

THE ULTRASONIC h-SHAPE SEPARATOR: HARVESTING OF THE ALGA SPIRULINA PLATENSIS UNDER ZERO-GRAVITY CONDITIONS

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Abstract

The forces on suspended particles in acoustic fields are reviewed briefly and the theoretical modelling of ultrasonic separators based on piezoelectrically excited layered resonators is described. The so-called h-shaped ultrasonic separator is analysed by combining the mathematical modelling of the laminar flow with the acoustic force based velocity field of the particles relative to the suspension medium. This allows a complete modelling of the resonator's particle separation performance. An example for separation chamber design optimized by use of the mathematical model is presented and the calculated particle traces in the h-resonator are shown and compared with experimental results. The presented results are of importance for ultrasonic separation of plant (algae) cells under low gravity conditions, where the sedimentation concept fails.

Introduction – the phenomenon

The use of acoustic standing waves to concentrate initially homogeneously suspended particles at acoustic pressure nodal, or antinodal, planes within a fluid was first described by Kundt und Lehmann [1]. The effect was originally used only to make ultrasonic fields visible. However, the interaction of standing ultrasonic waves with particles dispersed in a fluid produces forces on the particles which can be utilized for the separation of the dispersed particles from the fluid as well [2-36].

Fig.1 shows the effect of a standing wave on a suspension consisting of small Pyrex glass spheres with diameter $d \approx 100 \mu\text{m}$ in water. The standing wave of frequency $f = 0.67 \text{ MHz}$ is generated in an acrylic glass tube with an inside diameter of 40 mm, whereby the amplitude of the velocity field shows a symmetric distribution in lateral direction around the axis of symmetry of the tube. The theoretical explanation of the obeyed migration of the initially homogeneously dispersed glass spheres is given in the following Section, here it is only qualitatively described by means of Fig.1. At the beginning of irradiation, almost instantaneously the spheres are driven towards the acoustic pressure nodes, whereby the average distance between the particles considerably diminishes. Then the particles trapped within the planes migrate

closer together, whereby coagulation and even coalescence may be triggered.

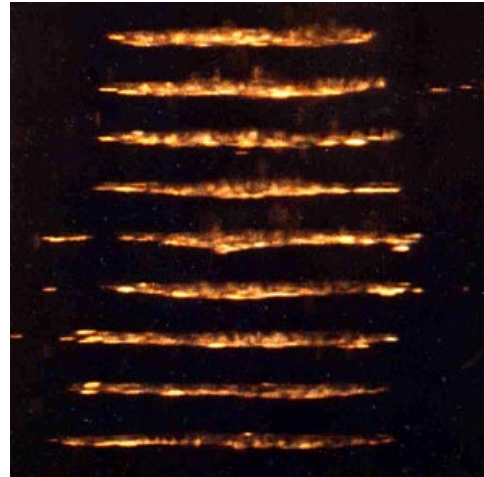


Fig.1. Photograph of the pattern of particle positions in a vertical ultrasonic standing wave field. The example shows spherical Pyrex glass particles with diameter $d \approx 0.1 \text{ mm}$, excitation frequency is 670 kHz corresponding to a half-wave-length $\lambda/2 = 1.1 \text{ mm}$, viewing window 1 cm x 1 cm.

The phenomenon is applicable to all kinds of dispersions. Table 1 shows the common characterization of dispersions according to the physical state of the particles and the dispersion medium. The particles can be gaseous, liquid, solid, or even biological cells. The dispersion fluid can be gaseous or liquid. The most important practical examples are particles of all kinds in air (aerosoles) or in water (hydrosoles).

Table 1. Kinds of Dispersions

Medium → ↓ Particle	Liquid	Gas
Solid	Suspension	Smoke
Liquid	Emulsion	Fog, Mist
Gas	Dispersed Bubbles	

The overview about all possible kinds of dispersions given in Table 1 reviews the established characterising terms and indicates the great potential of the ultrasonic separation technology. Nevertheless, the

phenomenon has not yet gained widespread industrial application as the process can be highly sensitive to disturbances and involves acoustic forces that have to be compared with the separation speed limiting viscous drag forces.

