Liquid Temperature Effect on Sound Propagation in Polymeric Solution with Gas Bubbles

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Abstract—Acoustic properties of polymeric liquids are high sensitive to free gas traces in the form of fine bubbles. Their presence is typical for such liquids because of chemical reactions, small wettability of solid boundaries, trapping of air in technological operations, etc. Liquid temperature influences essentially its rheological properties, which may have an impact on the bubble pulsations and sound propagation in the system. The target of the paper is modeling of the liquid temperature effect on single bubble dynamics and sound dispersion and attenuation in polymeric solution with spherical gas bubbles. The basic sources of attenuation (heat exchange between gas in microbubbles and surrounding liquid, rheological and acoustic losses) are taken into account. It is supposed that in the studied temperature range the interface mass transfer has a minor effect on bubble dynamics. The results of the study indicate that temperature raise yields enhancement of bubble pulsations and increase in sound attenuation in the near-resonance range and may have a strong impact on sound dispersion in the liquid-bubble mixture at frequencies close to the resonance frequency of bubbles.

Keywords—Sound propagation, gas bubbles, temperature effect, polymeric liquid.

I. INTRODUCTION

ACOUSTIC properties of polymeric liquids have importance for polymer processing technology and acoustic monitoring of polymer dynamic modules [1], [2]. Acoustic method is also one of the well-known tools for bubbles diagnostics, because both sound speed and attenuation are highly sensitive to free gas concentration [3]. Theoretical foundation of the method in the case of polymeric bubble-liquid mixture needs appropriate description of bubble dynamics with account for non-Newtonian properties of the liquid. Probably, first approach to modeling of bubble dynamics in liquid with complex rheology was formulated in [4], where effect of liquid shear-thinning was incorporated in the original Raleigh model of spherical cavity collapse. First results on bubble dynamics in viscoelastic liquids were reported in [5], small amplitude pulsations of gas bubble in Oldroyd liquid were studied in [6]. Detailed review of further publications in this field, both theoretical and experimental, can be found in the book [7]. More recent results were reported in [8]-10], etc. Comprehensive reviews of last years' publications on bubble dynamics in non-Newtonian liquids and certain relevant problems can be found in [11], [12]. Theory of sound propagation in viscoelastic liquid with bubbles was developed in [13], [14]. Viscoelasticity is the most important property inherent to polymeric liquids

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(solutions and melts). At a macroscopic level this kind of non-Newtonian behavior is described by relaxational models using time derivative of the stress tensor, or by equivalent integral equations [15]. Temperature has essential impact on both relaxation spectrum of liquid and its viscosity, which yields temperature dependence of losses at single bubble pulsations in the wave and may cause changes in sound speed and attenuation in the system. The target of the study is to describe this effect and to estimate its manifestation quantitatively for the case of polymeric solution with fine gas bubbles.

II. SINGLE BUBBLE DYNAMICS

A. Basic Model

General approach to modelling of single vapour-gas bubble dynamics in polymeric liquid was developed in [16]. As distinct to [16], the analysis presented below is limited to the case of pure gas bubble, which leads to essential simplification of the resulting relation for the amplitude of bubble pulsations in a sound field, found in [16]. It is supposed that rheological behaviour of polymeric liquid, surrounding the bubble follows generalized linear Maxwell model [15], which is used usually for description of polymer liquids dynamics:

$$\tau_{ij} = 2 \int_{-\infty}^{t} G(t - t_1)s_{ij}(t_1)dt_1 + 2\eta_s s_{ij} \tag{1}$$

$$s_{ij} = e_{ij} - \frac{1}{3}(\nabla \cdot \vec{v})I, \quad e_{ij} = \frac{1}{2}(\frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i})$$

Here $G(t - t_1)$ is the relaxation function; τ_{ij}, s_{ij} - deviators of stress and rate-of-strain tensors, respectively; η_s - low-molecular solvent viscosity; $\vec{v}$ - liquid velocity. Rheological equation (1) means that only shear viscoelasticity of liquid is accounted for hereafter, and relaxation features at bulk deformations of liquid in the wave are neglected. It was shown elsewhere [7] that the input of liquid volume viscoelasticity in bubble dynamics is small in a wide range of conditions, as compared with other sources of dispersion and dissipation.

