ICSV14
Cairns • Australia
9-12 July, 2007

14th International Congress on Sound & Vibration

ULTRASOUND PIEZOCERAMIC TRANSDUCER FOR UNDERWATER ACOUSTICS IN SONOCHEMISTRY

Irinela Chilibon[1], Mihaela Mocioi[2] and CarmencitaMateescu[3]

[1]National Institute of Research and Development for Optoelectronics, INOE- 2000,
PO Box MG-5, 77125, Bucharest-Magurele, Romania, qilib@yahoo.com
[2]University ”Politehnica” of Bucharest, Bucharest, Romania
[3]National Institute for Materials Physics, PO Box MG-7, Bucharest-Magurele, Romania

Abstract

The effect of ultrasound fields on reactive crystallization nucleation could produce homogeneous nucleation and improve the process reactivity into chemical solutions. Organic molecules or polymers can influence the crystallization of inorganic salts and may act as crystal growth inhibitors. It is not yet clear if the additive interacts with dissolved components or with the developing phase. Crystallization process may be controlled by various additives. Also, ultrasounds may also affect the process. Complex phenomena take place in liquids when an ultrasonic field is applied. Ultrasounds yield cavitation bubbles which accumulate gases and vapors from the liquid phase until they become too large and collapse, producing shock waves. The temperature and pressure inside the cavitation bubbles and in their vicinity are huge and influence all chemical reactions taking place in the liquid phase. Also, ultrasounds induce different reactions into the chemical mixed solutions, function of ultrasound duration, frequency, temperature, and application manner of solutions (dripping or pouring). Therefore a small size ultrasound piezoceramic transducer is suitable to the sonochemistry applications. The generation is made by a transducer supplied by a signal generator. The ultrasound piezoceramic transducer generates ultrasound field which yields caviational effects such as: enhancement of gas bubbles, temperature increasing, mechanical effects, dispersion of solids in liquids, and reactivity increasing. The transducer converts electrical signal into mechanical vibrations, which are propagated into chemical solutions. The small diameter of transducer case allows get to straiten glass tubes, such as the chemical flask necks. The ultrasound piezoceramic transducer is suitable to generate ultrasound in the same time with the chemical processes, at 46 kHz resonance frequency. A transducer application in sonochemistry process is the calcium carbonate crystallization, combining the effects of organic additives and ultrasonic field. As final result is calcium carbonate powder with large amount of nanometer size crystals.

1. INTRODUCTION

A possible improvement of the crystallization process is to use power ultrasound, but this requires an appreciation of all the phenomena occurring during sonocrystallization. It has been observed that ultrasonic waves have an influence on the growth rate and the crystals size distribution during crystallization in saturated solutions [1]. For instance, the nucleation rate of the $CaSCO_4$ crystals has been found multiplied by 10 at ultrasound application.

The induction time of nucleation is strongly reduced in presence of ultrasonic waves for different systems. The growth of sugar crystals has been found to be faster under ultrasound compared with mechanical agitation. The crystal growth rate depends on the ultrasonic frequency and intensity. Several authors have noted this acceleration of the crystals growth under ultrasound for different solutes, but today the mechanism of the ultrasound action is not yet well understood [1]. It has been also found that ultrasound can reduce or modify agglomeration and improves the product handling. It is possible that the ultrasonic wave increases the probability of collision between the particles as in the primary nucleation.
So, one can say that the effects of ultrasound on crystallization are very divers, mostly positive, but that they are numerous and difficult to analyse separately.

The implosion of the cavitation bubbles plays an important role in the disintegration of the imersed solid particles.

Chemical and some mechanical effects are given when ultrasound irradiates liquid, and most of these effects are a result of the implosive collapse of cavitation bubbles and/or bubble-induced microstreaming. Ultrasound is applied to various purposes because of these beneficial effects. For example, ultrasound is used to enhance reaction rates, to clean glassware, and to form nanoparticles of metals and pharmaceutical products. Particularly in the crystallization processes, the term “sonocrystallization“ is defined as applying ultrasound to crystallization process, and the following ultrasonic beneficial effects have been reported. It was so far confirmed that ultrasound excels in controlling primary nucleation [2], modifying crystal size and crystal size distribution (CSD) [3], inducing nucleation and enhancing crystal growth rate [4]. Because of ultrasonic ability to control the crystal properties, the emphasis is placed on the application of ultrasound to crystallize active pharmaceutical ingredients. Moreover the following applications are suggested as the potential applications of sonocrystalization [5].

