NUMERICAL CALCULATION OF PRESSURE FIELDS IN SONOCHEMICAL REACTORS – LINEAR EFFECTS IN HOMOGENEOUS PHASE

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Abstract

During the last 50 years sonochemistry was under active investigation. Nevertheless, there is still a lack of a sound theoretical basis for the design of sonochemical reactors. Furthermore, sonochemical reactions are not understood in detail on a molecular level. In order to calculate the yield of chemical reactions in reactors of different shapes one needs to know the number of cavitation bubbles in the reactor. These bubbles are generated by oscillating pressures in the liquid. Therefore, as a first step of the design of sonochemical reactors pressure fields in homogeneous media in reactors of different geometric shapes are calculated..

Keywords: sonochemistry, reactor design, pressure fields.

Introduction

Sonochemical effects have been known for about fifty years. During the last decade interest in sonochemistry has increased rapidly. A broad variety of chemical reactions may be influenced by sound (ABRAMOV 1994), (DUKE 1988), (LUCHE 1993), (SENAPATI 1991) and (SUSLIK 1989). Ultrasound enhances the rates of many chemical reactions. In homogeneous phase ultrasound generates cavitation bubbles which leads to sonochemical reactions. When the liquid is irradiated with ultrasound, it causes a series of compression and rarefaction cycles which create areas of high and low local pressures. Power ultrasound produces cavitation bubbles. It has been calculated that pressures of thousands of atmospheres and temperatures of thousands of degrees are generated on collapse of these bubbles. For example, sonicated water shows sonoluminescence. The details of how sound influences chemical reactions are still not clarified. As has been stated by BLAKE et al. (1994) and LEIGHTON (1994) application of detailed mathematical techniques may expand our understanding of the 'why' and 'how' of sonochemical effects.

Although sonochemical reactions could be explained for some cases, the behaviour of a given system has been either impossible to predict or in only a few cases hard to establish. One reason is that the research and development for sonochemical reactors are mostly based on empirical reaction data.

Our ultimate goal is to develop a model which is capable of predicting at least approximately the yield of sonochemical reactions in reactors of different designs which are in use. As a first step in this direction pressure fields in homogeneous media in reactors of different geometric shape were investigated.

In this paper we take only linear effects into account. In subsequent papers we will investigate nonlinear effects and two-phase media.The final result of the investigations should be a method for the rational design and scale-up of the chemical reactors. An understanding of chemical mechanisms of sonochemical reactions probably requires quantum chemical calculations.

Theory

Sound waves, in our case, are generated by the vibration of a transducer in contact with the liquid medium. The characteristics of the transducer determine the frequency and the direction of the generated sound field.

To calculate the sound field of an arbitrarily shaped body we begin with the determination of the time-dependent pressure field of a point source. In this paper nonlinear wave propagation is neglected. Let us consider a sphere with an average radius, a, which is expanding and contracting so that the radial velocity of its surface everywhere is the same function of time $U(t)$. The rate of fluid flow away from the surface of the sphere is

$$S(t) = 4\pi a^2 U(t) \ . \tag{1}$$

The wave equation for the pressure, p, of the fluid in spherical coordinates is written as

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial p}{\partial r}\right) = \frac{1}{c^2}\frac{\partial^2 p}{\partial t^2} \ . \tag{2}$$

c is the velocity of the acoustic wave and r the distance from the center of the sphere.

By using the first-order equation of (radial) motion in a fluid

$$\frac{\partial p}{\partial r} = -\rho\frac{\partial u_r}{\partial t} \ , \tag{3}$$

where u_r equals the radial velocity of the fluid particles and ρ the density of the fluid and the assumption of an arbitrary outgoing wave

$$p = \frac{P(r - ct)}{r} \ , \tag{4}$$

we derive the equation (MORSE and INGARD, 1968)

$$\frac{1}{r}\frac{\partial P(r-ct)}{\partial r} - \frac{P(r-ct)}{r^2} = -\frac{\rho}{4\pi a^2}\frac{\partial S(t)}{\partial t} \quad \text{at} \quad r = a \tag{5}$$

to calculate the shape of the pressure wave. $P(r-ct)$ is a function which describes the shape of an outgoing wave.

If the radius, a, of the sphere tends to zero (the sphere becomes a point source) the term P/r is much greater than $\partial P/\partial r$. Therefore, for the shape of the pressure wave, we get the relationship

$$P \approx \frac{\rho}{4\pi}\frac{dS}{dt} \quad \text{at} \quad r = a\ . \tag{6}$$

At time t and a distance r from the center of the point source the pressure wave fulfils the relationship

$$p \approx \frac{\rho}{4\pi r} \cdot S'\left(t - \frac{r}{c}\right)\ , \tag{7}$$

where $S'(z) = \frac{dS(z)}{dz}$.