It is well known [3] that gas bubble in a sound field of small amplitude can be treated as linear oscillator with effective elasticity and dissipation constants. Let denote the bubble radius in the wave as $a = a_0 + \Delta a$, $\Delta a / a_0 \ll 1$, where a_0 is the equilibrium radius. The pressure in liquid can be described by the relation $p = p_0 + p_a e^{i\omega t}$. Then, choosing p_0, a_0 and equilibrium liquid density ρ_0 for characteristic parameters, and searching the bubble radius variation in the form

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$\Delta a / a_0 = \delta e^{i\omega t}$, we can come [7] to the following relation for dimensionless amplitude of the bubble, normalized by the dimensionless sound pressure $\bar{p}_a = p_a / p_0$:

$$\delta_* = \delta / \bar{p}_a = [(\Omega^2 - \chi) - 2i\Omega \nu]^{-1}, \quad (2)$$

$$\nu = \frac{\Omega^2}{2\bar{c}_f} + \frac{2\,\mathrm{Im}\{G^*\}}{\Omega} + 2\bar{\eta}_s + \frac{\mathrm{Im}\{D_T\}}{2\Omega}$$

$$\chi = 4\,\mathrm{Re}\{G^*\} + \mathrm{Re}\{D_T\} - 2\bar{\sigma}$$

$$D_T = \frac{3\bar{p}_{g0}\beta^2\gamma}{\beta^2 - 3(1-\gamma)(\beta\,\mathrm{cth}\,\beta - 1)}, \quad \beta^2 = i\Omega\,\mathrm{Pe}_g$$

$$\mathrm{Pe}_g = \frac{a_0 (p_0 / \rho_0)^{1/2}}{a_g}, \quad a_g = \frac{k_g}{\rho_{g0} c_{pg}}, \quad \bar{p}_{g0} = \frac{p_{g0}}{p_0} \quad \rho_{g0} = \frac{p_{g0}}{T_0 c_{pg}(1-\gamma^{-1})},$$

$$\bar{c}_f = (t_0 / a_0)c_f, \quad \bar{\sigma} = \sigma / (p_0 a_0)$$

$$\Omega = \omega t_0, \quad t_0 = a_0 (\rho_0 / p_0)^{1/2}, \quad \bar{\eta}_s = \eta_s / (p_0 t_0)$$

Here p_{g0}, ρ_{g0}, T_0 are equilibrium pressure, density and temperature of the gas phase; k_g, c_{pg}, γ - the gas thermal conductivity, specific heat capacity at constant pressure and adiabatic exponent, respectively; Pe_g - Peclet number; η_s - solvent viscosity, c_f - sound speed in liquid, σ - liquid-gas surface tension coefficient, G^* - complex dynamic module of viscoelastic liquid:

$$G^* = \int_0^\infty \frac{(\Omega\bar{\theta})\bar{F}(\bar{\theta})(i + \Omega\bar{\theta})}{1 + (\Omega\bar{\theta})^2} d\bar{\theta}, \quad \bar{\theta} = \theta / t_0 \quad (3)$$

In (3) $\bar{F}(\bar{\theta})$ is the non-dimensional spectrum of relaxation times θ. Equation (2) is based on exact solution of the problem of bubble dynamics in viscoelastic liquid, derived in [16]. It is valid in a wide range of frequencies because accounts not only for rheological losses, but also for heat and acoustic dissipation at single bubble interaction with the liquid. When deriving (2), it was assumed that the thermal boundary layer thickness around pulsating bubble is much smaller than its radius and the pressure in bubbles doesn't depend on the spatial coordinate, that is $p_g = p_g(t)$ and homobaricity takes place in the gas phase [17]. Parameters ν, χ in (2) have the meaning of effective dissipation coefficient and elastic constant of the bubble, respectively. One can see that within the formulated model, rheology of the liquid influences both parameters in an additive manner. More exact analyses of bubble dynamics in viscoelastic liquid [16], taking into account cross effects of liquid rheology, heat transfer and liquid compressibility, has showed that their input in total dissipation and effective elasticity of the bubble is minor and can be neglected in the frequency range $\omega \ll 10^6\,Hz$.