- Manipulation of crystal distribution by controlled nucleation
- Ultrasonic irradiation at modest supersaturation to initiate controlled nucleation

Manipulation of crystal distribution by controlled nucleation was carried out for dodecanedioic acid [6]. As concern as the other substances, however, ultrasound cannot always induce primary nucleation. Therefore a fundamental study for investigating ultrasonic nucleation phenomenon is required to controlling primary nucleation.

The acoustical demands, for a good efficiency, are conditioned by the necessity to realize a working regime at the resonance frequency of the cavitation bubble, and limited by some factors, such as: the transducer type, PZT material properties, the transducer manufacture mode, etc.

The ultrasonic piezoceramic sandwich transducer has three parts: one central and active vibration source (piezoceramic tores), and two end-metal masses (reflector and radiator). The active source of vibration is placed inside the transducer body making one sandwich (metal-piezoceramic) structure bonded by two end-metal masses.

2. PIEZOCERAMIC SANDWICH TRANSDUCER DESIGN

The active element of the device is a sandwich transducer, made of two piezoceramic tores stack with metal cylinders (Figure 1). This electromechanical transducer works on the basis of the inverse piezoelectric effect, converting electrical power into a mechanical displacement in the range of tens of microns. All device elements are fixed and pre-stressed by a stainless steel screw, which induces an initial polarization of the piezoceramic stack. The vibration amplitude of the sandwich radiator depends on PZT tores number, PZT material elasticity coefficients and the metal elasticity coefficients. Maximum peak to peak displacements of the transducer radiating face would be in order of tens microns, when operating at resonance frequency.

The design of a simple metal - piezoceramic - metal sandwich transducer ([7], and [8]) to resonate at a given fundamental frequency, $2\pi\omega$, in the plate thickness direction is based on the following equation:

$$\frac{\varpi l_c}{v_c} + \operatorname{arctg}\left[\frac{A_1\rho_1 v_1}{A_c\rho_c v_c}\operatorname{tg}\left(\frac{\varpi l_1}{v_1}\right)\right] + \operatorname{arctg}\left[\frac{A_2\rho_2 v_2}{A_c\rho_c v_c}\operatorname{tg}\left(\frac{\varpi l_2}{v_2}\right)\right] = \pi \quad (\text{or } 180^0) \qquad (1)$$

where:

l_1, l_2, l_c: lengths of 1st metal, 2nd metal, ceramic sections
v_1, v_2, v_c: sound velocities in 1st metal, 2nd metal, ceramic sections
A_1, A_2, A_c: cross sections of 1st metal, 2nd metal, ceramic sections
ρ_1, ρ_2, ρ_c: densities of 1st metal, 2nd metal, ceramic sections

If the metal plates are assumed to be identical and the ceramic placed symmetrically between them, the calculation of the transducer dimensions can be greatly simplified by approximation. A graphical representation of the general solution is shown as being a rapid and easy solution of the transducer equation (1), and one can determine the optimum component lengths of the sandwich transducer.

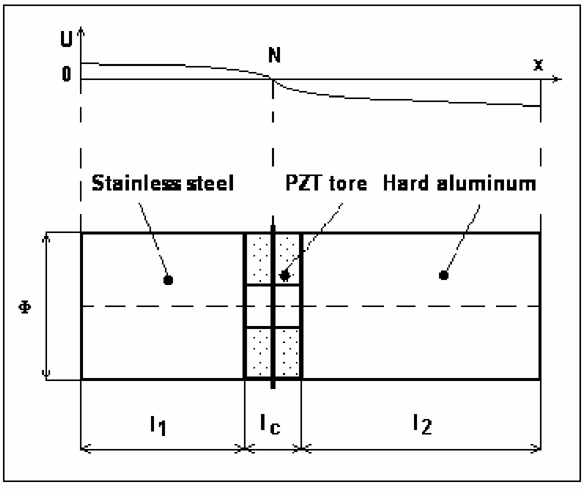

Figure 1 Simple sandwich piezoceramic transducer

In this case, the effect of the ceramic on the resonance frequency is almost entirely due to its compliance, and the effect of its mass is negligible. This is fairly obvious when one considers that the section at the center is subjected to high stress and strain levels and therefore contributes to the total potential energy, but experiences little vibratory motion and therefore contributes little to the total kinetic energy.

