# NONLINEAR ACOUSTICS PROCESS IN GRANULAR MEDIUM WITH VISCO-ELASTIC FLUID

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## ABSTRACT

Computer simulation of mesoscopic features of granular materials shows a significant nonlinear effect on the acoustic wave. Results of 2-D simulation are compared to experimental research of elastic properties of the granular medium sodden with visco-elastic fluid. Nonlinear relaxation processes are shown in this medium. Both the second order and the third order nonlinear terms are proved to be sensitive to the slow flow of visco-elastic fluid through the granular medium.

# INTRODUCTION

Problem of acoustical effect on fluid flow in granular medium is considered there as a process of acoustically stimulated visco-elastic transition. Visco-elastic properties become more pronounced for the dynamics of confined liquids, where shear stress is essential [1]. At low frequencies elastic media differ from liquids namely by their resistance to shear deformation, which is completely absent in liquids. There is the class of non-Newtonian fluids. For such fluids shear viscosity generally decreases with the increase in shear tension - in the vicinity of a solid boundary this leads to the facilitated slip of the strained fluid regarded. It is known that shear viscosity essentially decreases with the temperature increase as well. This changing in a state of fluids leads to noticeable changing in nonlinear properties of the media.

Beside this in studies of many rocks and other granular media nonlinear elasticity effects were revealed rather at relatively low strains  $\sim 10^{-6}$  or less. Experimental evidence for the highly nonlinear behavior of granular materials has existed for years from experiments of static stressstrain behavior and dynamic nonlinear wave interaction. Due to material nonlinearity, acoustical wave can distort, creating accompanying harmonics, multiplication of waves of different frequencies, and, under resonance conditions, changes in resonance frequencies as a function of drive amplitude. In homogeneous, undamaged materials, these phenomena are very weak and could be observed at essentially strong strains. The sensitivity of nonlinear methods to the detection of changing in the state of the medium (cracks, flaws, etc.) is far greater than that of linear acoustical methods (measures of wave speed and wave dissipation), and in fact, the nonlinear methods appear to be more sensitive than any other method currently available [2]. Nonlinear response of granular matters is very large and easy to measure in contradiction to homogeneous materials. The large nonlinear response arises from the complex compliance of local or volumetric ingomogenieties that are mesoscale (10<sup>-9</sup> m and larger) and entirely dominating the relatively small atomic nonlinearity. However, the nonlinear response is being also related to fluids in porous [3]. The full mechanism of the nonlinear response is not yet well understood. Multiple static and dynamic experiments show that granular materials cannot normally be described by classical theory. They usually call nonlinear mesocopic elastic materials and have at least one of the following properties: they are highly nonlinear, and/or they exhibit hysteresis and discrete memory in their stress-strain relation [4]. Numerous experimental and theoretical researches show hysteresis of mesoscale structures essentially changes nonlinear properties of the media, increases their damping.

Hysteretic media with visco-elastic properties is considered in this paper as a model of granular medium filled with visco-elastic fluid with non-Newtonian properties, oil for example. Hysteretic loops arising in stress-strain relation under cycles of acoustical excitation of mesoscale elements of the medium lead to partial transformation of deformation energy to heat. Therefore virtual heat sources are concentrated in such a medium in the points where mesoscale elements with the most pronounced hysteretic properties exist. When the heat energy (temperature) produced by these sources is sufficient, phase transition occurs from elastic to viscous state of non-Newtonian fluid. Computer simulation of finite amplitude sound wave excitation of 2-D medium with random mesoscale hysteretic elements has been done. It is shown that hysteretic process leads to 2-D percolation cluster formation in the medium structure under cyclic sound excitation. Method of acoustics resonance spectroscopy has been applied to experimental research of visco-elastic phase transition in a granular medium.

## NONLINEAR WAVE PROPAGATION

Let's regard the general equations, which define the process of sound propagation in nonlinear media.

