Microbubble histogram reconstruction by nonlinear frequency mixing

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In the 4th generation Sodium Fast nuclear Reactors (SFR), different phenomena can lead to gaseous microbubbles presence in the primary liquid sodium pool. This paper investigates the ability of nonlinear acoustics techniques to characterize these microbubbles presence. The goal is here to determine the void fraction (volume fraction of free gas) and the histogram of bubbles radius. Different acoustic techniques are currently developed at CEA. Among others, the nonlinear mixing of two frequencies [New84] is under study. Based on the nonlinear behavior of bubble resonance, this technique allows determining the radius histogram of a bubble cloud. Two different mixing techniques are here presented: the mixing of two high frequencies and the mixing of a high and a low frequency. The first step is an air-water experimental set-up. Microbubbles clouds are generated with a like dissolved air flotation process and an optical device gives us reference measures. Generated bubbles have radii in the range of several microns to several tens of microns. The developed experimental procedure allows us to determine the bubble size's histograms with accuracy never reported yet. [New84] V.L. Newhouse, P.M. Shankar - J. Acoust. Soc. Am., Vol. 75(5), p.1473-1477, 1984

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Introduction

The characterization of two-phase mixtures is a common problem. In the sea, in food, in many industrial processes... some bubbles are sometimes present and need to be characterized. Depending on the density and the size of the bubbles and the liquid phase nature, many different techniques are proposed in the literature: optical methods, acoustic methods, electrical or electromagnetic methods, methods based on the emission of ionizing radiation...

In the 4th generation nuclear reactors – specifically named SFRs (Sodium Fast Reactors) – liquid sodium is used as coolant. In SFRs, phenomena such as gas entrainment or nucleation can lead to the presence of gaseous microbubbles within the primary vessel liquid sodium. This presence has no direct impact on the core neutronics but it can lead among others to gas pocket accumulation and sodium acoustics properties (attenuation and speed of sound) modifications. To assess these consequences, different bubbles characterization techniques are currently developed at CEA.

The feedback of the French SFR Superphénix shows that we should expect a void fraction (volume fraction of free gas) value around 10^{-6} and bubble radii comprised between 10 and 100 microns. Adding the fact that the liquid sodium is opaque, we turn our attention to microbubbles clouds characterization techniques based on acoustics.

Several acoustical techniques are applicable in order to characterize gaseous bubbles in liquids: low-frequency speed of sound measurements based on Wood’s model, attenuation measurements... In 1973, Zabolotskaya and Soluyan proposed a method derived from Westervelt's parametric arrays: the nonlinear mixing of two frequencies.

The Principle of Nonlinear Frequency Mixing

When a system is acoustically nonlinear and is excited by two waves having frequencies \( \omega_1 \) and \( \omega_2 \), a particular phenomenon occurs: the mixing of the frequencies. It leads to emission of waves – independent from the initial ones – by the system named « sum » and « difference »: \( \omega_1 + \omega_2 \) and \( \omega_1 - \omega_2 \).

The bubble resonance phenomenon is highly nonlinear. The Keller-Miksis’ modeling of bubble dynamic – derived from Rayleigh’s one – shows it. Under Parlitz et al. reformulation, Keller-Miksis’s model gives:

\[
\left(1 - \frac{\dot{R}}{c}\right)R\dot{R} + \frac{3}{2} \ddot{R}^2 \left(1 - \frac{\ddot{R}}{3c}\right) = \left(1 + \frac{\ddot{R}}{c}\right) \frac{P(t)}{\rho} + \frac{R}{\rho c} \dot{P}(t)
\]

\[
P(t) = \left(p_0 + \frac{2\sigma}{R_0} - p_v\right) \left(\frac{R_0}{R}\right)^3 - \frac{2\sigma}{R_0} - \frac{4\mu}{R} \dot{R} + p_v - p_0 - P_i \sin(\omega t)
\]

where \( R \) is the instantaneous radius of the bubble, \( R_0 \) is its rest radius, \( c \) is the speed of sound in the liquid, \( \rho \) is the mass density of the liquid, \( p_0 \) is the static pressure, \( \sigma \) is the surface tension, \( p_v \) is the saturation vapor pressure, \( \gamma \) the gas polytropic index, \( \mu \) is the dynamic viscosity of the liquid and \( P_i \) the incident pressure amplitude.

The application of the mixing frequency technique to bubbles clouds is very interesting because the emergence of mixed frequencies in a linear medium will only be linkable with the presence of resonant(s) bubble(s) in the probed volume, contrary to most of the techniques based on linear acoustics. This technique also allows the deduction of the frequency of the resonant bubble; and as this frequency is directly linked with the bubble radius, this technique allows bubble characterization.

