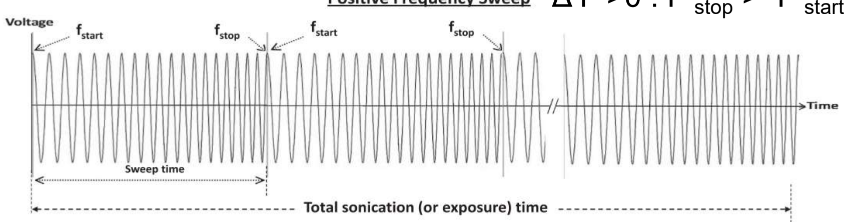


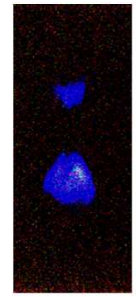
5°C cooling fluid

Gas saturation 61 mL.min^{-1} during 30 minutes

3.6 MHz HIFU

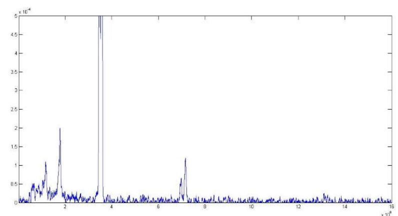
Positive Frequency Sweep $\Delta F > 0 : F_{stop} > F_{start}$



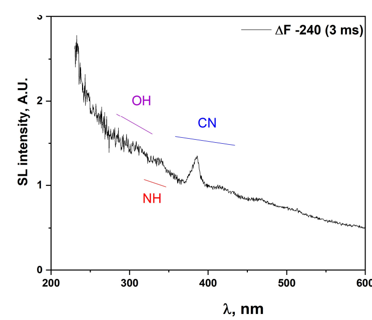


3600-3520 kHz
1 ms

- ❑ **Sonochemiluminescence SCL**
  - Distribution
  - Intensity

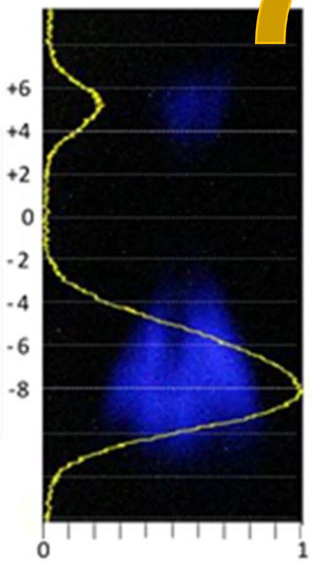


- ❑ **Acoustic Spectra**

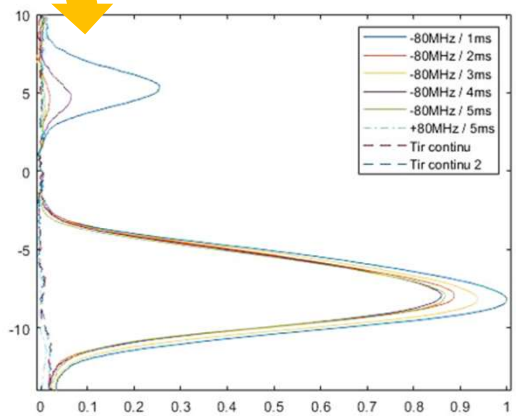


- ❑ **Sonoluminescence SL**
  - Plasma nature

Localising and quantifying active bubbles



300 ml of 0.01 M luminol solution (pH = 10.8, Na_2CO_3), Ar-20%O_2, exposure time 1 minute at 3600 kHz -80 MHz/s (1ms)



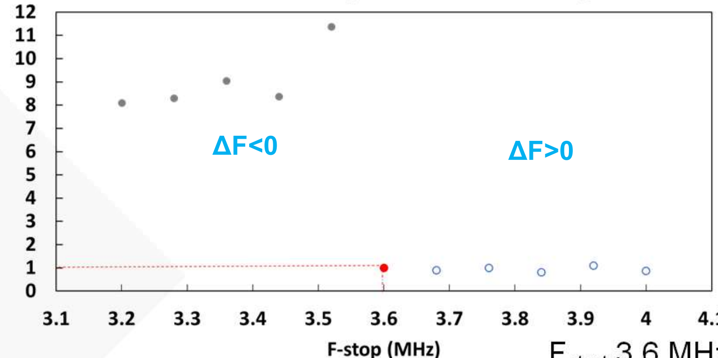
Scanning direction:

$\Delta F < 0 : F_{stop} < F_{start}$

$\Delta F > 0 : F_{stop} > F_{start}$

$$r_{sweep} = \frac{\Delta F}{T_{sweep}} \text{ (MHz/s)}$$

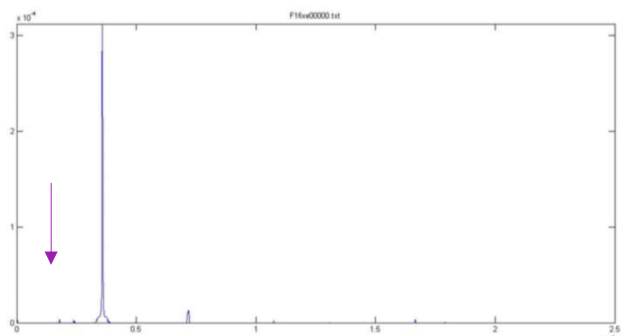
Normalized integrated luminosity for SCL



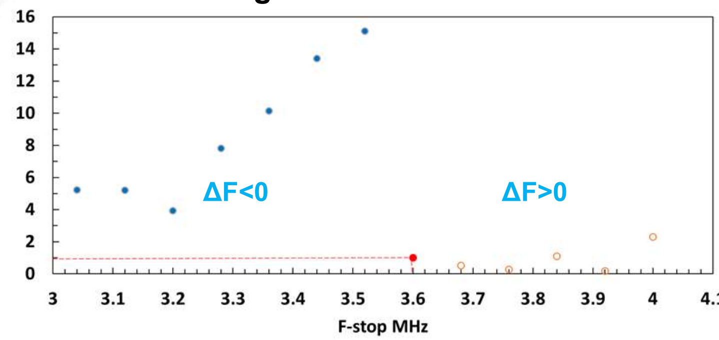
F_{start} 3,6 MHz

Rate ± 80 MHz/s

3,6 MHz fixe 100 mV



Normalized integrated Area below acoustic curve



- Post-focal
- Pre-focal, more bright = more active
- Focal zone non active

- Harmonics, subharmonics, ultraharmonics more visible ► active bubbles
- ↑ continuum ► inertial cavitation

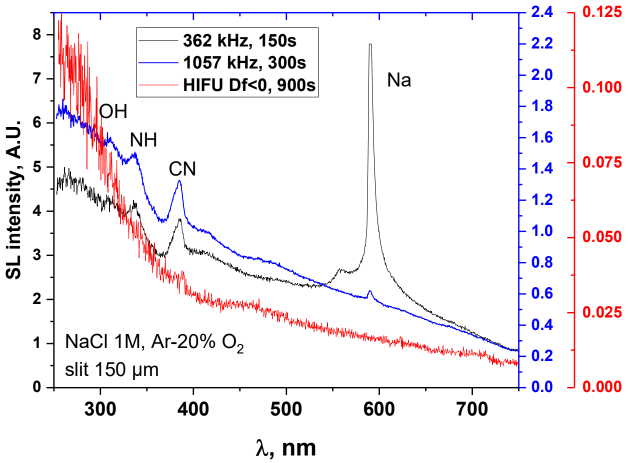
- Same trend cavitational activity
- ΔF >0 sonochemical activity is more strong, especially for short T_{sweep}

Plasma nature

SL spectra → identification of active species in the plasma generated in the bubbles

Above 500 kHz :

