Ultrasonics Sonochemistry 28 (2016) 346–356

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

journal homepage: www.elsevier.com/locate/ultson

Cavitation and non-cavitation regime for large-scale ultrasonic standing wave particle separation systems – *In situ* gentle cavitation threshold determination and free radical related oxidation

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ARTICLE INFO

Article history:
Received 7 April 2015
Received in revised form 17 July 2015
Accepted 7 August 2015
Available online 7 August 2015

Keywords:
Ultrasonic large-scale particle separation
Cavitation threshold
Luminol
Sonochemiluminescence
Lipid oxidation
Milk separation

ABSTRACT

We here suggest a novel and straightforward approach for liter-scale ultrasound particle manipulation standing wave systems to guide system design in terms of frequency and acoustic power for operating in either cavitation or non-cavitation regimes for ultrasound standing wave systems, using the sonochemiluminescent chemical luminol. We show that this method offers a simple way of *in situ* determination of the cavitation threshold for selected separation vessel geometry. Since the pressure field is system specific the cavitation threshold is system specific (for the threshold parameter range). In this study we discuss cavitation effects and also measure one implication of cavitation for the application of milk fat separation, the degree of milk fat lipid oxidation by headspace volatile measurements. For the evaluated vessel, 2 MHz as opposed to 1 MHz operation enabled operation in non-cavitation or low cavitation conditions as measured by the luminol intensity threshold method. In all cases the lipid oxidation derived volatiles were below the human sensory detection level. Ultrasound treatment did not significantly influence the oxidative changes in milk for either 1 MHz (dose of 46 kJ/L and 464 kJ/L) or 2 MHz (dose of 37 kJ/L and 373 kJ/L) operation.

1. Introduction

Ultrasound separation in large-scale systems is an emerging field [1] and the optimal parameter space for specific applications is yet to be determined. Often the frequency range of 0.4–2 MHz has been employed [1]. From basic knowledge of sonochemistry, a power dependent cavitation threshold is expected, around the 1–2 MHz range. Ashokkumar et al. [2] found OH^- radicals peaking at values at 0.358 MHz while maxima from 0.582 to 0.863 MHz were obtained in a study by Mason et al. [3] Since the pressure field is system specific the cavitation threshold is system specific (for the threshold parameter range). Cavitation phenomena are not usually evaluated for large-scale ultrasonic standing wave particle separation systems. High acoustic power in large-scale ultrasound separation is desired to enable high volume throughputs, but need to be below the level at which strong acoustic streaming is observed to disrupt particle alignment. We will demonstrate that for separation under strict non-cavitation conditions an additional restriction on the power level should be employed, especially when operating in the 1–2 MHz frequency. The power level varies with transducer driving parameters, geometric transducer and vessel design and with location in the standing wave field. The method presented here generates knowledge of the cavitation threshold *in situ* for a certain geometry, frequency and power level. This enables operating at the highest throughput conditions while remaining in the non-cavitation regime. If the sample properties differ significantly from water, this needs to be compensated for.

The parameter space desired for acoustic particle multi-node manipulation is outlined schematically in Fig. 1. For operation in non-cavitation ('gentle') conditions, low frequencies should be avoided. It is known that cavitation activity increases with increasing pressure amplitude and with increasing frequency until peak values are reached [4]. For the amplitude these peak values are higher than what we operate at here, but for the frequency the peak is around 0.4–0.9 MHz as described above, i.e. lower than the regular range for acoustic large-scale separation (see Fig. 2). While the power level cavitation onset is not usually studied in

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http://dx.doi.org/10.1016/j.ultsonch.2015.08.003
1350-4177/

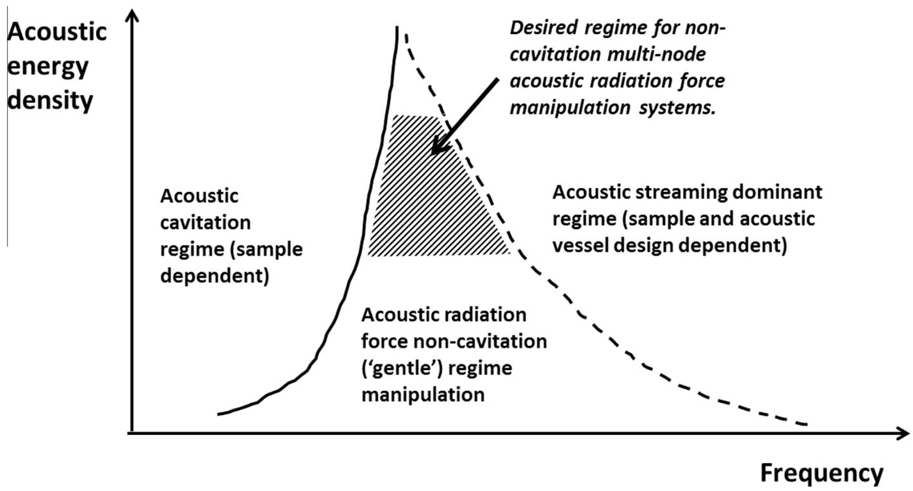

Fig. 1. Phenomenological schematic parameter space for operation of multi-node acoustic radiation force manipulation systems, in terms of acoustic energy density and ultrasound frequency. The desired operation range is at a frequency range above cavitation onset and below acoustic streaming onset and at high enough acoustic energy density to obtain a high enough level of acoustic radiation force.