Zabolotskaya et al. were the first to conduct theoretical work on waves interactions generated by bubbles. And it is only in the early 80’s that the first experimentations of frequency mixing on bubbles and bubble clouds have been conducted.

On a bubble cloud, different applications of the frequency mixing principle are possible: the mixing of a high frequency with a low frequency (HF-LF) and the mixing of two high frequencies (HF-HF).

Mixing of a high frequency with a low frequency (HF-LF)

A high frequency \( \omega_1 \) – named imaging frequency – is selected substantially higher than the resonance frequency of the smaller bubble (i.e. presenting the higher resonance frequency). The low frequency \( \omega_p \) – named pump frequency – is selected in the range of potentially present bubble resonant frequencies.
When the pump frequency matches the resonance frequency of – at least – one bubble present in the probed volume, this bubble resonates and induces the apparition, among others, of the sum and difference frequencies: \( \omega_i + \omega_p \) and \( \omega_i - \omega_p \).

![FIGURE 1. Illustration of the HF-LF frequency mixing principle and example of obtained spectrum when the pump frequency matches a bubble resonance frequency.](image)

Considering the link between the resonance frequency of a bubble and its radius, then an incremental construction (by counting) is possible. A sweep over the whole range of potential resonance frequencies is made with the pump frequency and after the detection of enough bubbles, a statistically representative histogram of the bubble radii is obtained.

As the imaging frequency \( \omega_i \) is selected high, its beam is narrow, which allows obtaining a restrained probed volume (especially if the used transducers are focalized). Then the result is a histogram of the radii of the bubbles present in this volume and averaged on the probing time. If the probed volume is known, a deduction of the void fraction value is possible.

HF-LF mixing is historically the first frequency mixing technique that has been experimented, by Newhouse et al.\(^{11}\) on a bubble clouds with as aim to obtain information on their sizes. Later, Chapelon et al.\(^{12}\) then Koller et al.\(^{13}\) obtained histograms of the bubble radii with a validation by an optical measurement. In both cases, the correlation coefficient was average: around 0.8.

![FIGURE 2. The histograms of the bubble radii obtained by Koller et al. with a frequency mixing technique and a camera.](image)

More recently, Buckey et al.\(^{14}\) and Leighton et al.\(^{15}\) have obtained histograms with a HF-LF mixing technique but without validation of the obtained results.

**Mixing of two high frequencies (HF-HF)**

With this other implementation, the frequencies of the two emitted waves are substantially higher than the resonance frequencies of the present bubbles. One of the high frequencies \( \omega_1 \) is fixed and a sweep is operated with the second one \( \omega_2 \). If the difference between the two emitted frequencies \( \omega_2 - \omega_1 \) matches the resonance frequency of
– at least – one bubble present in the probed volume, the induced resonance of this bubble is responsible for the emission of a new wave which frequency equals this difference.

Like with the HF-LF technique, the sweep with $\omega_2$ is operated in order to cover the whole range of resonant frequencies of potentially present bubbles with $\omega_2 - \omega_1$, allowing the construction of the histogram of the bubble radii.

Compared to HF-LF technique, few experimental validations of HF-HF technique have been detailed in the scientific literature.

In 1980, Kobelev et al.\textsuperscript{16} was the first to show experimentally the emission of a low frequency by a bubble cloud excited with two high frequencies. After him, Sandler et al.\textsuperscript{17} and Gimenez et al.\textsuperscript{18} studied bubble detection with a HF-HF mixing technique. Sutin's work\textsuperscript{19}, published in 1998, is to our knowledge the first and only so far on the construction of a complete histogram of the bubble radii of a cloud with this technique. The results were considered consistent by their author but have not been validated by a reference measurement. So far, mixing HF-HF has therefore never allowed obtaining validated histograms of bubble radii.

The main advantage of the HF-HF mixing technique in the HF-LF technique is that with the HF-HF technique, it is technologically much easier to cover a very wide range of resonance frequencies of bubbles and thus bubble sizes.

**Experiments**

Since 2010, several experiments have been conducted at CEA. Both techniques HF-LF and HF-HF have been tested in water-air devices. Microbubbles clouds are generated thanks to an aeroflotation set-up. This technique is based on cavitation in air-oversaturated flows. It allows the nucleation of microbubbles which radii are comprised between few microns and less than 100 microns. Acoustical construction of the histograms of bubbles radii needs a reference measurement for validation. This reference is issued from the image processing of videos given by an immersed camera.
**HF-LF mixing technique experimentations**

The configuration of the implemented acoustic device used is described in figure 5. A LF transducer centered at 250 kHz and connected to a function generator, produces the pump frequency. The imaging frequency is emitted by a HF transducer – connected to a function generator – centered at 2.25 MHz, which is focused at a distance of 30 mm. An identical transducer is used as receiver. This last is connected to a 12 bit analog-to-digital converter, which is itself connected to a personal computer.