In the case of a discrete spectrum (3) takes the form

$$(i\Omega)^{-1} G^* = \frac{\bar{\eta}_p - \bar{\eta}_s}{z(\alpha)} \sum_{k=1}^\infty \frac{k^\alpha - i\Omega\bar{\theta}_1}{k^{2\alpha} + (\Omega\bar{\theta}_1)^2} \quad (4)$$

where η_p is Newtonian (low-frequency) viscosity of polymeric solution, and $z(\alpha)$ is the Riemann zeta function of the spectral distribution parameter α, introduced according to the Spriggs law [15]: $\theta_k = \theta_1 / k^\alpha$.

B. Temperature Effect

The main relaxation time in the spectrum θ_1 can be estimated from the Rouse formula [18]:

$$\theta_1 = 0.608 \frac{(\eta_p - \eta_s)M}{\phi R_G T} \quad (5)$$

where M, ϕ are molecular mass of the polymer and its concentration. The dependency of the solution Newtonian viscosity, η_p, on the polymer concentration ϕ at not too large values of ϕ is described usually by the Martin relation: $\eta_p = (1 + \beta \exp(k_M \beta))\eta_s$. Here k_M is the Martin constant, $\beta = \phi[\eta]$ - reduced polymer concentration, $[\eta]$ - characteristic viscosity of solution, R_G is the universal gas constant and T the absolute temperature. The value of $[\eta]$ can be estimated from the Mark-Houwink relation:

$$[\eta] = KM^b \quad (6)$$

where K and b are constants for a given polymer-solvent pair at a given temperature over a certain range of molecular mass variation. The parameter b (the Mark-Houwink exponent) lies in the range 0.5 to 0.6 for solutions of flexible chains polymers in thermodynamically bad solvents and in the range 0.7- 0.8 for good solvents [10]. For the former ones the constant $K \approx 10^{-2}$ (if the intrinsic viscosity $[\eta]$ is measured in cm^3/g), while for the latter $K \approx 10^{-3}$. Temperature dependence of the relaxation characteristics of polymeric liquids is described by the time-temperature superposition (TTS) principle [18], which states that with change in temperature the spectrum of relaxation times shifts as a whole in a self-similar manner along t axis, according to the value of the temperature-shift factor a_T, defined by:

$$a_T = \frac{\rho(T_0)T_0(\eta_p(T) - \eta_s(T))}{\rho(T)T(\eta_p(T_0) - \eta_s(T_0))} \quad (7)$$

where ρ is the liquid density. With Ωa_T for an argument it becomes possible to plot temperature-invariant curves $\mathrm{Re}\{G_1^*(\Omega a_T)\}$ and $\mathrm{Im}\{G_1^*(\Omega a_T)\}$. The temperature dependence of viscosity is described by activation theory:

$$\eta_p = \eta_{p0} \exp[E_p(R_G T_0)^{-1}(T_0/T - 1)] \quad (8)$$

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$$\eta_s = \eta_{s0} \exp[E_s(R_G T_0)^{-1}(T_0/T - 1)]$$

where E_p, E_s are activation energies of the solution and the solvent, respectively; $\eta_{p0} = \eta_p(T_0)$, $\eta_{s0} = \eta_s(T_0)$. The E_s value is usually about 10 to 20 kJ/mol. For low-concentrated solutions of polymers with moderate molecular masses, the difference between these two activation energies, $\Delta E = E_p - E_s$, does not exceed usually 10 kJ/mol [10]. For low-concentrated solutions of certain polymers in thermodynamically bad solvents negative ΔE values were reported [19].