This conclusion may be proven by applying the equation to the case, in which the ceramic is replaced by a metal of the same type and cross-section as that used for the outer plates, and of length given by:

$$l^1 \approx l_c \frac{A_1 Y_1}{A_c Y_c} \tag{2}$$

then Y_1, Y_c and Y_c^E has to be defined the solid metal bar of length $\mathbf{2l_1 + l^1}$ has the same resonance frequency as the original sandwich. This result is derived by assuming that the θ phase angle across both ceramic and equivalent metal section (i.e. $\omega l_0/v_0$ and $\omega l^1/v_1$) are small enough for the approximation $\tan\theta = \theta$ to be used. This formula gives a section of the same total compliance as the ceramic. The sound velocity value of v_c is obtained from the relation:

$$v_c = \sqrt{\frac{Y_c^E}{\rho}} \tag{3}$$

If the sound velocity in the ceramic is the modulus relating to open circuit or constant displacement conditions, then the antiresonance frequency of the transducer is found. The coupling coefficient of the whole transducer may be calculated from the resonance and antiresonance frequencies, f_r and f_a as follows:

$$k^2 = \frac{f_r^2 - f_a^2}{f_a} \approx \frac{2\Delta f}{f_r} \qquad (\Delta f = f_a - f_r) \tag{4}$$

For the most effective use of a transducer, it is usually desirable to achieve as high a value of the coupling coefficient, k, as possible. The approximate formula for k^2 shows that this means making the proportional difference between f_r and f_a as great as possible.

To do this, the piezoelectric material must be incorporated into the sandwich in such a way that the effect of the boundary conditions (short or open circuit at the electrodes) on its elastic modulus will have the maximum influence on the resonance of the sandwich.

The simplified model of the piezoceramic transducer can only approximate the resonance frequency for the entire device. Therefore, it is necessary to manufacture several sandwich piezoceramic transducers with different dimensions. The electromechanical sandwich transducer works on the reverse piezoelectric effect, converting an electrical power into a mechanical microdisplacement. Each PZT tore of the stack contributes to the entire device displacement.

The ΔL total displacement for a stack made by n piezoceramic tores with L length is a function of d_{33}, the piezoelectric coefficient of the piezoceramic material and U the sandwich stack applied voltage, and the relation between them is:

$$\Delta L = d_{33}\, n\, U \tag{5}$$

The simple sandwich piezoelectric transducers produce motion of a too small amplitude for high power applications. It is common practice to amplify the mechanical motions by means of a tapered resonant horn and the shape of the taper most commonly chosen is exponential. Also, other configurations can be utilized, for example to obtain two resonance frequencies.

The stepped horn consists of two cylinders of different diameter placed end to end concentrically, and its most useful characteristic is that large motion amplification is obtainable. In high power applications of ultrasound, an exponentially tapered solid horn in half-wave resonance is used. The solutions apply to loosely systems and assume Poisson's ratio may be neglected i.e., lateral dimensions are small compared with the length (Figure 2).

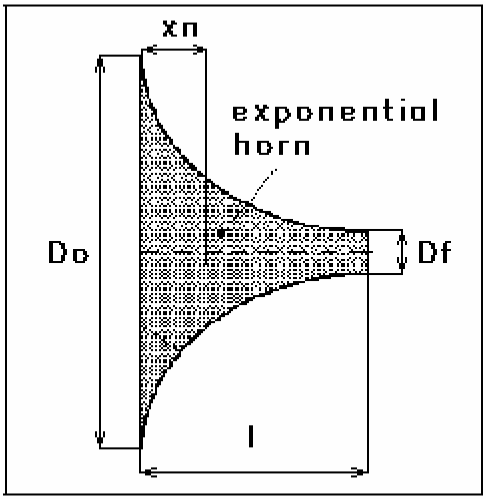

Figure 2 Ultrasonic exponential solid horn

$N = D_0 / D_f$ magnifying factor of the vibration amplitude

The length of the ultrasonic exponential solid horn is given by the relation:

$$l = \frac{c}{2f}\sqrt{1+\left(\frac{\ln N}{\pi}\right)^2} \tag{6}$$

$$x_n = \frac{l}{\pi}\arctan\left(\frac{\ln N}{\pi}\right) \tag{7}$$

As an improvement, the ultrasound acoustical power of the sandwich transducer can be increased in the following ways:

- take an optimum PZT material and metallic elements (for radiator and reflector)
- increase the size (diameter, thickness) of the piezoceramic tores
- take a greater number of piezoceramic tores
- use a proper acoustical transformer (different forms and sizes) to amplify the vibrations
- realize a narrow acoustical angle of the directivity pattern.