Suppose a spatial field of distributed spheres with vanishing radius, a, which all perform radial simple-harmonic movements, so that $S(t)$ becomes $S_{j,\omega} \cdot \exp(-i\omega t)$ (j = number of source). In this case, $p(\mathbf{r})$ is the result of a superposition of all sources. If the source distribution assumes the shape of a surface and the quantity of sources increases to infinity, which is a possible model to describe a vibrating body, the sum of the point sources changes to an integral equation.

As the coverage of the surface with point sources in this case becomes continuous, the velocity perpendicular to the normal vector of the body's surface vanishes.

Therefore the harmonic fluid motion $S(t)$ can be expressed as $U_0(\mathbf{r}')\exp(-i\omega t)$, where U_0 is the velocity amplitude of the vibrating body and $\mathbf{r}'$ a point on its surface.

As a result, we get the first equation for the calculation of the pressure field of any harmonic vibrating body

$$p(\mathbf{r}, t) = \frac{i\omega\rho}{4\pi} \cdot \iint_{S'} U_0(\mathbf{r}') \cdot \frac{e^{i(k|\mathbf{r}'-\mathbf{r}|-\omega t)}}{|\mathbf{r}'-\mathbf{r}|} dS'\ , \tag{8}$$

where dS' equals the body's surface.

At this place, we have to compare similar theories (LOCKWOOD and WILLETTE 1973), (STEPANISHEN, 1971) and (TJØTTA and TJØTTA, 1982) in which the last equation was derived in a different way. Referring to the majority of papers, the acoustic field of a planar transducer in an infinite rigid baffle was calculated by the following procedure :

Due to the influence of the reflected field a point source in front of an infinite rigid plane with a distance, d, can be described by two sources with distance $2d$. By calculating the pressure field of both point sources one gets the same field as with the model of one point source in front of the plane (JUNGER and FEIT, 1986). This procedure is analogous to the method of image charges in electrodynamics. Therefore, the field of this point source in contact with that plane is approximated by two sources with vanishing distance to each other. The result is a pressure field twice as large as one with a single source.

With this model an integral nearly identical with *Eq.* (8) is obtained. The difference is that the pressure field calculated by this formula gives pressure values twice as large as with the aid of *Eq.* (8). As will be shown in *Eq.* (14) the reflected sound field has to be added to *Eq.* (8). In our work the application of image sources is not possible because the assumption of an infinite (reactor) wall is not fulfilled.

Therefore, we have to calculate the pressure field of an ultrasonic source by *Eq.* (8) and then add the reflected field of the transducer surface and the finite rigid baffle to the primary field.

Before we derive formulae to add reflected fields to the original field, the term in the double integral of *Eq.* (8) has to be discussed.

The fraction $g(\mathbf{r}', \mathbf{r}) = \exp(-ik|\mathbf{r}' - \mathbf{r}|)/|\mathbf{r}' - \mathbf{r}|$ is a Green's function and obeys the equation

$$\Delta g(\mathbf{r}', \mathbf{r}) + k^2 g(\mathbf{r}', \mathbf{r}) = -\delta(\mathbf{r}' - \mathbf{r}) , \tag{9}$$

where $\delta(\mathbf{r}' - \mathbf{r})$ is the delta function in three dimensions. By this definition, $g(\mathbf{r}', \mathbf{r})$ is a special solution of (8) for a source at the point $\mathbf{r}'$ in an unbounded medium. The most general solution of *Eq.* (9) for a bounded medium is $g(\mathbf{r}', \mathbf{r})$ plus a solution of the homogeneous equation

$$\Delta g'(\mathbf{r}) + k^2 g'(\mathbf{r}) = 0 . \tag{10}$$

So, by the assumption of an acoustic field produced by distributed sources in a bounded medium we have to solve an equation analogous to *Eq.* (9)

$$\Delta p(\mathbf{r}) + k^2 p(\mathbf{r}) = -f(\mathbf{r}) . \tag{11}$$

By multiplying *Eq.* (9) with $p(\mathbf{r}', \mathbf{r})$ and subtracting $g^*(\mathbf{r}', \mathbf{r}) = g(\mathbf{r}', \mathbf{r}) + g'(\mathbf{r}', \mathbf{r})$ multiplied by *Eq.* (11) we obtain

$$g^*(\mathbf{r}', \mathbf{r}) \cdot \Delta p(\mathbf{r}) - p(\mathbf{r}) \cdot \Delta g^*(\mathbf{r}', \mathbf{r}) =$$

$$= p(\mathbf{r}) \cdot \delta(\mathbf{r}) - g^*(\mathbf{r}) \cdot f(\mathbf{r}) \; . \tag{12}$$