Equation of motion under elastic stress for the medium without dampening is

$$\rho_0 \partial^2 \xi_i / \partial t^2 = \partial \sigma_{ij} / \partial x_j, \tag{1}$$

where  $\rho_0$  is the density of the matter at the rest,  $\xi_i$  is a particle displacement along coordinate *i*,  $\sigma_i$  is *ij* component of the stress tensor. For simplicity, we will consider plane wave propagation in a single direction *x*. Continuous equation, which relates disturbed density  $\rho$  with deformation of the medium can be presented in this case in the obvious form

$$\rho_0 = \rho \left( 1 + \partial \xi / \partial x \right) \tag{2}$$

The equation of state we present as an expansion up to the first nonlinear term (up to second approximation).

$$\sigma = \sigma_{o} + \left( \partial \sigma \right)_{\rho_{0}} \left( \rho - \rho_{0} \right) + \frac{1}{2} \left( \partial^{2} \sigma \right)_{\rho_{0}} \left( \rho - \rho_{0} \right)^{2}$$
(3)

Accord to Eq. (2) and Eq. (3) we can connect stress and strain relation in second approximation as

$$\sigma = \sigma_{o} + \left( \partial \sigma \right)_{o} \varepsilon \left( 1 - \varepsilon \right) + \frac{1}{2} \left( \partial^{2} \sigma \right)_{o} \varepsilon^{2}, \qquad (4)$$

where  $\varepsilon = \partial \xi / \partial x$  is a strain of medium. Substitution Eq. (4) leads us to nonlinear wave equation

$$\frac{\partial^2 \xi}{\partial t^2} - c_0^2 \frac{\partial^2 \xi}{\partial x^2} = 2c_0^2 \left( \frac{\partial c}{c_0 \partial \varepsilon} - 1 \right) \frac{\partial u}{\partial x} \frac{\partial^2 \xi}{\partial x^2}, \tag{5}$$

where  $c^2 = \partial \sigma / \rho_0 \partial \varepsilon$  and  $c_0^2 = \partial \sigma / \partial \varepsilon_0 1 / \rho_0$  is a squared sound velocity in the medium. Typically for granular medium term  $\partial c / \partial \varepsilon >> c_0$ , another word nonlinearity in stress-strain relation determines mainly the nonlinear acoustical processes in granular medium.

Let's come to retarded coordinates  $\tau = t - x/c_0$  and x = x. Then  $\partial^2 / \partial t^2 \rightarrow \partial^2 / \partial \tau^2$ ,  $\partial / \partial x \rightarrow \partial / \partial x - \partial / c_0 \partial \tau$  and  $-\partial / c_0 \partial \tau >> \partial / \partial x$ , therefore  $\partial^2 / \partial x^2 \approx \partial^2 / c_0^2 \partial \tau^2 - 2\partial^2 / c_0 \partial \tau \partial x$ . After that transformations wave equation (5) can be written as

$$\frac{\partial u}{\partial x} = \left( \frac{1}{2\rho_0} c_0^4 \right) \frac{\partial K}{\partial \varepsilon} \frac{\partial \varepsilon}{\partial u} \frac{\partial \tau}{\partial \tau}$$
(6)

Here  $u = \partial \xi / \partial \tau$  is a velocity of medium oscillations in the field of acoustical wave and  $K = \partial \sigma / \partial \varepsilon$  - elasticity modulus.

We can solve this equation by method of successive approximation  $u = u_1 + u_2$ , where  $u_1$  is a solution of linear equation  $\partial u_1 / \partial x = 0$ . It could be any function of  $\tau$ . Then in second approximation we can write

$$\partial u_2 / \partial x = (1/2\rho_0 c_0^4) \partial K / \partial \varepsilon ) u_1(\tau) \partial u_1(\tau) / \partial \tau .$$
(7)

#### MODEL OF HYSTERESIS PROCESS

Based on the work of Preisach and Mayergoyz [5], McCall and Guyer have developed a phenomenological model to describe the quasistatic stress-strain relationship in rocks [6]. In this model, called the P-M space model, the bond system of a rock is represented as an assemblage of hysteretic elastic elements that can be in only one of two states, open or closed. Fig. 1 shows the behavior of a single elastic element as the applied stress varies. The element, originally open with length  $L_0$ , closes to length  $L_c$  as the stress increases to  $\sigma_c$  and it remains closed as the stress continues to increase. When the stress is decreased, the element opens at  $\sigma_0$ , changing back to length L, and remaining there as stress decreases further. We used in our calculations the simplest example of such hysteretic element – an elastic spring with dry friction (Fig. 2). Here  $\Delta L = L_0 - L_c$ . A large number of such elements with differing

 $L_0$ ,  $\sigma_0$  and  $L_c$ ,  $\sigma_c$ ,-models the heterogeneous elastic properties of the rock's bond system.