3. RESULTS

The technological manufacture requires special mechanical processes to realize the piezoceramic stack, metallic elements and then to encapsulate all together. In this construction (Figure 3), four piezoceramic tores are bolted between a pair of end-metal elements (steel cylinder reflector and exponential hard aluminum radiator).

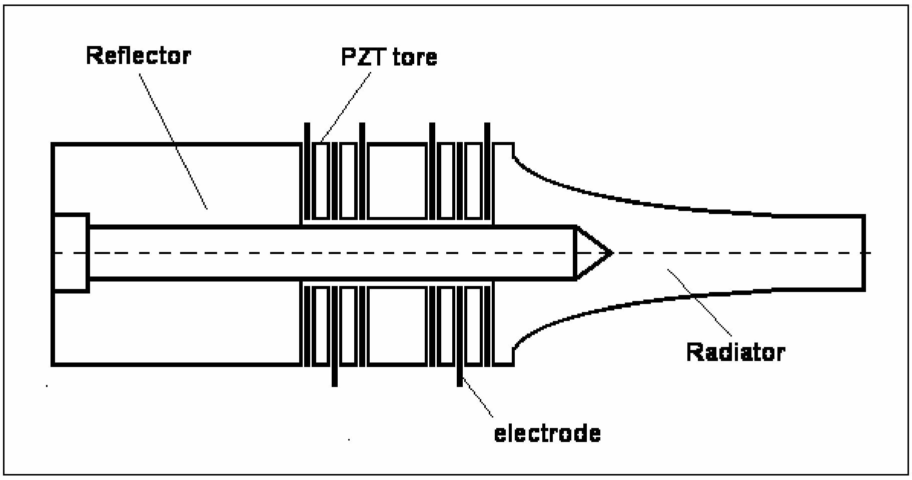

Figure 3 Experimental transducer cross – section

The piezoceramic elements are of pre-polarized lead titanate zirconate composition, which exhibit high activity coupled with both low loss and aging characteristics. They are ideally suited to form the basis of an efficient and rugged transducer. The assembly is clamped together by means of a high tensile bolt, which ensures the ceramics are in compression mode at maximum transducer displacement. Between the PZT tores there are copper electrodes, to make the electrical connection. The PZT-5A tores dimensions are: ϕ25 mm external diameter, ϕ11 mm inner diameter and 4 mm thickness, the steel cylinder reflector is 50 mm length and the hard aluminium reflector is 70 mm length.

The Fig. 4 shows the impedance vs. frequency characteristic of ultrasound piezoceramic transducer, measured by RLC Hioki, 3532-50 equipment, and Fig. 5 presents the phase vs. frequency characteristic.

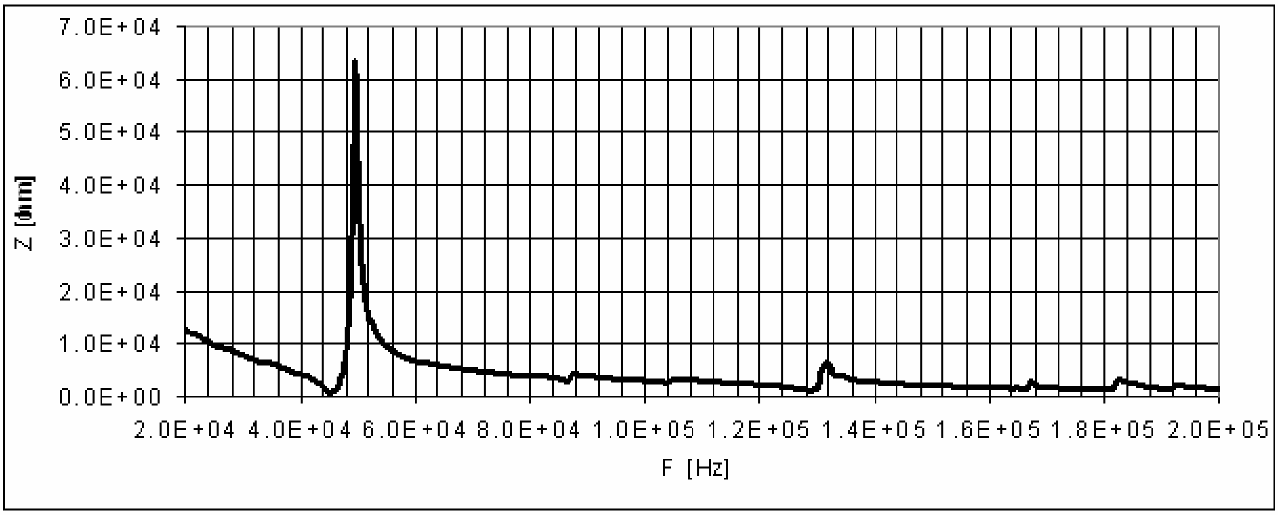

Fig. 4 Impedance vs. frequency characteristic of ultrasound piezoceramic transducer