Now this equation will be integrated over the whole volume occupied by the fluid. Because of the symmetry of g^*, $[g^*(\mathbf{r}', \mathbf{r}) = g^*(\mathbf{r}, \mathbf{r}')]$, and $\delta(\mathbf{r}' - \mathbf{r})$ and the fact that

$$g^*(\mathbf{r}', \mathbf{r}) \cdot \Delta p(\mathbf{r}) - p(\mathbf{r}) \cdot \Delta g^*(\mathbf{r}', \mathbf{r}) =$$

$$= \nabla[g^*(\mathbf{r}', \mathbf{r}) \cdot \nabla p(\mathbf{r}) - p(\mathbf{r}) \cdot \nabla g^*(\mathbf{r}', \mathbf{r})] \tag{13}$$

we derive a volume integral which is, due to the divergence theorem of Gauss, convertible into a surface integral. This leads us to the governing equation for the calculation of the pressure field in any ultrasonic reactor employing a homogeneous phase :

$$p(\mathbf{r}, t) = \iiint_{V'} f(\mathbf{r}) g^*(\mathbf{r}', \mathbf{r}) dV'$$

$$+ \iint_{S'} \mathbf{n}_{S'}(\mathbf{r}') \cdot [g^*(\mathbf{r}', \mathbf{r}) \cdot \nabla' p(\mathbf{r}) - p(\mathbf{r}) \cdot \nabla' g^*(\mathbf{r}', \mathbf{r})] dS' \; . \tag{14}$$

$\mathbf{n}_{S'}(\mathbf{r}')$ is the unit normal vector of the surface over which integration has to be done.

This integral equation enables us to calculate the acoustic pressure field within and on the surface forming the boundary of the medium. If $g^*(\mathbf{r}', \mathbf{r})$ is the field at point $\mathbf{r}$ due to a point source at $\mathbf{r}'$, the resultant field of distributed sources in the volume V' is calculated by the summation of the fields of all the differential sources $f(\mathbf{r})dV'$ (first term) plus the reflected waves from the boundaries (second term). Thus, if there are no boundaries at all, the second integral will vanish and $p(\mathbf{r}, t)$ is a result of the volume integral which in our case is identical with *Eq.* (8).

The foregoing equations depend on a linear and undamped wave propagation. To embed damping effects we have to consider the energy loss of the wave depending on viscosity and thermal conductivity (the classical absorption). For a propagating wave, the intensity I at a distance x from the source is given by BATHIA (1967) as

$$I = I_0 e^{-2 \cdot \alpha \cdot x} \; . \tag{15}$$

α = classical absorption coefficient.

This relationship will be introduced into *Eqs.* (8) and (14) by using a complex wave number

$$\overline{k} = k + i\alpha \ . \tag{16}$$

For liquids or for very high frequencies in gases one has to solve the following equations to calculate $\overline{k}$ as outlined by MORSE and INGARD (1968)

$$\overline{k}^2 = \frac{i\omega^2}{2\Omega c^2}\left(\frac{1 - i(Y + \gamma\Omega \mp Q)}{1 - i\gamma Y}\right) ,$$

$$\Omega = \frac{K\omega}{\rho c^2 C_p} ,$$

$$Y = \left(\eta + \frac{4}{3}\mu\right)\frac{\omega}{\rho c^2} ,$$

$$Q^2 = (1 + i(\gamma\Omega - Y))^2 - 4i(\gamma - 1)\Omega \ . \tag{17}$$

The upper minus sign in the first formula corresponds to an adiabatic, and the lower plus sign to an isothermal wave propagation. The propagation mode depends mainly on the circle frequency ω and the thermal conductivity of the fluid. In water and with frequencies higher than $5 \cdot 10^4$ [1/s], like in our case, we can assume an adiabatic wave propagation.

In this work, the approach to calculate the pressure field in a reactor with an arbitrary shape is as follows:

First of all the pressure field generated by the modelled transducer is calculated numerically in the reactor fluid and on the boundaries with the aid of *Eqs.* (8) and (17). As the values of the primary field at the walls are provided by this sum, we can calculate them for the total field by taking the following assumptions into consideration:

- For a rigid wall a doubling of the pressure values due to reflection at the wall is supposed.
- The free surface of the fluid nearly fulfils the condition of an ideal 'pressure-release' boundary. As one is not able to determine the derivative of the total acoustic field at this surface until the reflected field is known, we calculate the reflected field of a rigid wall and then convert it by a phase-shift of π (reversed sign of $p_R(\mathbf{r}, t)$) with the result that the total field vanishes at the liquid surface.