Relation (2) was studied numerically with account for the temperature dependence of rheological parameters, according to (4), (5), (7), (8), which were written in the dimensionless form as:

$$\bar{\theta}_{10} = 0.608\bar{\eta}_{s0}A\exp(k_M\beta), \quad A = ([\eta]p_0 M)/(R_G T_0)$$
$$\bar{\theta}_1(T^*) = a_T\bar{\theta}_{10}, \quad a_T = (\bar{\rho}T^*)^{-1}(\bar{\eta}_p - \bar{\eta}_s)/(\bar{\eta}_{p0} - \bar{\eta}_{s0})$$
$$\bar{\eta}_p = \bar{\eta}_{p0}\exp[\bar{E}_p(T^{*-1}-1)], \quad \bar{\eta}_s = \bar{\eta}_{s0}\exp[\bar{E}_s(T^{*-1}-1)] \quad (9)$$
$$\bar{\rho}_{f0} = [1 + \bar{\alpha}_1(T^*-1)], \quad \bar{\rho}_{f0} = \rho_{f0}(T)/\rho_{f0}(T_0), \quad \bar{\alpha}_1 = \alpha_1 T_0$$
$$\{\bar{\eta}_p, \bar{\eta}_s, \bar{\eta}_{p0}, \bar{\eta}_{s0}\} = \{\eta_p, \eta_s, \eta_{p0}, \eta_{s0}\}/(p_0 t_0)$$
$$\{\bar{E}_p, \bar{E}_s\} = \{E_p, E_s\}/(R_G T_0), \quad T^* = T/T_0, \quad \theta_{10} = \theta_1(T_0)$$

Here α_1 is the liquid thermal expansion coefficient. Plots of the non-dimensional normalized bubble amplitude $\delta_1 = |\delta_*|$ versus frequency Ω and temperature T^* were obtained for different values of reduced polymer concentration β. The values of the system parameters, chosen for simulations, are collected below; they were estimated for air bubble in solution of polymer with $M \sim 10^6$ [9]:

$$p_0 = 10^5 Pa, \ \eta_s = 0.1 Pa \cdot s, \ \rho_0 = 10^3 kg/m^3 \quad (10)$$
$$c_f = 1500 m/s, \ k_M = 0.4, \ \bar{E}_p = 6.6, \ \bar{E}_s = 4.9$$
$$A = 500, \ \bar{\alpha}_1 = 0.3, \ \bar{\sigma} = 0.03$$

The non-dimensional values of activation energies correspond to $E_s = 12 kJ/mol$, $E_p = 16 kJ/mol$; the studied temperature range is equal approximately to $60K$. The Spriggs distribution parameter α was chosen equal to 2 (Rouse distribution).

Results of simulations are presented on Figs. 1-3. For all plots $a_0 = 5 \cdot 10^{-4} m$, the studied non-dimensional frequency plots $a_0 = 5 \cdot 10^{-4} m$, the studied non-dimensional frequency range for the chosen parameter values corresponds approximately to dimensional frequency $1.5 < f < 11 kHz$, $f = \omega/2\pi$. The 3D plot on Fig. 1 was calculated for $\beta = 4$.
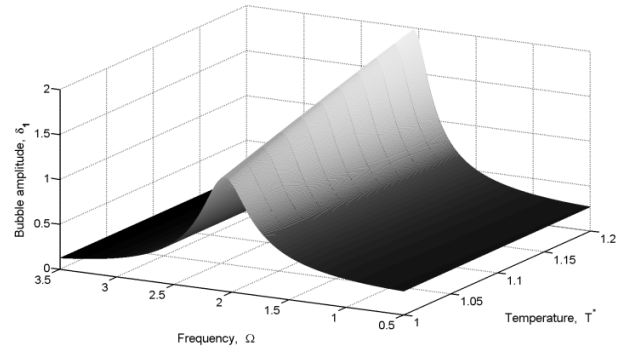