Although an extensive literature on the theory of the interaction of ultrasonic waves with particles already exists, there is still a gap between interesting research projects and an efficient acoustical separation technology of practical importance. However, with nowadays available highly advanced piezoelectric transducers and driving electronics, some commercial applications of this separation effect became visible [13, 37, 38], and the mentioned gap has been reduced significantly by the progress made in a joint European effort, the EUSS network, within the European Commission's Training and Mobility (TMR) Program for young researchers [14, 19]. This paper is focussing entirely on the application of the ultrasonic separation technology to hydrosols including biocell suspensions, where most recent progress was made.

Acoustic radiation forces on suspended particles

Interaction between single particle and acoustic field

Acoustic mean forces on suspended particles can be subdivided into forces on an individual particle (single particle and acoustic field interaction), i.e., the primary radiation force, and into the interparticle force between two or more particles in a sound field, i.e., the secondary radiation force. At the beginning of irradiation, the strong axial component F_A of the primary radiation force drives the spheres towards the displacement velocity antinodes, by which the average distance between the particles considerably diminishes. Then, due to the transversal component F_T of the primary radiation force and the secondary radiation force F_I (Interparticle Force), the particles come closer together, whereby coagulation and even coalescence may be.

All the forces mentioned above can be derived as special limit cases from one common physical principle, the momentum flow analysis. This analysis of the total momentum flow passing through the regarded particle was first described by Gor'kov [39]. According to Gor'kov, the radiation force caused by a standing wave is usually several orders of magnitude larger than that caused by a travelling wave of the same amplitude.

The primary ("sound radiation") force in the propagation direction of the standing sound wave is the strongest force, which almost instantaneously drives the particles towards the antinode or node planes (dependent upon the acoustic properties of the dispersion) of the applied alternating acoustic displacement velocity field

$$\vec{F}_A(\vec{r}_p) = 4\pi\bar{k}a^3 \langle E_{ac} \rangle K(\rho_f, \rho_p, c_f, c_p) \sin(2\bar{k}\vec{r}_p), \quad (1)$$

whereby r_p is the locus vector, \bar{k} the wave vector, a the particle radius, $\langle E_{ac} \rangle$ the time averaged acoustic energy density, $K(\rho_f, \rho_p, c_f, c_p)$ the acoustic contrast factor, ρ_f, ρ_p density of fluid and particle, c_f, c_p sound speed in fluid and particle, respectively.

The derivation of this equation is based on the assumption that the particle radius is much smaller than the wavelength. From this equation it can be learned that the force is proportional to the third power of the particle radius, and thus is proportional to the particle volume. Further the acoustic radiation force is proportional to the time averaged acoustic energy density in the liquid. The spatial dependence has twice the periodicity of the standing wave, the direction of the force to or from the displacement velocity antinodal planes is determined by the sign of the acoustic contrast factor that depends on the ratio of the densities and sound speeds in the liquid and the particle material. As a general rule, solid particles are driven towards the displacement velocity antinode planes, while gas bubbles are driven towards the node planes.

Instead of the commonly used term "acoustic particle velocity" (in the german language "Schallschnelle") here and throughout this paper the term "acoustic displacement velocity" is used to avoid at one hand any confusion with the velocity of the dispersed particle and at the other hand with the propagation velocity of the acoustic wave.