In this paper only first-order reflections are used to determine the entire acoustic field.

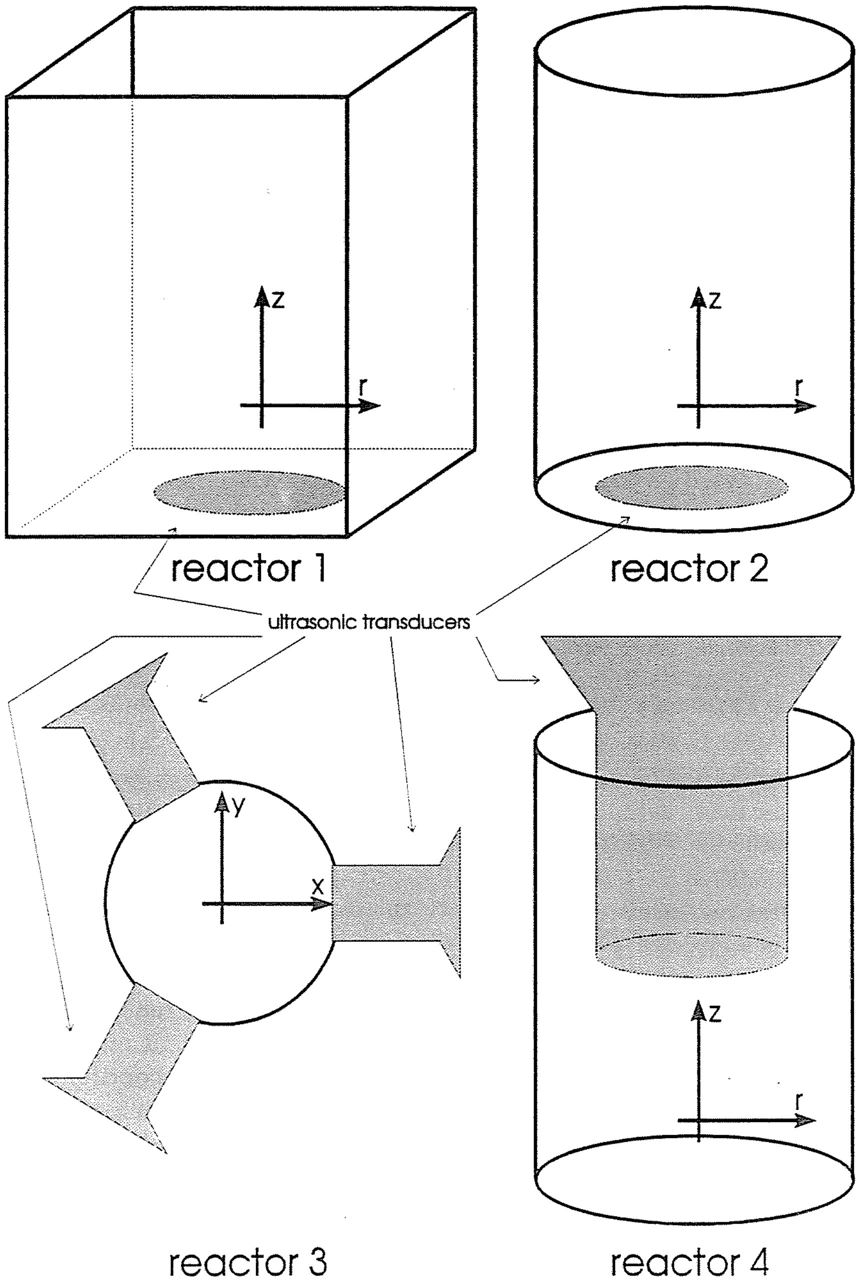

Fig. 1. Different types of modelled sonochemical reactors

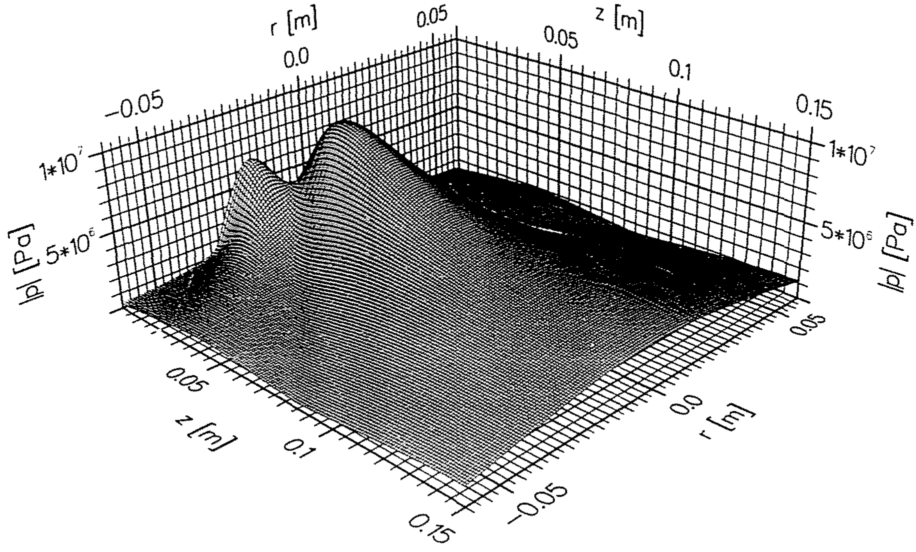

Fig. 2. Acoustic field of an ultrasonic transducer without any boundary conditions

Types of Sonochemical Reactors Modelled

Four different types of ultrasonic reactors (see *Fig. 1*) were analysed and their pressure fields calculated as a function of the frequency and amplitude of the transducers' vibration. All the transducers described have the same diameter of 0.06 m.

The first two reactors have nearly the same geometry: the ultrasonic transducer is a circular plate which transmits acoustic waves from the bottom of the two reaction vessels. The first vessel has a quadratic shape with a length of a side of 0.12 m and the vessel of the second reactor is a circular tube with a diameter of 0.12 m. Both vessels are 0.15 m high. Reactor 3 is a circular tube with the same diameter as the second one. The acoustic waves are transmitted by three ultrasonic horns which are placed in an equilateral triangle in the same plane around the tube. The centers of the three transducer surfaces have their positions at the coordinates ($r = 0.06$ m, $\varphi = 0$, $z = 0$) and ($r = 0.06$ m, $\varphi = \pm 2\pi/3$, $z = 0$).

For this reactor the pressure values were calculated for two planes:

One is the usual $r - z$-plane in cylindrical coordinates as used for the other reactor types. The other is the plane rectangular to the $r - z$-plane and the axes are described as the x- and y-axis like in the normal Cartesian sytem. Pressure values are calculated up to a height of 0.15 m.