The LF chirp is emitted between 10 and 500 kHz thus allowing the full range of resonant frequencies of the present bubble sizes (between 7 and 330 microns) to be covered.

**FIGURE 5.** Acoustic devices configuration for the HF-LF mixing technique experimentations.

For more details on the HF-LF experiments, especially about the used devices, see references 10 and 20.

**HF-HF mixing technique experimentations**

The HF-HF mixing configuration is really close: the same HF transducers are used as emitters and the LF transducer is substituted by a Brue & Kjaer 8103 hydrophon.

A chirp is emitted with one of the HF transducers between 2.1 and 2.55 MHz and the other HF transducer emits at 2.05 MHz. It allows covering a range of bubble resonant frequencies which radii are comprised between 7 and 70 microns.

**FIGURE 6.** Acoustic devices configuration for the HF-HF mixing technique experimentations.

For the time being, due to experimental configuration, it has not been possible for us to conduct the two different mixing techniques on the same bubble cloud, in parallel of an optical validation measure. So here we give the optically validated results obtained with the HF-LF mixing technique, as previously published\(^\text{[20]}\), and the results
obtained in a second phase with the HF-HF mixing technique applied on a bubble cloud that was generated by the device used for the HF-LF mixing experiments.

**Results**

Figure 7 shows a spectrum revealing the presence of a bubble, which is resonant at the pump frequency: 280 kHz. Sum and difference frequencies can be observed at 1.97 MHz and 2.53 MHz. Many others frequencies – proof of the strong nonlinearity of bubble resonance – are present: harmonics, ultraharmonics and maybe subharmonic of the pump frequency and harmonics, subharmonics and unexplained “sub-subharmonics” of the sum and difference frequencies.

![FIGURE 7. Example of obtained spectrum during the resonance of a bubble excited with a HF-LF mixing technique.](image)

The construction of the histograms of the bubble radii has been based on the sum and difference frequencies detection only. A threshold is defined and each time that a sum or difference frequency appears, its resonance frequency, and thus the bubble’s radius is deducted. The corresponding radius of the histogram is incremented and after the detection of enough bubbles, a complete and representative histogram is obtained.

Figure 8 is an example of a histogram obtained with HF-LF mixing technique and an optical device. The histograms obtained by optical and acoustic methods present a good agreement with a correlation coefficient of 0.94. Such result has, to the best of our knowledge, never been reported before.

![FIGURE 8. Normalized histograms of the bubble radii, obtained thanks to an optical method and a HF-LF mixing frequency technique.](image)

HF-HF mixing technique has been conducted with the same microbubble generation device but not exactly in the same conditions. Figure 9 is the obtained histogram. HF-HF technique does not allow to detect as many bubbles as the HF-LF one. In consequence, the histogram presented here is not perfect yet but it is in coherence with the histograms presented on figure 8.
FIGURE 9. Normalized histogram of the bubble radii, obtained thanks a HF-HF mixing frequency technique.

We hope to obtain soon as good results with the HF-HF technique as with the HF-LF technique. Awaiting this, the results obtained here are promising for our industrial application: the SFRs liquid sodium. Indeed CEA has so far only one kind of hot sodium-proof ultrasonic transducer: the TUSHT. This transducer has several resonances frequencies comprised between 0.5 MHz and 5 MHz and a HF-HF mixing technique seems to be easier to implement with it than a HF-LF mixing technique.

Conclusions and Prospects

So far, no miraculous technique has yet been developed to characterize all types of two-phase liquid-gas media. For low density microbubble clouds encountered in Sodium Fast Reactors, results obtained with frequency mixing techniques are promising. It has been proven that these techniques are appropriate for the construction of histograms bubble radii with a good accuracy.

We hope to be soon able to get simultaneous optical, HF-HF based and HF-LF based histograms. We also work on the exact probed volume knowledge in the aim to deduct from the histograms the void fraction values.

We also plan to try to exploit other mixed frequencies than "simple" sum and difference frequencies. According to Leighton et al. this could increase the results quality.

In order to implement these techniques on Sodium Fast Reactors, we are actually developing the ACWABUL (Acoustic Characterisation in WAter of BUbbLes) set-up. This bench should allow us to further experiments under conditions closer to real industrial conditions: large and fully bubbly volume, variable void fraction and bubble radii histograms... In addition it will allow us to try to validate these techniques with TUSHT before considering experiments in liquid sodium.

Seen the acoustic similarities between water at 20°C and sodium at 550°C, if the implementation of frequency mixing techniques were validated in water with TUSHT, there could be a good chance that it would work in liquid sodium and thus in SFRs.

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