- Very low SL intensity
- Broad molecular emission difficult to study



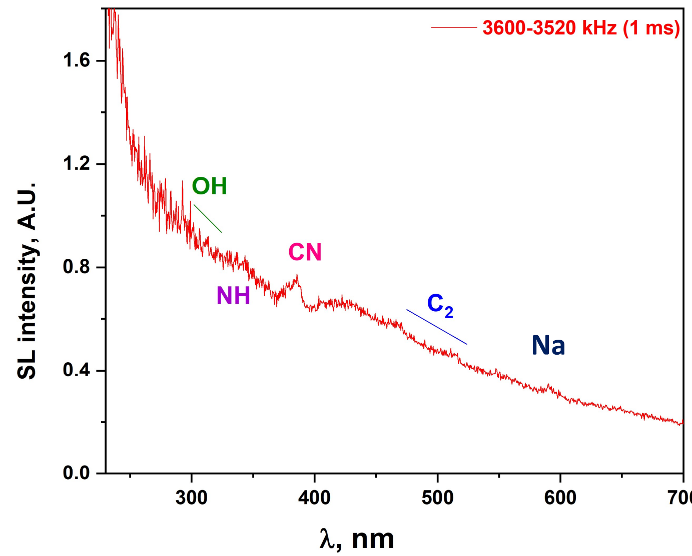
Detected species:

OH 310 nm; **NH** 337 nm ;**CN** 386 nm

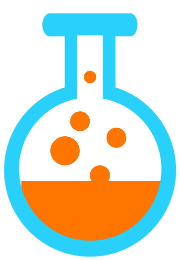
C_2 470 and 515 nm ;**Na** 589 nm

- Molecular emission of OH, NH, Na, C_2 very low
- **CN emission (B2Σ+ - X2Σ+) the most intense = Spectroscopic probe**

CO_2→ US →CO + O
N_2 + CO→ US →**CN** + NO

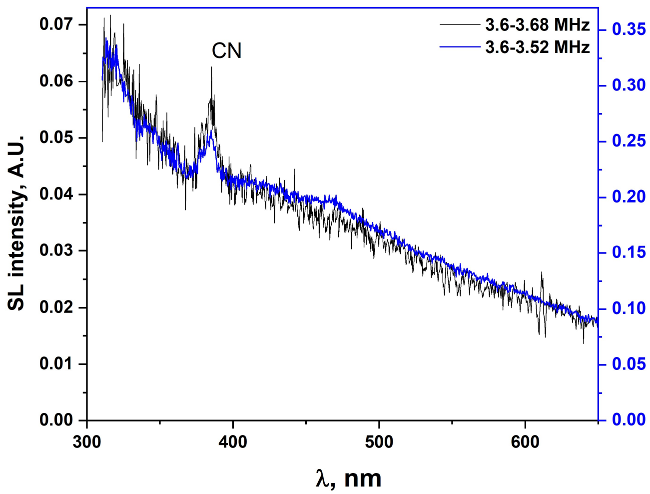


SL emission spectra of HIFU in sweeping mode, in 1 M NaCl +3.7 mM 2-propanol under Ar-15.5%O_2-2.2%N_2 at f= 3600 -3520 kHz, -80 MHz/s (1ms), merged with the corresponding spectra using high pass Filter 320 nm.

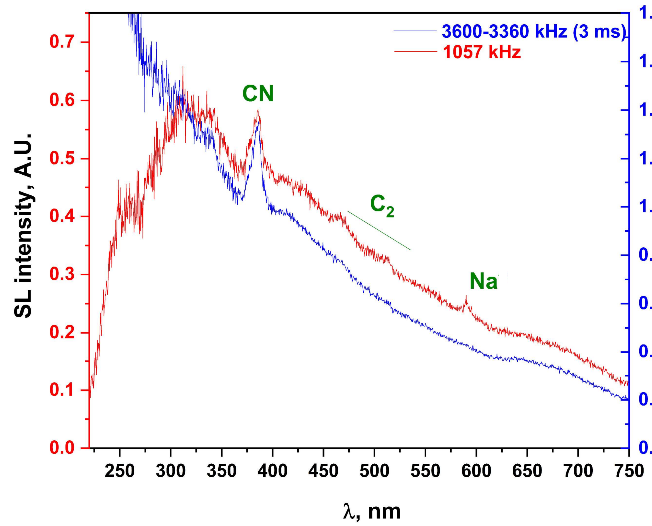


- ❖ Solution : **1M NaCl, 3.7 mM 2-propanol**
- ❖ Gas: **Ar/O_2 (15,5% $_{vol}$)/ N_2 (2.2% $_{vol}$)**
- ❖ Acquisition time: **15 minutes**