The last reactor which is examined consists of a cylindrical tube with the same dimensions as reactor 2. An ultrasonic horn emits the acoustic waves downwards to the bottom from a height of 0.075 m.

All the calculations were carried out for a transducer frequency of 50 kHz and an amplitude of 10^{-5} m.

Details of the Program Implementation

The analytical calculation of *Eqs.* (8) and (14) is possible only in a few special cases with several simplifying assumptions (HARRIS, 1981), (SEYBERT et al. 1985), and (TJØTTA and TJØTTA, 1982). As the modelling of pressure fields of several ultrasonic reactors is necessary and in most of the cases the shape of the transducer or the wall does not have a special symmetry, a numerical calculation of the integrals was chosen.

The surface of the walls and the transducer was subdivided into pieces of equal size. The integrals have been calculated by a simple Riemann integration. Already with a subdivision of the surface into 50 × 50 pieces we obtain solutions of sufficient accuracy.

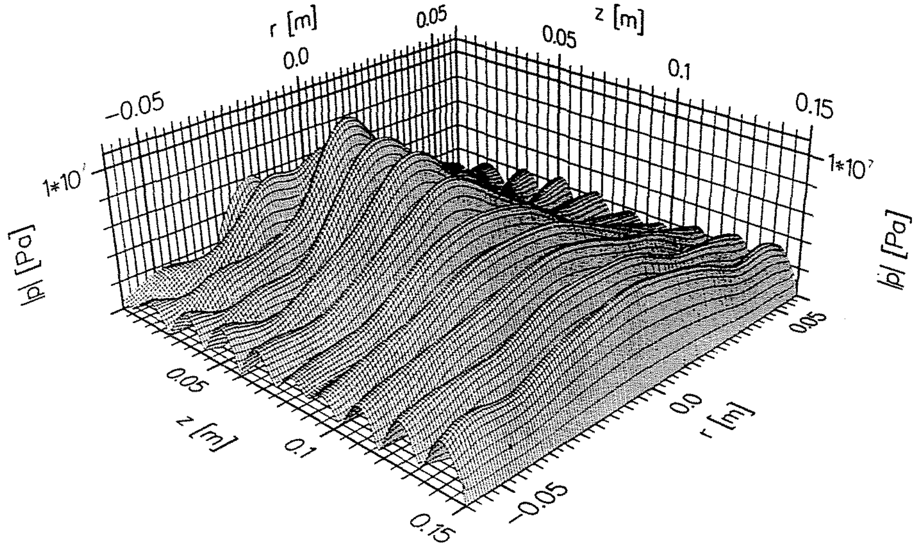

Fig. 3. Acoustic field of reactor type 1

To carry out the calculations a Convex C3840 and an IBM Workstation RS/6000 Model 3bt were used. With a quantity of 100 × 100 pressure values and a surface subdivision into 50×50 pieces the computing time needed on the IBM was three hours for the reactor of type 2. Considerably longer calculation times were needed for reactors which do not show e.g. cylindrical symmetry. One reason is that the pressure values at the boundaries must be calculated additionally to the primary field of the transducer which is only determined for one plane. The other reason is that one has to subdivide the surface into 300×300 pieces to obtain a sufficiently accurate solution. For these reactors the calculation time can rise up to one week for the Workstation.