Fig. 1 Liquid temperature and frequency dependence of the bubble amplitude in polymeric liquid

As it follows from Fig. 1, bubble amplitude grows with temperature. The result is explained by the dynamic viscosity reduction. It can be seen also that the temperature has minor effect on the resonance frequency of bubbles in the studied range. This effect is clearly seen on the plots presented on Fig. 2, which were calculated for different concentrations of polymer: the curves $1, 1'$ correspond to $\beta = 0.2$; the curves $2, 2'$ - to $\beta = 4$; and curves $3, 3' -$ to $\beta = 6$. The resonance amplitude of bubbles drops with polymer concentration and for each value of β the amplitude-frequency curves shift slightly to the left with temperature. The effect is explained by reduction of viscous losses and liquid elasticity with growth of T^*, accompanied by narrowing of the relaxation spectrum, and is more pronounced for larger polymer concentrations. Resonance frequency of bubbles is increased with β due to increase in the liquid elastic modules (the effect was described also in [20] for free pulsations of bubbles in viscoelastic fluid).
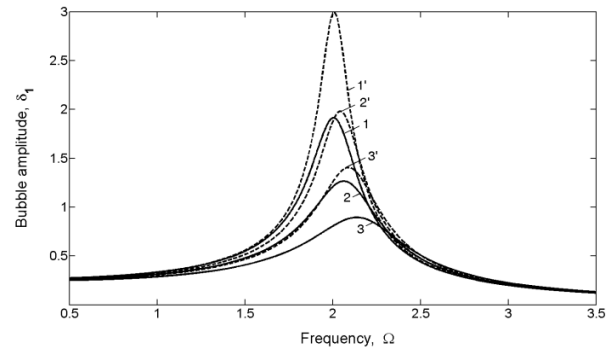

Fig. 2 Liquid temperature effect on bubble amplitude at different concentrations of polymer: Solid lines - $T^* = 1$, dashed lines - $T^* = 1.2$

Growth of the bubble resonance frequency with polymer concentration is responsible for another result, illustrated by plots on Fig. 3, where the influence of liquid temperature on bubble amplitude was studied separately at sub-resonant and super-resonant values of Ω. Despite all curves demonstrate amplitude growth with temperature, the plot, corresponding to sub-resonant frequency (solid line 1) at small polymer

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concentration $\beta = 0.2$ (slightly viscoelastic solution with $\bar{\eta}_s \approx 0.02$, $\bar{\eta}_p \approx 0.024$)), is located above the dashed line 1, calculated for super-resonant frequency. For larger values of β the relative position of solid and dashed lines is opposite. This result is explained also by shift to the right of the resonance curves because of changes in resonant frequency of bubbles with growth of polymer concentration.
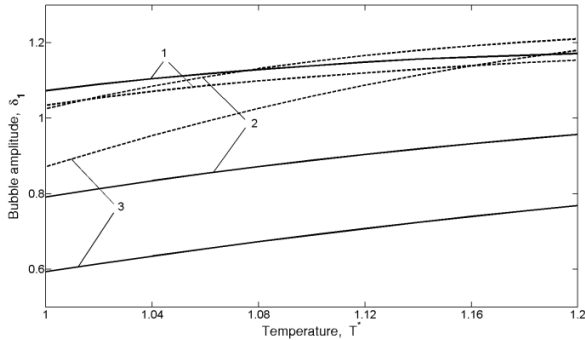

Fig. 3 Liquid temperature effect on bubble amplitude at sub-resonant ($\Omega = 1.8$, solid lines) and super-resonant ($\Omega = 2.2$, dashed lines) frequencies: Curves 1- 3 correspond to $\beta = 0.2, 4, 6$