Further concentration of the particles within the antinode or node planes of the displacement velocity amplitude occurs by the transversal component of the primary radiation force that is proportional to the gradient of the acoustic energy density. Within the antinode planes suspended solid particles are therefore driven by forces perpendicular to the direction of the sound wave propagation and pointing towards the lateral displacement velocity amplitude maxima. [40]

Interaction between two particles in an acoustic field

The forces between two particles situated in an acoustic standing wave field have first comprehensively been described by Bjerknes [41]. Within the displacement antinode planes, acoustic interparticle forces increase with the fourth power of the reciprocal distance between the particles and are therefore negligible for low particle concentrations. Since the concentration of the particles within the planes is much higher than in the initially homogenous dispersion, the average distance is much smaller and the chance for agglomerations due to interparticle forces increases. Nonetheless, agglomeration caused by these secondary forces typically needs several minutes. Besides that, the application of the effect to particle separation

is restricted to particles which tend to coalesce and flocculate. This property is needed since the final macroscopic particle separation in the conventional coagulation approach is performed by gravity forces which are effective only on the bigger flocculated clusters. These gravity forces are rather weak and, after switching off the acoustic standing wave field, again need typically several minutes to sediment the flocculated particles on the bottom of the resonance cell. Hence, the use of conventional acoustic standing wave fields dramatically restricts the application to a small share of the numerous technically important suspensions and needs relatively long periods of sound treatment. A more comprehensive review of quantitative relations for the various types of forces acting on spherical obstacles freely suspended in a fluid medium has been given recently by Gröschl [42].

Resonator analysis

Layered piezoelectric resonator model

For obtaining a well defined acoustic plane standing wave field, composite piezoelectric resonators are used that can be schematically described by a layered structure according to Fig.2. The electric excitation of the acoustic field is usually performed by PZT (Lead-Zirconate-Titan-ate) piezoceramic plates vibrating primarily in thickness extensional modes.

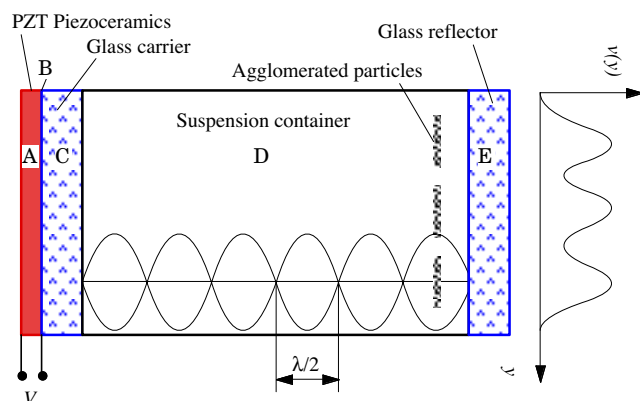


Fig.2. Scheme of a layered resonator. The outermost interface planes to the surrounding air function as almost totally reflecting acoustic mirrors for the acoustic wave excited (free surface boundary conditions). The glass plates function simultaneously as walls of the vessel for the liquid medium.

One-dimensional theory

The transfer matrix model of piezoelectric multilayer resonators used in this work for calculation of filter properties was developed by Nowotny and Benes [43, 44]. Application of the model to piezoelectric resonators for particle separation was described first by Gröschl [42]. The model is based on the fundamental equations of piezoelectricity [45] that relate

the coupled electroacoustic field quantities, acoustic displacement u , mechanical stress T , electric potential ϕ (quasistatic approximation), and dielectric displacement D . The model is generally restricted to harmonic time-dependence and to the one-dimensional case, that is, all considered quantities are assumed to show space-dependence in only one direction (direction of sound propagation, thickness direction of the layers). Furthermore, in some situations, the displacement of the sound wave may be restricted to this direction as well (longitudinal waves only). This treatment is justified here, because the piezoceramic plate transducers used essentially permit electrical excitation of longitudinal waves only. As a consequence of this restriction to a single displacement direction, all material constants, which are tensor quantities in the general case, are reduced to scalars, whereby the medium (layer) under consideration is described by its relevant elastic stiffness constant c , piezoelectric constant e , and dielectric constant ϵ relevant for the regarded extensional mode. "Relevant" quantity in this context means the parameter values valid for the regarded wave.

The electrodes of the piezoelectric layer have to be treated as separate films, but may be regarded as massless if they are sufficiently thin. On the outer free surfaces of the total sandwich arrangement, stress must be zero and dielectric displacement is assumed to be zero. This general free surface boundary condition establishes an expression for the electrical admittance Y between the electrodes. The explicit result for Y as a function of angular frequency ω and electrode area A was given by Nowotny and Benes [43].

