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**Phone:** 2-2716

**E-mail:** santos@isc.tamu.edu

**Name:** juan santos

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Classification

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## Very slow propagating mode at audiofrequencies in gel

J. C. Bacri, J. Dumas and A. Levelut

Laboratoire d'Ultrasons (\*), Université Pierre-et-Marie-Curie, Tour 13, 4, place Jussieu, 75230 Paris Cedex 05, France

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**Résumé.** — Nous avons mis en évidence un mode élastique (longitudinal) de gonflement à très basse fréquence (de 100 Hz à 2 000 Hz) dans un gel polyacrylamide à 10 %. Nous avons trouvé une vitesse de phase très faible (2,3 m/s) directement reliée à la constante de cisaillement du réseau. Nous mesurons une viscosité liée au polymère.

**Abstract.** — An elastic dilatational mode at very low frequency (100 Hz to 2 000 Hz) has been found in a 10 % polyacrylamide gel. The phase velocity, which is directly related to the polymer shear constant, is very low (2.3 m/s). A polymer network viscosity has also been measured.

**1. Introduction.** — A gel is an infinite crosslinked polymer network imbedded in a fluid solvent. Mechanical vibrations in gel can be studied in three regions of frequencies which give different kinds of information :

— at high frequencies ( $\geq 100$  MHz), the elastic constant measured is that of water slightly modified by the polymer, and the sound attenuation is related to the friction between the network and the solvent [1] ;

— at intermediate frequencies ( $\approx 1$ -20 MHz) the relaxations of internal motions in monomers are the main parameters affecting ultrasonic measurements [2] ;

— at very low frequencies (0-10 kHz), static measurements and light scattering experiments [3, 4, 5] give very low values of the elastic constant of the gel,  $10^4$  times smaller than high frequency values.

The aim of this letter is to present direct dynamic measurements of the elastic shear constant in this very low frequency range, and some information about the viscosity of the polymer network. These results are obtained by the observation of an axially symmetrical dilatational mode in a gel cylinder.

**2. Theory.** — In order to interpret the present very low frequency experiment we consider the gel as a single-component solid. Our reasons for adopting this description rather than the two component model used in interpreting high frequency experiments [1]

are the following. In the two-component model the displacement  $r$  of the polymer network and the displacement  $s$  of the solvent are coupled through a friction term  $f(\dot{r} - \dot{s})$  [1, 6]. This term (which varies as  $\omega$ ) is dominant at low frequencies as compared to the inertial terms and to the restoring force terms (which vary as  $\omega^2$ ). As a consequence, in our frequency range ( $\nu < 2\ 000$  Hz), the movements of the two components are in phase and they may be accurately described by a single variable. In fact the value of the friction coefficient  $f \approx 10^{10}$  dynes s.cm<sup>-4</sup> corresponds to a swelling time of the order of some days [7] : the solvent does not flow out of the polymer mesh during the duration of the experiment. Therefore, we suppose the network and solvent movements are in phase in the gel which may be considered as a single homogeneous isotropic solid. Under these conditions we have to study a sample mainly composed of a solvent held in the shape of a rod by the polymer network. This sample is a solid : it has a shear modulus which is due to the polymer. Then we may put

$$\mu_g \approx \mu_p$$

where  $\mu_p$  is the shear modulus of the polymer network. Static measurements give an order of magnitude for this coefficient :  $\mu_p \approx 2 \times 10^5$  dynes cm<sup>-2</sup> for a 10 % acrylamide gel [3]. This solid sample has a compressibility or a bulk modulus  $K_g$  which may be measured, in principle, in a hydrostatic compression measurement. The compressibility is mainly due to the solvent

(\* ) Associé au Centre National de la Recherche Scientifique.

$$K_g \approx K_s$$

This very low value shows that the acoustic impedance mismatch between transducer and gel is high and therefore the reflection is quasi total. The wave velocity in gel is about  $10^2$  lower than in the surrounding air : the elastic energy cannot be radiated because of total reflection. Moreover, the losses due to viscoelastic wave emission in air are negligibly small. Scattering from sample inhomogeneities is also not a relevant cause of attenuation considering the wavelength of the waves (0.1 cm to 1 cm) and the excellent optical quality of the sample on this scale.