III. Sound Propagation in Polymeric Liquid with Bubbles

A. Basic Model

Sound propagation in polymeric liquid with bubbles is described within homogeneous approach [21], based on the following assumptions: bubbles in the mixture are spherical; their radii are much smaller than the typical distance between the bubbles, while the latter is smaller than the size of an averaging volume, within which a representative number of bubbles can be found. Liquid density in the wave and its equilibrium value ρ, ρ_0, respectively, are related to the pressure in the wave by the state equation $p = p_0 + c_f^2(\rho - \rho_0)$. Here c_f is the sound speed in a pure liquid without bubbles, and p, p_0 represent the instant pressure in liquid and its equilibrium value, respectively. Solution of the problem is searched in the form of a wave, travelling in the positive direction of x - axis, which is described in a non-dimensional form by a standard multiplier $\exp[i\Omega(\tau - \bar{x}/\bar{c}_m)]$. Here τ, $\bar{x}$, $\bar{c}_m$ are the non-dimensional time, longitudinal coordinate and complex sound speed in the mixture, respectively. It is well known that bubbles, even a minor amount, are the basic source of dissipation at sound wave propagation, and that sound absorption in a pure liquid is small, as compared with dissipation arising from radial oscillations of bubbles in the wave [3]. The physical reason for this result is that dissipation at sound propagation in bubbly liquids is located mainly in thin spherical liquid layers with large velocity gradients around pulsating bubbles [7]. Therefore, when formulating the dispersion equation for the mixture, the liquid rheology terms can be retained solely in

equations describing bubble dynamics, and they may be omitted from the momentum balance equation for the bulk of liquid. Such approach has gained wide acceptance when modelling wave propagation in pure viscous bubbly liquids [21]. For the case of viscoelastic liquid, this approach was verified in [14], where dispersion equation for vapor-gas bubble mixtures in polymeric solutions was obtained by the multiple scattering method, and rheology terms were retained in both the momentum balance equation for the bulk of liquid and the equations describing single bubble dynamics. In the case of a monodisperse mixture of gas bubbles the resulting dispersion equation is simplified essentially and can be written in the form (the details may be found in [22]:

$$\frac{1}{\bar{c}_m^2} = \frac{1}{\bar{c}_f^2} - 3\varphi_0(1-\varphi_0)\frac{3\varphi_0(1-\varphi_0)}{(\Omega^2 - \chi) - 2i\Omega\nu} \quad (11)$$

Here φ_0 is the equilibrium volume concentration of free gas, and effective elasticity and dissipation coefficients of a bubble are defined by (2).

B. Temperature Effect

Results of simulations for the case of air bubbles with equilibrium radius $a_0 = 5 \cdot 10^{-4}\,m$ and volume concentration $\varphi_0 = 10^{-5}$ are presented on Fig. 4 in the form of 3D plot of non-dimensional sound speed $C = \text{Re}\{\bar{c}_m\}$ versus dimensionless frequency Ω and temperature T^*. The data correspond to polymeric solution with $\beta = 4$; the rest of parameters are the same as for Fig. 1.
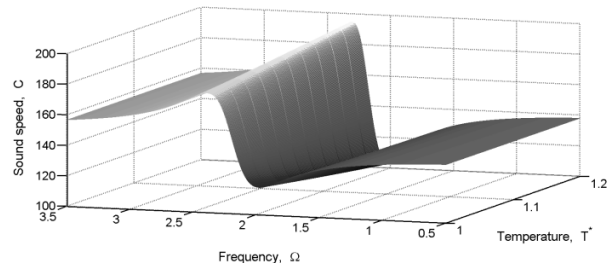