Viscoelastic and dielectric losses can be expressed by an acoustic quality factor Q and a dielectric loss angle d , that form the imaginary parts of the material constants c and ϵ , respectively. (Piezoelectric losses can be neglected in most practical cases.) Other loss mechanisms relevant for acoustic particle filters, like sound attenuation due to bubbles or dispersed particles, losses due to divergence of the sound beam, etc., can be accounted for in a global way by using reduced Q -values (effective quality factors) for each material layer, as described by Gröschl [42].

For a given multilayer resonator, typically comprising an active piezoelectric layer followed by several passive solid and/or liquid layers, in a first step, the transfer matrix model allows calculation of the electrical admittance (real and imaginary parts) between the electrodes of the active layer as a function of frequency. In a second step, the spatial progress of the primary electroacoustic field quantities u , T , ϕ , D within each layer, can be determined for a fixed frequency and a given voltage amplitude applied across the electrodes. From the primary field quantities the spatial progress of other quantities of interest, like

displacement velocity or acoustic energy density in the considered layer, can be derived easily [42]. Practical application of the model requires knowledge of all material parameters involved, these are for each layer: thickness d , mass density ρ , elastic stiffness constant c , effective quality factor Q_{eff} . Furthermore, if the considered layer is located between electrodes: dielectric constant ϵ and dielectric loss angle d ; and, in addition for piezoelectric layers: piezoelectric constant e (or electromechanical coupling factor). The parameter values used for the calculations presented in this work are listed in [33]. For the mathematical expressions used see Gröschl [42] and Nowotny [43].

Electrical admittance

Fig.3 shows the electrical admittance spectrum (absolute values) of the multilayer resonator section of the h-shape separator (see Fig.5) with water filling, measured with a specialised computer-controlled electrical admittance measurement system [46], compared to the values predicted by the transfer matrix model described in the previous section.

Two characteristic frequencies are marked: f_1 coinciding with the fundamental resonance frequency of the piezoceramic (this coincidence is a consequence of chosen dimensions of layers A and C) and which is electrically strongest pronounced, and f_2 , which is electrically much weaker pronounced and nonetheless will be identified as one of the frequencies of optimum filter performance (see below).

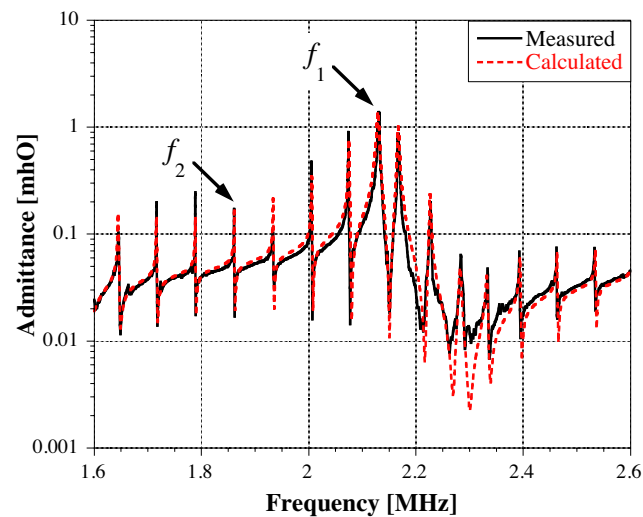


Fig.3. Measured and calculated admittance versus frequency curves of the resonator filled with water (PZT-ceramic/glass/water/glass composite resonator).

Spatial dependences in axial direction

Fig.4 shows the calculated spatial progress of displacement velocity amplitude and stored acoustic energy density, respectively, along the axial direction within the layers A, B, C, D, E of the h-shape separator (see also Fig.2).