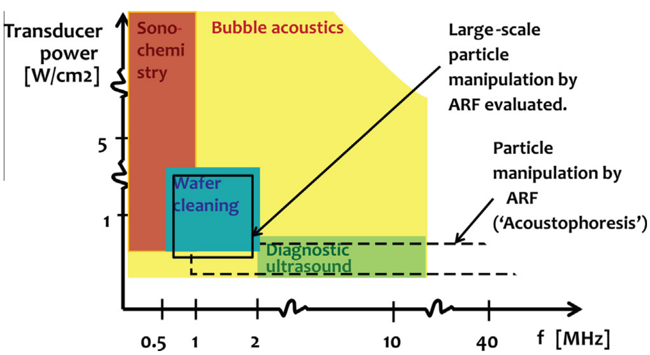

Fig. 2. Estimated parameter space from an equipment point of view indicating the mainly employed regions for selected fields of ultrasound research and applications.

detail around 2 MHz and above for processing applications, the cavitation power threshold is known to shift to higher energy densities for higher frequencies. This is demonstrated for instance in biomedical applications by theoretical relations of peak negative pressure cavitation threshold as a function of initial nucleus radius for 1, 5 and 10 MHz for short-pulse (μs range) low duty-cycle by Apfel and Holland [5], in numerical modeling up to 1 MHz by Merouani et al. [6], experimental results of 3 W at 1.7 MHz and 8 W at 2.4 MHz in the system studied by Kirpalani et al. [7] and theoretically for stable cavitation (measured as sub-harmonic emission) by Bader et al. [8], see further Section 4). The exact appearance of the cavitation threshold in this high frequency range is not known but here we illustrate it with a curve with increasing slope, in analogy with measurements by Barger up to 1.16 MHz [9] and with the upper part of a cavitation activity versus frequency curve, see for instance Koda et al. [10]. Acoustic streaming will limit the multi-node system operation at high frequencies due to the onset of Eckhart type streaming, where the exact threshold level depends on factors such as the acoustic energy density (gradient) level, the sample type and the vessel design [11]. The absorption coefficient increases with frequency squared for many samples. The frequency dependency of the acoustic streaming onset is conceptually illustrated as nonlinear (dotted line in Fig. 1), rising and falling more rapidly than would a linear trend. Due to the inertial nature of acoustic streaming the streaming onset only occurs at high enough energy densities and frequencies. Further, it can be described that the onset frequency will occur at a lower frequency for higher energy density (here schematically illustrated by the dotted line). Generally, high power enables operation at high volume throughput, i.e. short residence times and or large vessels. Higher power levels are required also for manipulation of smaller particles and low acoustic contrast factor particles, for which the acoustic radiation force is lower [12,13]. Hence, based on these three phenomena the desired operation range for multi-node acoustic radiation force manipulation systems is believed to be within a relatively narrow frequency range and at high enough energy density to provide the high enough level of acoustic radiation force, as indicated in Fig. 1 (striped area).

Particle manipulation systems have been extensively evaluated mainly since the early 1990s in batch design as reviewed by Coakley [14] and since around 2001 [15] in microscale systems as reviewed by Laurell et al. [16]. In the field of microfluidics, cavitation effects are generally avoided by not operating below 1 MHz [17]. By analyzing the literature in view of different research fields and applications it can be observed that the large-scale ultrasonic separation systems evaluated more recently [1] usually employ lower frequency and higher transducer power levels, see Fig. 2, in addition to batch system and multi-node design [1,17]. According to the authors' knowledge frequencies above 2 MHz have not been employed for these large scale batch systems [18]. The transducers employed to date for large-scale batch operation are not seldom the ones developed for semiconductor silicon wafer cleaning applications, for which cavitation is mandatory.

One application evaluated for the multi-node large throughput acoustic radiation force manipulation is milk fat separation [1,19]. In this case operating in non-cavitation conditions firstly avoid exposure to cavitation generated radicals that oxidize free fat and reduce the nutritional related antioxidant activity provided by other components in the sample. Secondly, high temperatures and shear stresses reached very locally, close to a cavitating bubble, can potentially also modify molecules and structures in the sample. Thirdly, operating in the non-cavitation regime is also advantageous for avoiding the risk of generating micro and nano sized metal particles deposited in the sample from the transducer and vessel surfaces by cavitation related effects as recently described and evaluated in an enlightening article by Mawson et al. [20]. Mechanisms such as cavitation pitting of the surface, ultrasonic peening, oxidation related corrosion and enhanced corrosion by cavitation disruption of a surface passivation layer were