Refer to statistical distribution of  $L_0$ ,  $\sigma_0$  and  $L_c$ ,  $\sigma_c$  one could obtain different type of hysteretic dependence  $\sigma(\varepsilon)$ . Fig. 3 shows schematically a hysteretic loop in stress/strain relation typical for the granular medium. The specific feature of the media with hysteretic stress/strain relation is that the sign of  $\partial K / \partial \varepsilon$  in Eq. (7) is changing with the sign of medium deformation velocity. This feature leads to essential difference between classical nonlinear and hysteretical behavior. The third harmonics of the fundamental sounding frequency for the pure hysteric material arises in the first nonlinear approximation and this signal distortion is quadratic in the fundamental strain amplitude, whereas a cubic dependence is predicted by classical nonlinear theory.

To extend the hysteric model to 2-D let's regard an elementary scheme of elastic cell with the diagonal bonds shown at Fig. 4. Hysteric properties of such a cell are defined by the diagonal bonds, which could be modeled by elastic elements with dry friction (as shown at Fig. 2). The other bonds of this cell could be regarded as perfect springs with equal elasticity. As while as diagonal bonds make the cell to sustain the shear stress and therefore it could be regarded as a model of elastic state of matter. Some energy will be released in process of cycling load of each hysteretic element  $\delta E \approx \oint \sigma d\varepsilon = kT$ . Accord to P-M approach we regard the difference

between opened and closed state of the hysteretic element  $\Delta L = L_0 - L_c$  is constant. Therefore the energy released in each hysteretic element will be defined by the difference





Fig. 1. Sketch of stress/strain relation for hysteretic element





between  $\sigma_c$  and  $\sigma_0$ , specified for this element. We restrict further our consideration only by the process of energy releasing and will not regard it dispersion due to e.g. heat conduction. That leads to the state when the temperature of each hysteretic element increases in proportion of the difference between threshold stresses  $\sigma_c$  and  $\sigma_0$ , and number of cycles of a

load. When this energy (or temperature) reaches some definite value  $E_0$ , sufficient for phase transition from elastic to fluid state, we come to the acoustical model of fluid state element without diagonal bounds in Fig. 4. 2-D elastic cell without diagonal bonds can't sustain any shear stress like a perfect fluid but conserve it elastic properties to provide sound wave propagation.

Let's regard 2-D array of originally opened hysteretic cells with equal elasticity of each elastic element and with random  $\sigma_c$  and  $\sigma_0$ , obeyed to homogeneous statistical distribution for hysteretic diagonal elements. When we apply quasistatic cycling stress to this array  $\sigma = \sigma_m \sin \omega t$ , elements with  $\sigma_c \leq \sigma_m$  will be involved in hysteretic process. Further one should follow  $\oint \sigma d\epsilon \approx n (\sigma_c - \sigma_0)$  for each hysteretic element and remove diagonal bonds, when this value reaches to  $E_0$  and phase transient occurs. Another words, weak (fluid) cells arise in the array and spatial distribution of elastic properties of the array changes under the cycling load: strains will be concentrated near the fluid points and stimulate the phase transition in the cells next to the fluid ones. Thus, the order arises in originally random process of phase transition in the array under the number of cycles of the load. As a result this ordered process generate a percolation cluster of fluid cells, which provides a fluid flow through the originally elastic array. Fig. 5 shows an example of simulated history of percolation cluster development in the 16×16 array. Grey symbols correspond to fluid cells and darkness shows number of cycles needed for phase transition: less darkness – more cycles. Elastic hysteretic cells are colored in white.



Fig. 5. Scheme of percolation cluster development

As a result of such a modal consideration one could obtain the total elasticity decreasing with the amplitude or time of cycling load application. It is known as slow dynamics effect [7]. It worth to note that heat conduction will be an additional reason which put in order the process of phase transition in random media. Therefore taking into account heat conduction beside with acoustical oscillations make percolation cluster arising more pronounced in the random array of hysteretic elements. We used for the computer modeling a homogeneous statistical distribution for random hysteretical elements. It is clear that the result will be dependent on the type of this distribution. Role of statistical distribution in percolation cluster formation should be investigated additionally.