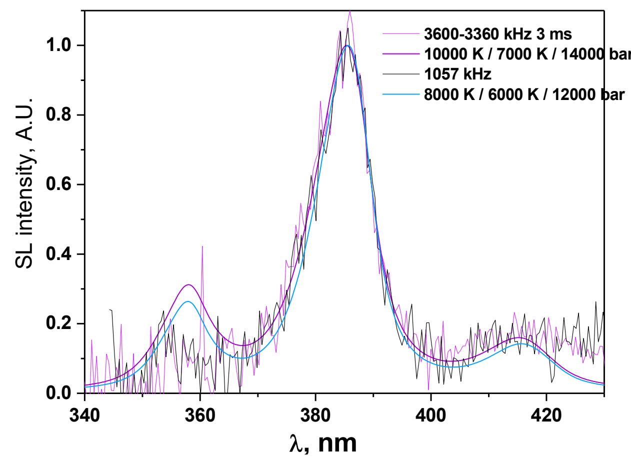
Simulation of CN molecular emission



SL emission at sweeping rate of ±80 MHz/s and a sweeping time of 1 ms for different F_{start}



SL emission spectra of HIFU at f= 3600 - 3360 kHz, -80 MHz/s (3ms) and fixed frequency of 1057 kHz,

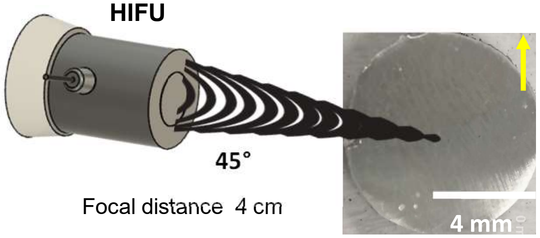


- Df>0 and Df<0 spectra are **similar** and **overlap**
- No spectral differences
- Difference in **intensity** (different number of bubbles)

- Plasma characteristics inside the bubbles do not depend on the frequency
- SL/SCL intensification is related to the number of bubbles

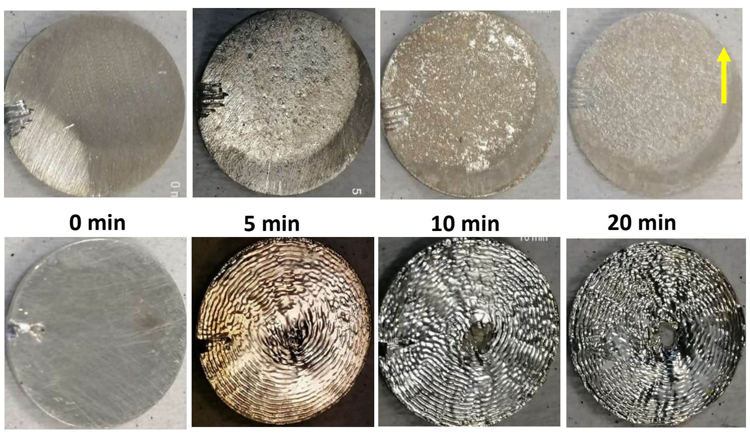
Rovibronic temperatures	**HIFU 3.6 MHz**	**1057 kHz**
T_v (K) ± 2000	10000	8000
T_r (K) ± 2000	7000	6000
P eff (bar) ± 2000	14000	12000