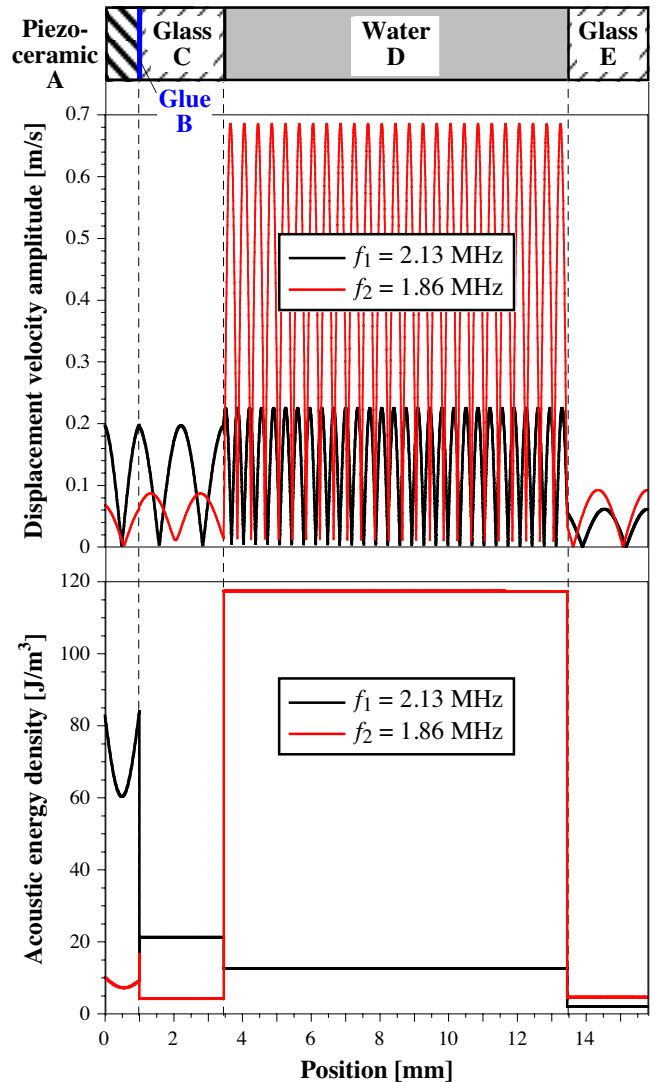


Fig.4. Spatial course of acoustic displacement velocity amplitude and acoustic energy density in axial direction of the PZT-ceramic/glass/water/glass composite resonator.

The graphs represent the results for the two selected frequencies f_1 and f_2 (see Fig.3). Surprisingly enough, the maximal displacement velocity amplitudes as well as the maximal energy densities in the water layer do not appear at the electrically strongly pronounced piezoceramic's fundamental resonance f_1 but at the electrically much less pronounced composite system's resonance f_2 . With respect to filtration efficiency, for a given electric energy supplied, maximum stored acoustic energy in the water (liquid) layer is desired. Thus, optimum system performance is achieved at resonance frequencies not coinciding with eigenfrequencies of the piezoceramic (layer A) or the ceramicglass composite transducer (empty chamber, layers A, B, C), as already shown by Gröschl for ultrasonic filters based on the ultrasonically enhanced sedimentation principle [42]. For the calculations shown in Fig.6, an electric driving power of $4 W_{rms}$

has been assumed, according to the experimental conditions applied.

Spatial amplitude distribution in lateral direction

The very good agreement of the electrical admittance spectra obtained from the one-dimensional model with that obtained from the measurements of the actual composite resonator in Figs.3 indicates that couplings to lateral modes are only spuriously affecting the electrical behaviour.

Ultrasonic h-shape separator

In Fig.5 the scheme of the h-shape resonator is introduced. In contrast to the ultrasonically enhanced sedimentation based separators, the h-shape resonator utilizes the acoustic radiation forces directly for separating the liquid flow lines into the cleaned outlet from the particle traces into the particle enriched outlet. Therefore the direct ultrasonic separation concept of the h-resonator is not relying on gravity.