# **EXPERIMENT**

Visco-elastic behavior of fluid in granular medium was experimentally investigated by nonlinear acoustics resonance method, installation used in experiments is shown at Fig. 6. Cylindrical metal container was filled with pressed quartz sand with granular dimension 0.18-0.23 mm and fixed at magnetostriction vibrator. Container was excited in the range of it resonance frequency (2 kHz - 3.5 kHz). The tar solution in diesel fuel of different concentrations was flowed through the sand during the experiment. First of all a resonance frequency shift downward with the level of excitation was observed. It is a typical feature of granular matter and was registered by numerous researchers. Q-factor of resonance vibrations has been investigated as well as the relative change in the rate of the tar solution flow in dependence on the amplitude of acoustical vibrations. Q-factor changing (spreading of the resonance curve) reflects the increasing of the effective value of viscosity of the medium. Another words the visco-elastic balance in the parameters of the sample shifts to the viscosity increase and to the elasticity decrease. Both resonance frequency shift downward and Q-factor decreasing with the level of excitation proves that the sample under investigation rather looses it properties as solids and obtains more emphasized properties of fluid with the vibration amplitude increase.

Another evidence of hysteretic properties of granular medium filled with visco-elastic fluid is it capability to distort acoustical signal and generate the higher harmonics. Fig. 7 shows the dependence of the first three harmonics on the level of signal excitation.. It is seen that amplitude of the third harmonic response is rather higher then the same amplitude for the second one in the whole range of signal level variation. While the signal level is linear increasing (in log scale) in time at the fundamental frequency, amplitude of response at the



Fig. 6. Experimental setup



Fig. 7. Amplitude dependence for the first three harmonics for signal in granular medium.

third harmonic rises in proportion to squared amplitude at the fundamental frequency as the second one does. Such feature is a typical one for the granular media and is related with the hysteresis in equation of state for this matter [8]. Fig. 8 shows a rate of the tar solution flow dependence on the external pressure under the acoustical vibration of the maximum amplitude at the eigen frequency of resonator. While acoustical vibrations don't effect on the diesel fuel (0% of tar concentration) flow through the container with sand, they influence essentially changes when we deal with more viscous fluids. Rate of the flow of tar solution of 18% concentration (10 mPas viscosity) becomes in proportional to external pressure  $\Delta P$  $(U_{0} \text{ at the graph})$ , which corresponds to Poisel low. Whereas acoustical vibrations are absent it is shown the effective permeability of the medium continuously decreases (U at the graph). Non-integral power in  $U(\Delta P)$ dependence points out at the fractal pattern of permeability development in granular medium. Fluid can suddenly stop to flow under external pressure less then 5 mm. It looks like 'frozen' medium. But this phase transition from elastic state processes fluid to randomly. 'Frozen' fluid occasionally can flow again. Acoustical vibrations



Fig. 8. Flow rate of visco-elastic fluid through granular medium versus external pressure  $\Delta P$ . U – flow with sound, U<sub>0</sub> flow without sound.

stimulate fluid to follow the Poisel low as well and let the fluid flow the regular character.

Random hysteretic media is considered as a model of granular medium filled with visco-elastic fluid with non-Newtonian properties, oil for example. This model was applied to consider the process of acoustical effect on visco-elastic fluid flow in porous or granular medium. It was shown that acoustical vibrations lead to phase transition in elementary hystretic cells and the matter rather looses it properties as solids and obtains more emphasized properties of fluid with the vibration amplitude increase. It is rather rough model, which involve only the hysteretical type of acoustical effect to

the medium. But such an approach apparently defines the main features of this phenomenon – slow dynamics and fractal nature of percolation cluster development. The model regarded provides origin 2-D or rather 3-D structures in granular medium filled with visco-elastic fluid due to single dimension acoustical excitation. Inhomogeneous hysteretical cells transform the longitudinal acoustical oscillations to shear ons. And shear perturbations essentially change of properties of non-Newtonian confined fluid, they transform visco-elasic matter from elastic to fluid state.

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