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● *Original Contribution*

NONLINEAR ULTRASONIC PROPAGATION IN BUBBLY LIQUIDS: A NUMERICAL MODEL

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Abstract—In this paper, we investigate the problem of ultrasonic propagation in liquids with bubbles. A new numerical algorithm is constructed to solve the acoustic field-bubbles vibration coupled system. For this purpose, a second-order equation written in a volume formulation is considered for bubbles vibration and coupled with the linear nondissipative wave equation, *i.e.*, attenuation and nonlinear effects are supposed to occur exclusively because of the presence of bubbles. Nonlinear characteristics of the phenomenon are particularly analyzed and illustrated. Plane harmonic waves are first considered in a mixture of air bubbles in water, and conclusions about changes in the wave speed, attenuation, harmonic distortion, effective nonlinearity parameter and nonlinear effects with distance are given. In particular, a law relating the second-harmonic progression with the density of bubbles is found. The propagation of plane pulses is also analyzed to give results on nonlinear attenuation, changes of frequency, and self-demodulation. The influence of the resonance frequency of bubbles on the nonlinear field is then determined. Differences and similarities with nonlinear acoustics in homogeneous fluid are shown and commented. The possibilities and limits of an equivalent nonlinear fluid are then discussed. The propagation of a high-frequency pulsed signal in a bubbly liquid used in a biological application is also the subject of numerical experiments, for frequencies near and beyond the resonance frequency of the bubbles.

Key Words: **Nonlinear acoustics, Ultrasonic waves, Bubbly liquids, Numerical acoustics, Biological applications, Contrast agents, Cavitation.**

INTRODUCTION AND LITERATURE

Linear and nonlinear propagation through a bubbly liquid is an active area of research in acoustics. This is because of scientific interest in the subject as well as the numerous applications for which the presence of bubbles in a liquid is of definitive importance (Hamilton and Blackstock 1998; Jordan and Feuillade 2004, 2006; Kargl 2002; Leseduarte and Quintanilla 2006; Naugolnykh and Ostrovsky 1998).

Microbubbles are used in therapeutic and diagnostic medical applications of ultrasound. For example, they act as contrast agents because their harmonic response in blood allows to distinguish the acoustic scattering of bubbly blood from the surrounding medium. The understanding of microbubbles dynamics is also essential in high-intensity focused ultrasound treatment, lithotripsy and sonodynamic therapy (Matsumoto et al. 2005). These applications have favored a number of publications that analyze the linear and nonlinear ultrasonic propagation in liquids with bubbles: diagnostic applications (Bouakaz et al. 2002; Chen and Zhu 2006; Chin and Burns 2000; Church and Carstensen 2006; Forsberg et al. 1999; Kviklienė et al. 2004; Zhang et al. 2000) and therapy (Fong et al. 2006; Lo et al. 2006; Postema et al. 2004). The ultrasonic propagation in bubbly liquids is also a subject of great interest in underwater acoustics. Important contributions to the understanding of acoustic propagation in bubbly liquid come from the interest in these applications (Karpov et al. 2003; Sutin et al. 1998). In particular, linear and nonlinear wave propagation in bubbly layers was analyzed by several authors (Ng and Ting 1998; Sutin et al. 1998). The shielding and resonance effects of the bubbly layer were studied. The nonlinear response to harmonic waves was analyzed carefully by Karpov et al. (2003), including several discrete frequencies waves. This was motivated by the possibility of generating low-frequency waves by nonlinear parametric excitation and using the bubble layers

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as active media in parametric arrays (Karpov et al. 2003), as well as for characterization (Sutin et al. 1998). Finally, the more general interest of the authors comes from the medical and industrial applications of power ultrasound in multiphase medium (Proyecto CICYT 2005–2008).

It is well known that a liquid containing small gas bubbles has an acoustic behavior dramatically different from that of a homogeneous liquid. The important decrease of propagation velocity, the particular dispersive properties and the increase of losses are still the object of important scientific work. Moreover, the high compressibility of the bubbly liquid causes very important nonlinear acoustic behavior of the fluid. These characteristics represent the objective of the study presented in this paper.