Thus the exponentially decreasing echo amplitude  $A_n$  ( $n$  : transmitted echo number) gives a measurement of the true attenuation of the mode

$$\frac{A_{n+1}}{A_n} \propto e^{-q''l}$$

where  $l$  is the sample length. Figure 2 shows the exponential envelope of the transmitted echoes which corresponds to an attenuation

$$q'' \approx 2 \times 10^{-6} \nu^2 \text{ cm}^{-1}$$

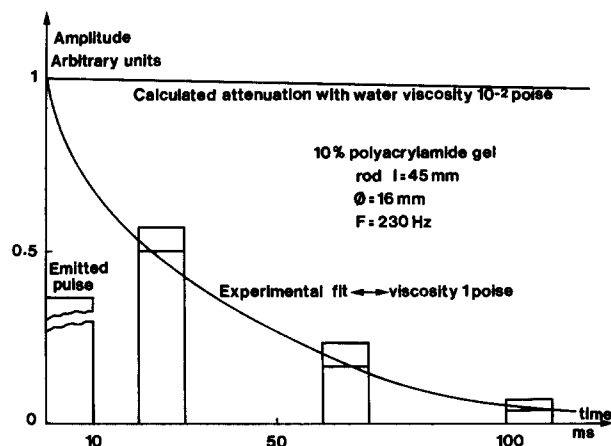


Fig. 2. — Measurement of transit time and attenuation of the dilatational mode for transmitted pulses. — Experimental fitting corresponding to a 1 P viscosity ; — Calculated curve for attenuation in the same length of water, viscosity  $\eta = 0.01$  P.

where the frequency  $\nu = \omega/2\pi$  is given in Hz. This corresponds to a measured viscosity :

$$\eta' \approx \eta_g \approx 1 \text{ P.}$$

$\eta_g$  is then, roughly, 100 times the solvent viscosity ( $\eta_{\text{water}} \approx 10^{-2}$  P).

The total viscosity is the result of several contributions :

- the viscosity of the network due to deformation of the polymer chains,
- the shear viscosity of the solvent,
- the friction term which arises from the relative movement of solvent and solute : this term can be ruled out since solvent and solute move in phase in the modes we consider.

The viscosity we measure seems very likely to be, under these conditions, the viscosity of the polymer network :

$$\eta_p \approx 1 \text{ P.}$$

The experimental conditions of measurement on three pulses show that this value should be taken only as an order of magnitude.

3.3 CONTINUOUS WAVES IN THE DISC. — The disc of gel is maintained directly between the same two transducers. The lateral surface is kept free. The emitting transducer is driven by a continuous wave and the transmitted amplitude of sound is measured as a function of frequency : maxima in the signal are observed for sound frequencies for which the sample has a length of  $n \frac{\lambda}{2}$ ,  $n = 1, 2, 3, \dots$  Figure 3 gives

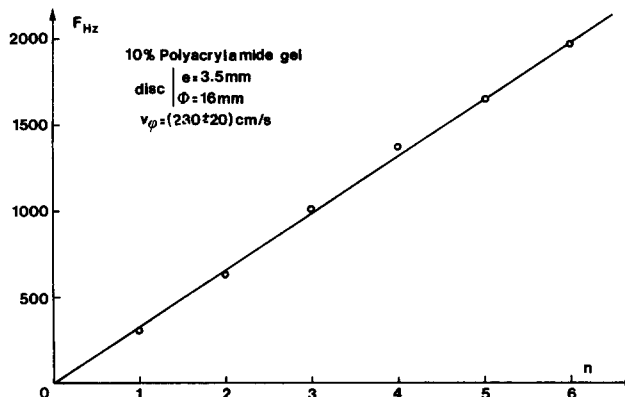


Fig. 3. — Experimental dispersion curve of the dilatational mode obtained from CW method in 10% polyacrylamide gel. The resonance frequencies of the sample are plotted against the order of excited harmonics. The first harmonic corresponds to a wavelength about 7 mm.

the resonance frequency as a function of  $n$ . It is possible to deduce a phase velocity

$$v_\phi = (230 \pm 20) \text{ cm/s}$$

which shows no dispersion within our experimental accuracy.