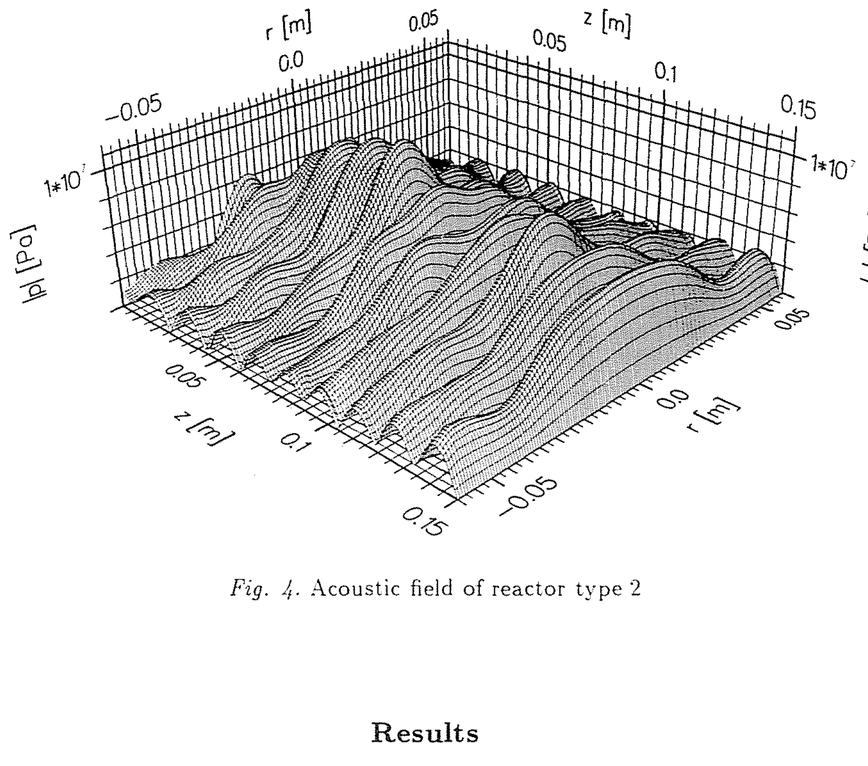

Fig. 4. Acoustic field of reactor type 2

Results

In this section some results of the calculations are presented for the four different types of sonochemical reactors. To make the difference clear between the acoustic field of a transducer in an infinite planar baffle and the values found within the reactor vessel, the undisturbed field of a transducer with a diameter of 0.06 m is shown in *Fig. 2*. It vibrates with a frequency of 50 kHz and an amplitude of 10^{-5} m.

In this figure, like in the following ones, the absolute pressure $|p|$ as a function of the position is presented. Therefore, only the pressure

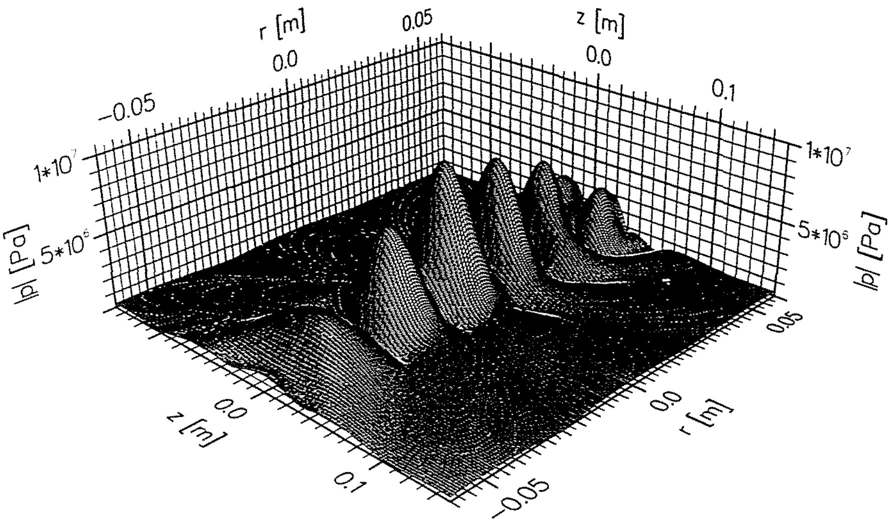

Fig. 5. Acoustic field of reactor type 3 in the $r - z$-plane where $z \in [-0.15 \text{ m}; 0.15 \text{ m}]$