General conclusions can be found in classical references and nonlinear acoustics books (Hamilton and Blackstock 1998; Naugolnykh and Ostrovsky 1998; Zabolotskaya 1974; Zabolotskaya and Soluyan 1973). These pioneers references, based on analytic perturbation techniques, gave a good physical knowledge of the acoustic behavior of a bubbly liquid. In particular, the second harmonic amplitude was calculated, but in these perturbation and frequency domain techniques, the shock formation and nonlinear attenuation could not be predicted. In fact, one of their main conclusions is the dramatic increase of nonlinear features of the liquid when bubbles appear, and then the impossibility to be described quantitatively with perturbation techniques (Hamilton and Blackstock 1998). The bubble equation given by Zabolotskaya and Soluyan (1973) is used in the present article, including the restrictions mentioned in this reference, but it is solved numerically without approximations and in the time domain. In this way, the strongly nonlinear propagation is modeled, including weak shock, as well as strong nonlinear bubble vibration generated by this multifrequency wave. More recently, the same authors also analytically and numerically investigated the nonlinear scattering of gas bubbles in liquids by taking into account the compressibility of the liquid and bubble interaction through the acoustic field (Ilinsky and Zabolotskaya 1992).

A considerable number of works dedicated to the numerical study of the bubble response to a harmonic acoustic field has been published (see for example the work by Lauterborn 1976). More recently, some works considered different acoustic pressure waveforms to excite the bubble and obtained effects of the excitation type on the sonoluminescence efficiency (Chen et al. 2002). Here we also consider the response of the bubble to any type of excitation, but the bubble vibration is coupled to the acoustic field, and we consider the evolution of the bubble vibration when the acoustic field is nonlinearly distorted as well. The main restriction of our model (adiabatic dynamic of the bubble) is also used by Chen et al. (2002).

Models of the propagation in bubbly liquid have been developed for several years from the principle of coupling bubble and propagation equations, but in most of the cases, equations are linearized (Commander and Prosperetti, 1989). Some coupled models including strongly nonlinear waves can also be found in the literature. In particular, Karpov et al. (2003) gave a very exhaustive analysis of the propagation in bubble layers by means of a numerical model. The system bubble-acoustic field was coupled and the dynamics of the bubble vibration did not contain the adiabatic restriction. However, in contrast with what we are assuming in this article, only thin bubble layer and single or dual-frequency excitations were considered. Sutin et al. (1998) developed an analytical model for the nonlinear incoherent scattering, with a physical approximation similar to the one used by Ilinsky and Zabolotskaya (1992). The work of Van Wijngaarden (1968) gave also a good theoretical analysis of the description of the bubbly liquid by an equivalent homogeneous fluid, in particular by means of the KdV equation.

The effect of the shell of encapsulated microbubbles on the nonlinear behavior of the bubbly liquid was studied by Ma et al. (2004). They used an analytical second-order and perturbation model, in the frequency domain, to analyze the nonlinear bubbly liquid behavior by defining a nonlinearity parameter derived from the second harmonic amplitude. Their main result indicates that, when bubbles are encapsulated, the nonlinearity parameter of the liquid decreases with respect to the behavior of free bubbles.

In the medical applications framework, some numerical models of high-frequency pulses propagation have already been proposed. In particular, Church and Carstensen (2006) calculated the responses of free bubbles to realistic incident pulses. Kvikliené et al. (2004) studied the propagation of pulses and their interaction with microbubbles by means of a numerical model in which the complex process is separated: first, the quasi-linear propagation of a transient wave is analyzed; second, the answer of a bubble to the pulse is studied; and finally, the propagation of the scattered echo is evaluated. Stride and Saffari (2005) developed a numerical model to study the attenuation dependence on the frequency (in the MHz frequency range), including nonlinear terms in the bubble vibration. The physical approximation analyzed the nonlinear propagation through a bubbly liquid in a physical approximation quite similar to the one used here but with a different mathematical and numerical model. They compared their numerical results to the ones obtained from experiments showing the importance of considering nonlinear terms.