Fig. 4 Sound speed in the bubbly mixture versus frequency and liquid temperature

As it is typical for bubbly mixtures [21], the low and high-frequency dispersion (for $\Omega \ll 2$ and $\Omega \gg 2$, respectively) is negative, while in the vicinity of the resonance frequency the dispersion sign is changed for an opposite. Strong changes of sound speed in this narrow frequency range appear even at very small gas concentrations. The dispersion is enhanced with the temperature growth. Effect of liquid temperature on the sound speed is well seen on Fig. 5, obtained by projection of 3D plots $C = C(\Omega, T^*)$ on $C - \Omega$ plane for two bubbly mixtures with different polymer concentrations. The data presented on Fig. 5 correspond to $\beta = 0.2$ (plot 1) and $\beta = 6$ (plot 2), respectively; the vertical arrow indicate direction of

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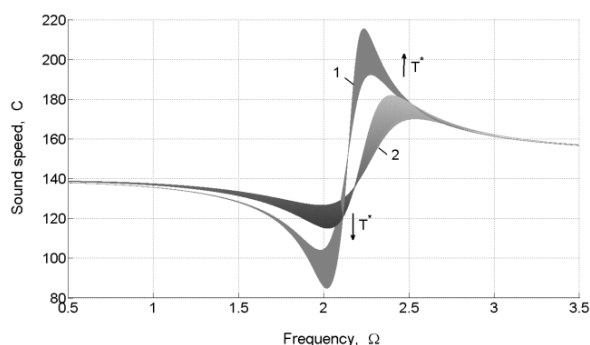the temperature raise across the plots on two branches of the dispersion curves.

Fig. 5 Temperature effect on the dispersion range

As it follows from Fig. 5, the temperature influence on the sound speed in liquid with bubbles is located in the bubble resonance range, in accordance with results, discussed in the previous section. The effect of liquid temperature is small far from the strong dispersion region.

The last figure is aimed to demonstrate the liquid rheology effect on sound attenuation at different temperatures. Curves 2, 2' were calculated for pure viscous liquid with viscosity η_p, equal to Newtonian viscosity of polymeric solution with $\beta = 4$; the curves 1, 1' correspond to the same liquid with account for its viscoelasticity.
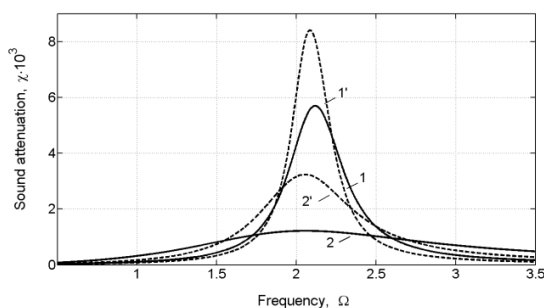

Fig. 6 Temperature effect on sound attenuation in pure viscous (curves 2, 2') and viscoelastic liquids (curves 1, 1') For the lines 1, 2 - $T^* = 1$; for 1', 2' - $T^* = 1.2$

The data presented on Fig. 6 indicate that temperature raise yields attenuation enhancement (both in viscous and viscoelastic liquid). However, it takes place only in the range of strong sound dispersion, were temperature changes lead to essential increase in the bubble pulsations amplitude. Far from this region (for sub- and super-resonant frequencies) temperature raise leads to attenuation reduction. Note that in this region attenuation of sound in viscoelastic solution of polymer is less than in an equivalent pure viscous two-phase mixture.

IV. CONCLUSION

The study has revealed essential impact of liquid temperature on single bubble dynamics and sound propagation in polymeric liquid with bubbles. The main effect of the temperature is localized in the vicinity of the bubbles resonance frequency. Growth of temperature yields increase of the amplitude of acoustically induced pulsations of bubbles and leads to the sound dispersion enhancement. The scale of the effect is sensitive to polymer concentration – its growth is responsible for the bubble amplitude and sound dispersion reduction. Dispersion curves for more concentrated solutions of polymer shift to the range of larger frequencies, which is explained by the impact of liquid elasticity on the natural frequency of bubbles. Temperature raise also yields the attenuation enhancement in the near-resonance range. For sub- and super-resonant frequencies the temperature raise leads to reduction of the sound attenuation.

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