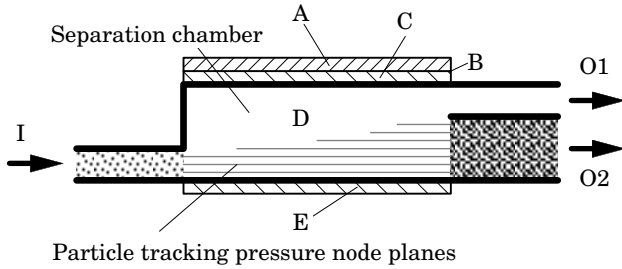


Fig.5. Scheme of the h-shape separator. On the left side the suspension is fed into inlet I of the separator, on the right side the upper outlet O1 is for the separated clean liquid and the lower outlet O2 for the particle enriched suspension.

Flow and force field simulation

Calculation of the particle traces was performed in two steps. First the velocity and the streamlines of the fluid was determined by solving the Navier-Stokes equations with the software Package FLUENT® (see Fig.6).

Input data are a) the h-separator geometry (Fig.5) analytically given by the graphic boundary condition defining software GAMBIT®, b) the material data mass density and viscosity of the fluid, the volume flow rate V_1 in the inlet I and the output volume flow ratio V_{O1} / V_{O2} .

In the second step Newton's equation of motion

$$\vec{F}_A(\vec{r}_p) + \vec{F}_{St}(\vec{r}_p) + \vec{F}_g - \vec{F}_b = \frac{4\pi a^3}{3} \rho_p \frac{d^2}{dt^2} \vec{r}_p \quad (2)$$

for the suspended particles were solved to obtain the particle traces $\vec{r}_p(t)$, whereby $F_A(\vec{r}_p)$ is the primary

acoustic radiation force in axial (vertical) direction given by Eq. (1), $F_{St}(\vec{r}_p)$ is the Stokes force

$$\vec{F}_{St}(\vec{r}_p) = -6\pi\eta a(\vec{v}_p - \vec{v}_f), \quad (3)$$

η the viscosity of the liquid, a the particle radius, \vec{v}_p and \vec{v}_f are the velocities of the particle and the fluid, respectively, F_g is the gravity and F_b is the buoyancy force and ρ_p the mass density of the particle. The results for the particle traces are given in Fig.6. The cross section of the separation chamber was 10 mm in height and 19 mm in width.

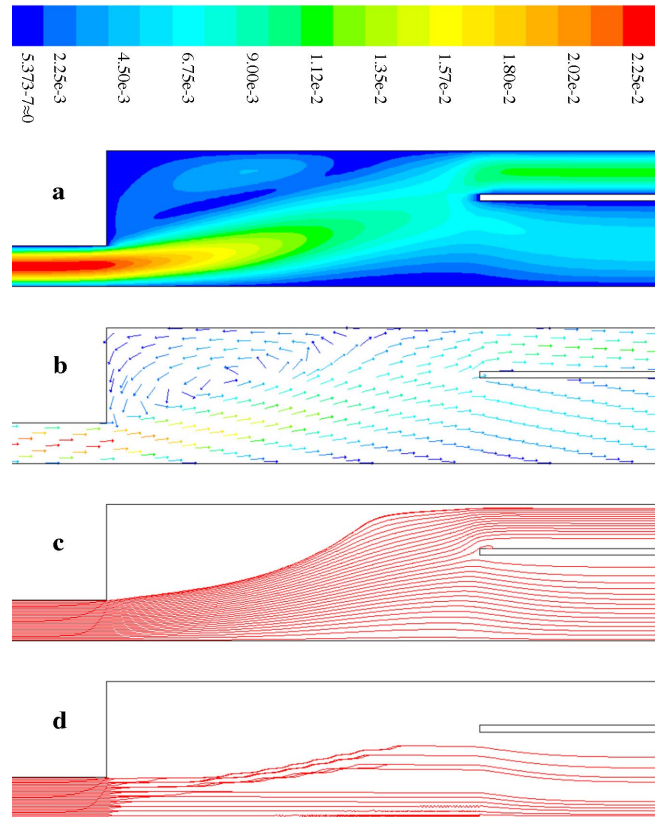


Fig.6. h-Separator performance simulation: a) color coded absolute value of the flow velocity in m/s; b) direction (unit flow velocity vector); c) streamlines of the liquid flow; and d) particle traces. Polystyrene particles $a = 20 \mu\text{m}$; $V_1 = 3 \text{ L/h}$ (liter per hour); $V_{O1} / V_{O2} = 1$.