Magnesium erosion



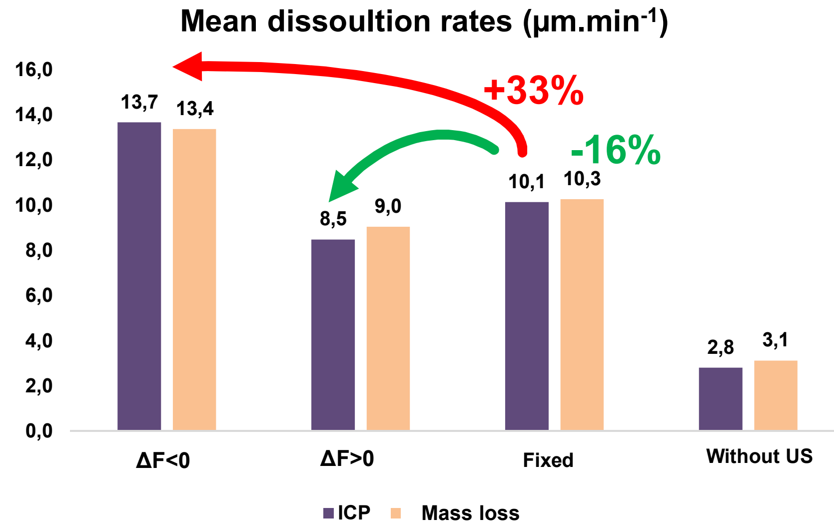
Flow direction of the acoustic stream.

Mg sample
Ø 9 mm
H 1.5 mm



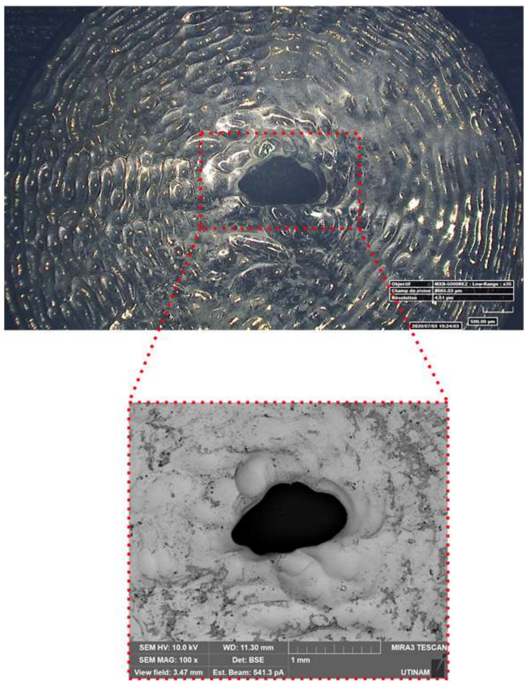
Mg evolution in 0.1 M oxalic acid with and without US for 20 minutes 3.6-3.44 MHz (2 ms), sweeping rate of -80 MHz/s.

- Brighter surfaces
- Appearance of macro-roughness

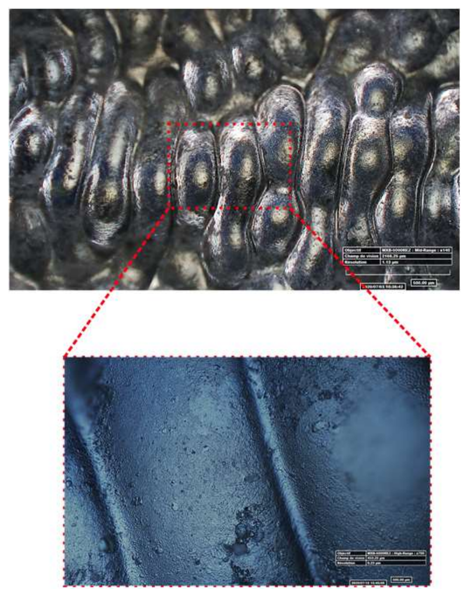


- Highest cavitation ΔF<0
- Fastest dissolution ΔF<0
- Up to 1.5 mm hole
- Dissolution kinetics in the center of the sample : 150 $\mu m.min^{-1}$
- Controlled dissolution

Magnesium erosion



Hole 1.4 mm x 0.8 mmx 1.5 mm
► **surface selectivity**



Increase for **surface roughness**
2.821µm → 12.961 µm
Appearance of wave

- ❖ Spectroscopic probe : HIFU 3.6 MHz + Sweeping frequency
- ❖ Unlock the hidden keys behind HIFU's plasma: T_v ;T_r
- ❖ HIFU's a workable solution for magnesium erosion :
  - Surface selectivity
  - High dissolution rates
  - Control surface roughness
- ❖ Other applications: decontamination, polishing, local and precise treatment (existing patent)

Thank you for your attention !

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