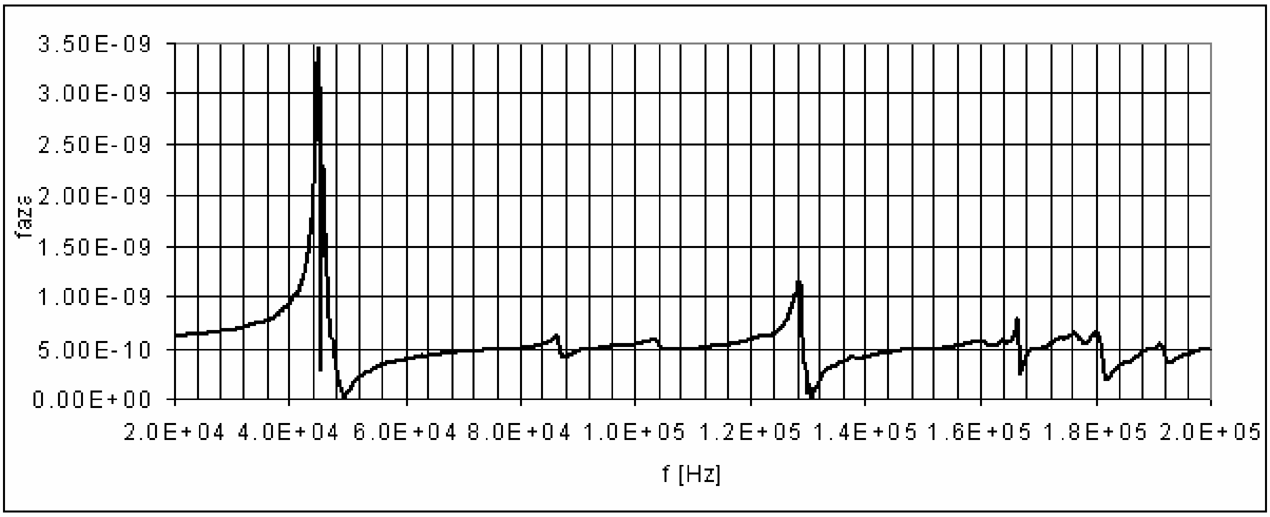

Fig. 5 Phase vs. frequency characteristic of ultrasound piezoceramic transducer

4. CONCLUSION

The ultrasound piezoceramic transducer for sonochemistry applications presents the following advantages:

- it is possible to achieve much grater vibration amplitudes by utilizing a horn with an exponential shape as radiator; as a result of this, much grater energy densities can be achieved
- the acoustical power can be increased by utilizing a greater number of tores and a large tore surface.

ACKNOWLEDEGEMENTS

This study was supported by CEEX X1C15/2005 contract, and the authors wish to thank CEEX program for the financial supports.

References

[1] N. Amara, B. Ratsimba, A. Wilhelm, H. Delmas, *Ultrasonics Sonochemistry*, **11**, 17-21 (2004)

[2] H. Izumi, T. Masakazu, H. Hideto, K. Hiroyuki, *Proceedings of The 5th International symp. Tech.* Korea and Japan Seoul, Korea, August 19-21, (1999)

[3] A. Nacera, B. Ratsimba, A. Wilhelm, H. Delmas, *Ultrasonics Sonochemistry*, **11,** (2004)

[4] L. Hong, J. Wang, Y. Bao, Z. Guo, M. Zhang, Journal of Crystal Growth, **247**, 192-198, (2003)

[5] C. Price, “Ultrasound-The key to better Crystals for the Pharmaceutical Industry”, Pharmaceutical Technology Europe, Publ, No.0240, 59-65, Oct (1997)

[6] J. Linda, P.W. Cains, P.D. Martin, “Use the power of sonocrystallization for improved properties”, *Chem. Eng. Prog.* **97**, 56-61, (2001)

[7] Morgan Matroc Limited, *Transducer Products Division*, Technical Publication TP-235, The design of piezoelectric sandwich transducers

[8] M. Wevers, J.P. Lafaut, L. Baert, I. Chilibon, “Low-frequency ultrasonic piezoceramic sandwich transducer”, *Sensors and Actuators A: Physical*, **122, (2),** 284-289, (2005)