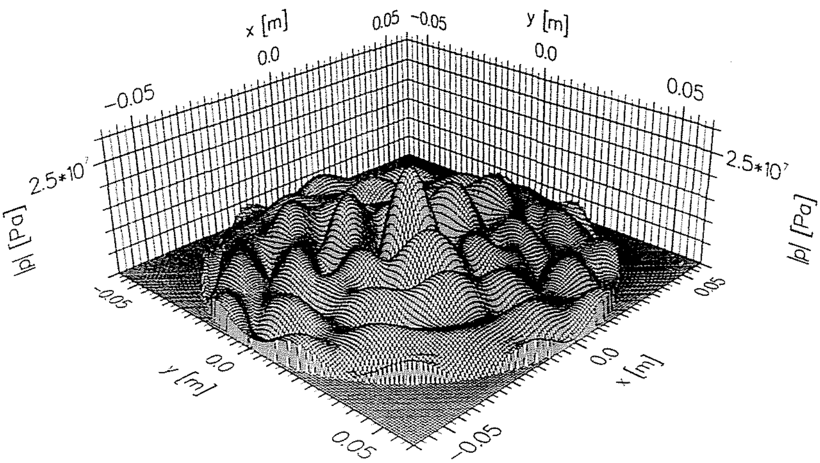

Fig. 6. Acoustic field of reactor type 4 in the $x - y$-plane where $z = 0$

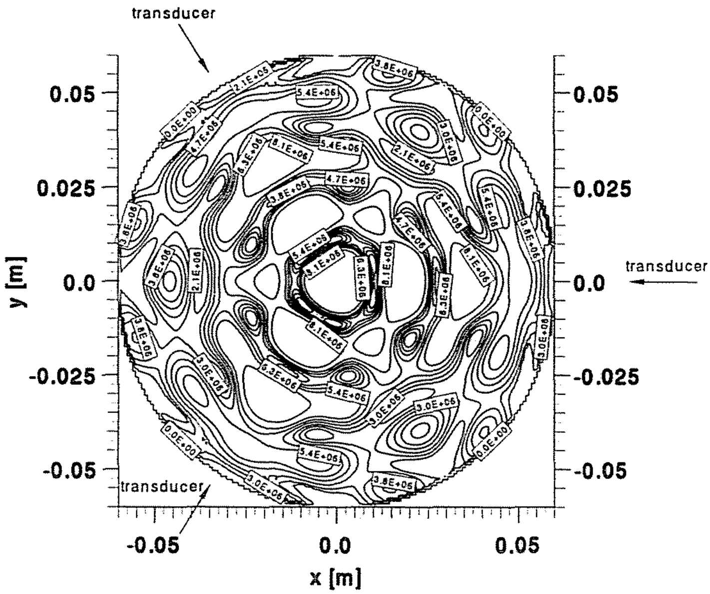

Fig. 7. Contour plot of the pressure field belonging to *Fig. 6.* The numbers in boxes represent the pressure amplitudes.

amplitudes but not the spatial phases of the pressure fields are represented. Although it is possible to determine the local phase relations of the acoustic field, for the intended application of these results it is not necessary to know them. *Fig. 3* shows the acoustic field within reactor 1. It is easy to recognize by comparing *Fig. 2* and *Fig. 3* that the number of pressure maxima has nearly doubled. This effect may be compared to the pressure field in a Kundt's tube.

The acoustic field of reactor 2 is shown in *Fig. 4*. One can recognize that this field has a similar shape to reactor 1. Different are the higher pressure values on the z-axis in the center of the reaction vessel. This is a result of the interference of the reflected waves due to the curvature of the reactor wall in contrast to the plane walls of the reactor type 1 where the amplitude of the reflected wave decreases with increasing distance from the wall.