4. Discussion. — The phase velocity measurements are made between 200 and 2000 Hz ; the corresponding wavelengths are between 1 and 0.1 cm and therefore the ratio  $R/\lambda$  varies between 0.8 and 8. In this region the expected variations of  $v_\phi$  are less than 5% (see [9, 10] and our figure 1a). Our experimental results which give a dispersionless velocity with an accuracy of 10% are in agreement with the theory.

For our group velocity measurement made with  $R/\lambda \approx 0.8$ , it is expected that  $v_{gr} \approx 0.85 v_R \approx 0.85 v_\phi$ . Our experimental result (which is less accurate than for  $v_\phi$ ) is compatible with the theoretical prediction. Taking

$$v_\phi = 0.55 \sqrt{\frac{3 \mu_p}{\rho}} = 230 \text{ cm/s,}$$

Table I. — Comparison between the different elastic coefficients of the solvent, the polymer network and the whole gel.

	Compressibility modulus $K$	Young modulus $E$	Poisson ratio $\sigma$	Shear modulus $\mu$	Longitudinal elastic constant $C_{11}$
Solvent	$K_s$	0	$\approx \frac{1}{2}$	0	$K_s$
Polymer	$\frac{2}{3} \mu_p$	$2 \mu_p$	0	$\mu_p$	$2 \mu_p$ <sup>(c)</sup>
Gel	$K_s + \frac{2}{3} \mu_p \approx K_s$	$3 \mu_p$ <sup>(b)</sup>	$\approx \frac{1}{2}$	$\mu_p$	$K_s + 2 \mu_p \approx K_s$ <sup>(a)</sup>

<sup>(a)</sup> Measured in reference [1].

<sup>(b)</sup> These measurements and static measurement [3, 13].

<sup>(c)</sup> Elastic constant measured in light diffusion experiment [3, 13, 14].

we find

$$\mu_p = \mu_g = (5.8 \pm 0.3) 10^4 \text{ dynes/cm}^2.$$

Static measurement in a 10 % polyacrylamide gel [3] has given  $E \approx 20 \times 10^4 \text{ dynes/cm}^2$ . Our own measurements give

$$E = 3 \mu_g = (17.5 \pm 1) 10^4 \text{ dynes/cm}^2.$$

These two results are in agreement.

The dispersion curves drawn in figure 1a and b are relative to a mode which has no nodal cylindrical surface. Other modes exist which correspond to vibration with one or more nodal cylindrical surfaces and have different dispersion curves [9, 11]. But a uniform excitation on the end surface is not likely to excite these modes which correspond to a very non-uniform distribution of displacements over the cross-section of the cylindrical rod [11].

In table I, we recapitulate the various elastic coefficients involved in a gel and we indicate which experimental method allows one to obtain some of them.  $\mu_p$  which is a particularly important parameter of the gel, has been measured in our experiment. It may also be obtained by light scattering experiments [3, 13, 14].

**5. Conclusion.** — We have reported on a simple experimental method which gives the shear modulus  $\mu_p$  and the polymer network viscosity of a gel.

Other methods could give the same type of information. For instance, transverse bulk elastic wave propagation may provide values of  $\mu_p$  and  $\eta_p$  but it is more complicated to achieve. On the other hand, static experiments measure  $\mu_p$  but are unable to give the viscosity. It seems that our technique has some advantages over the above methods.

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