h-shape separator sample experiments

Fig.7 shows a photograph of the h-shape resonator used in the experiments.

In Fig.8 a typical frame picture of the DV (digital video) movie documentation of the performance of the h-shape separator is shown for polystyrene particles with radius $a = 20 \mu\text{m}$, particle concentration $C_I = 0.314 \%$, $f = 1.96 \text{ MHz}$, $P = 3 \text{ Wrms}$, $V_{O1} / V_{O2} = 1$, $V_1 = 2.75 \text{ L/d}$ (= critical input flow rate).

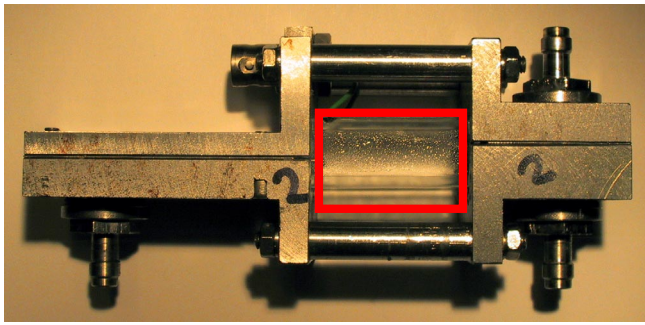


Fig.7. Photograph of the h-shape separator. On the left side the input silicone tube for the suspension and on the right side the two outlet silicone tubes for the separated clean water and for the particle enriched suspension, respectively, can be seen. The black cable on top connects the electrodes of the PZT plate with the drive electronics.

The chosen concentration of the particles was the limit value where the assumption of the simulation of single particle versus acoustic field interaction was still justified. As further sample suspensions yeast cell cultures of various concentrations have been investigated. The h-separator has also been tested successfully with spirulina platensis algae suspensions under microgravity conditions in the recent ESA 29th zero g parabolic flight campaign within the frame of the ESA Melissa project [47].



Fig.8. Frame picture of the h-separator performance. The output flow ratio $V_{O1} / V_{O2} = 1$ corresponds to a respective ratio of the mean flow velocities of 2:1, because the cross sectional area of output O1 was chosen half the value of the cross sectional area of output O2.

Performance comparison

For the comparison of different ultrasonic separator concepts the ratio of volume flow [L/h] over the needed true electrical power [W] is defined as the performance figure of the separator concept under investigation

$$S_p = V / P_{rms} = [L / Wh] \quad (4)$$

The presently most used term "separation capacity" in liters per day is, of course, directly dependent on the size of the separator and the electrical power

supply, while the reduced quantity suggested as performance figure allows a comparison of the concepts itself independent on the upscaling grade. In addition, of course, the meaning of the figure is dependent on the sample (application), especially on its concentration.

Summary

The most recent progress in the ultrasonic separation technology is based on the availability of powerful new tools described in this paper: The rigorous one-dimensional model of the layered resonator, a highly specialized electrical admittance measurement system, a LDV vibration amplitude distribution measurement system, highly specialized drive electronics with automatic resonance frequency and true input power control, and the mathematical model combining for the first time the hydrodynamic flow and the acoustic field influence on the traces of the suspended particles. For comparing the separation performance of different resonator concepts the acoustic separation performance figure has been introduced. For higher particle concentrations, the ultrasonically enhanced sedimentation is the state of the art bio-cell filter concept.[33] For very dilute suspension, the h-separator exhibits a significantly higher separation performance figure and has been proven to work also under zero g conditions.

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