In *Fig. 5* the pressure field on the $r-z$-plane is presented up to a height of 0.15 m in both directions. The shape of the field differs significantly from the previous one. In the vertical vicinity of the three transducers considerable pressure amplitudes may be found only. Because of the interference of the primary fields due to the transducers' position the highest values are in the middle at the origin of the system of coordinates. The maximum values

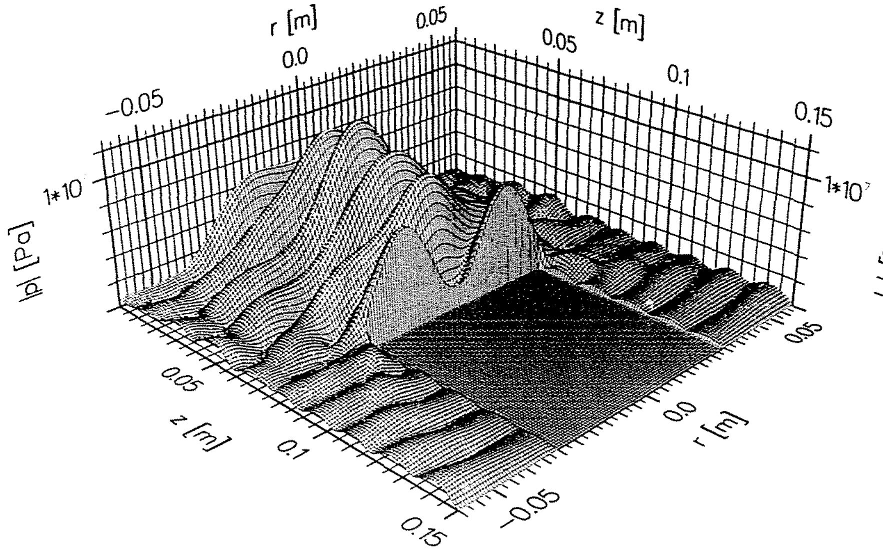

Fig. 8. Acoustic field of reactor type 4. The area where $z > 0.075$ m and $x < 0.03$ m belongs to the solid of the acoustic horn.

are more than doubled compared to the data of the first two reactor types. The acoustic field in the $x-y$-plane is represented in *Fig. 6.* Like in *Fig. 5*, the highest pressure values are in the vicinity of the reactor center. It has a rotational symmetry of the angle $2\pi/3$ like the positions of the transducers.

To give a better overview of the spatial distribution of this field a contour plot is shown in *Fig. 7.* The direction of oscillation and the position of the transducers are marked by little arrows. The numbers in small boxes represent the pressure amplitude in Pascal.

In the last figure (8) the acoustic field of reactor 4 is shown. The pressure amplitudes in the plane of the ultrasonic horn are not defined and therefore these values are set to zero. One recognizes that the amplitudes for z greater than 0.075 m are small compared to the rest of the area. This is due to a shielding effect of the transducer.

This effect has been approximated by a simple exclusion of those reflecting surface elements which have not a direct connection to the calculated point (simple shadow construction).

Conclusions

In this paper an algorithm and the first results are presented to compute the acoustic field at any point in sonochemical reactors of any geometrical shape. Calculations are based on the neglection of viscosity effects and nonlinear wave propagation. Furthermore, only first-order reflections are considered. In subsequent studies these points will be taken into consideration and their effects will be examined.

A first step is made to predict the probability of the emergence of cavitation bubbles and so predict at least approximately the spatial distribution of bubble clouds. If these predictions are possible one can determine which type of reaction will occur and where. One will then be able to optimize the type and the shape of a sonochemical reactor as a function of the chemical reaction executed.

Symbols

a	radius of oscillating sphere
c	velocity of acoustic waves in the fluid
$g(\mathbf{r}', \mathbf{r})$	Green's function for a source at the point $\mathbf{r}'$ in an unbounded medium
$g^*(\mathbf{r}', \mathbf{r})$	Green's function for a source at the point $\mathbf{r}'$ in consideration of boundary conditions
$f(\mathbf{r}')$	spatial factor of the pressure wave from a simple harmonic differential source
k	wave number of the acoustic wave
$\overline{k}$	complex wave number
p	acoustic pressure
$p_R(\mathbf{r}, t)$	pressure field due to a reflection
r	radial coordinate
t	time parameter
u_r	radial fluid velocity
C_p	heat capacity at constant pressure
C_v	heat capacity at constant volume
P	function which describes the shape of an outgoing wave
$S(t)$	time dependent radial fluid flow
S'	surface of the transducer or the boundary
$U(t)$	radial velocity of the sphere's surface
γ	ratio of specific heats ($= C_p/C_v$)
η	coefficient of bulk viscosity
μ	coefficient of viscosity

ρ	density of the fluid
ω	circle frequency of the harmonic vibrating transducer
$\mathbf{n}_{S'}(\mathbf{r}')$	unit normal vector of the transducer's or boundary's surface
$\mathbf{r}$	coordinate vector
$\mathbf{r}'$	coordinate vector on a vibrating or reflecting surface
∇	Nabla operator
Δ	Laplacian operator
Indices	
0	amplitude
S'	belonging to the transducer's or boundary's surface

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