**Supplementary data**

# Stress amplification and relaxation imaging around cracks in nanocomposite gels using ultrasound elastography

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## Theoretical background

Many theoretical works have been devoted to the expression of the propagation velocity of elastic waves in a stretched non-linear elastic soft solid by revisiting the seminal work of Landau and Lifshitz (Landau and Lifshitz, 1959). Here, we summarize the general equations.

### Mechanics of a continuous medium

A disturbance in a medium is represented by a particle displacement with time dependent position u(x,t). The general equation of motion is given by:

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \frac{\partial \sigma_{ij}}{\partial x_j} \tag{S1}$$

with $\rho$, $\sigma$ and $\frac{\partial^2 u_i}{\partial t^2}$ designate the density, the second Piola-Kirchhoff stress tensor and the particle acceleration, respectively.

The Piola-Kirchhoff stress tensor is given by:

$$\sigma_{ij} = \frac{\partial W}{\partial\left(\frac{\partial u_i}{\partial x_j}\right)} \tag{S2}$$

with W, the strain energy density. In a general elastic medium, the strain energy density developed to the third order is:

$$W = \mu e_{ik}^2 + \frac{\lambda}{2}e^2 + \frac{1}{3}Ae_{ik}e_{il}e_{kl} + Be_{ik}^2 e + \frac{1}{3}Ce^3 \tag{S3}$$

where $e$ is the Lagrangian finite-strain tensor:

$$e_{ik} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} + \frac{\partial u_l \partial u_l}{\partial x_i \partial x_k}\right) \tag{S4}$$

Also defined as the Lagrangian finite strain or Green-Lagrange strain tensor:

$$e = \frac{1}{2}(F^T F - I) \tag{S5}$$

with $C = F^T F$, being the right Cauchy-Green deformation tensor, which is commonly used in materials science. So, $e$ is a measure of how much C differs from identity I. In purely homogeneous deformations, the deformation gradient F has a diagonal representation: $F = \mathrm{diag}[F_{11}, F_{22}, F_{33}] = \mathrm{diag}[\lambda_1, \lambda_2, \lambda_3]$.

**Non-linear acoustoelasticity (AE): elastography of soft solids under stress**

In contrast to a linear analysis, where we may apply a linear strain measure (engineering strain) for infinitesimal deformation, a finite strain measure is used to represent local deformations in a large deformation nonlinear analysis. In this particular case, we use the full equation of the strain energy, eq. S3, and the tensors are no longer symmetrical.

Landau and Lifshitz have established the general expression for the elastic energy density of an isotropic body in the third-order approximation introducing the third-order coefficients (A, B, C) (Landau and Lifshitz, 1959).

From the components of a symmetrical tensor of rank two, we can form two quadratic scalar ($\varepsilon_{ik}^2$ and $\varepsilon^2$) and three cubic scalar ($\varepsilon^3$,$\varepsilon\varepsilon_{ik}^2$, $\varepsilon_{ik}\varepsilon_{il}\varepsilon_{kl}$. Hence, the most general scalar containing terms quadratic and cubic in $\varepsilon_{ik}$, with scalar coefficient (since the body is isotropic), is:

Catheline et al. (Catheline, Gennisson, et Fink 2003) have expressed the elastic strain energy density as function of the invariants and develop to the fourth-order with the fourth-order coefficients (E, F, G, H):

$$W = \mu I_2 + \frac{\lambda}{2} I_1^2 + \frac{1}{3} A I_3 + B I_1 I_2 + \frac{1}{3} C I_1^3 + E I_1 I_3 + F I_1^2 I_2 + G I_2^2 + H I_1^4 \quad \text{(S6)}$$

with the invariants of the Lagrangian (Green-Lagrange) strain tensor defined as follow:

$$I_1 = tr(e) \qquad I_2 = tr(e^2) \qquad I_3 = tr(e^3) \quad \text{(S7)}$$

The Lamé and the Landau coefficients as a function of the elastic moduli in the Voigt's notations are:

- $\lambda = c_{12}$
- $\mu = c_{66}$
- $A = 4c_{456}$
- $B = c_{144}$
- $C = c_{123}/2$ etc.

From the articleS99 of (Catheline et al. 2003). $c_{ij}$and $c_{ijk}$ designate the component of the second-order elastic tensor and the third-order elastic tensor, respectively.

It is possible to simplify equation S6, using the logarithmic strain tensor $\bar{e}$ defined as:

$$\bar{e} = \frac{1}{2} ln(\delta + 2e) \quad \text{(S8)}$$

with the invariants:

$$i_1 = tr(\bar{e}) \qquad i_2 = tr(\bar{e}^2) \qquad i_3 = tr(\bar{e}^3) \quad \text{(S9)}$$

For incompressible material, det(F) = 1.

We can then define the determinant of the finite strain tensor as: $\det(\mathbf{e}) = \frac{1}{2}\left[det(F^T F) - 1\right] = 0$. Now, using the well-known identity log(det(A)) = tr(log(A)), the incompressibility condition can be easily exffpressed in terms of $i_1$:

$$i_1 = \mathrm{tr}(\bar{e} = \frac{1}{2}\mathrm{tr}(ln(\delta + 2e) = \frac{1}{2}\ln(\det(\delta + 2e = \frac{1}{2} ln(1 + 2det(e)) = \frac{1}{2} ln(1) = 0.$$

For the sake of simplicity, the following calculations will be shown to the third order terms of the strain energy function.

The Lagrangian strain tensor as a function of the logarithmic strain tensor can be expressed using the Taylor series of the exponential function:

$$e = \bar{\mathrm{e}} + \bar{\mathrm{e}}^2 + \frac{2}{3}\bar{\mathrm{e}}^3 \tag{S10}$$

and its invariants can be expressed as:

$$I_1 = i_1 + i_2 + \frac{2}{3}i_3 \qquad I_2 = i_2 + 2i_3 \qquad I_3 = i_3 \tag{S11}$$

It follows that the third-order expansion of W in terms of the invariants of the logarithmic strain $\bar{\mathrm{e}}$ reads:

$$W = \mu i_2 + \frac{\lambda}{2}i_1^2 + \left(\frac{A}{3} + 2\mu\right)i_3 + (B + \lambda)i_1 i_2 + \frac{1}{3}Ci_1^3 \tag{S12}$$

which, if we take into account that $i_1 = 0$, equals to:

$$W = \mu i_2 + \left(\frac{A}{3} + 2\mu\right)i_3$$

or

$$W = \mu(i_2 + 2i_3) + \left(\frac{A}{3}\right)i_3$$

$$W = \mu I_2 + \frac{A}{3}I_3$$

If we extend this analysis to include fourth-order terms in the strain energy functions, it leads us to the famous expression of Hamilton and Zabolotskaya (Hamilton et al. 2004; Zabolotskaya et al. 2004):

$$W = \mu I_2 + \frac{1}{3}AI_3 + DI_2^2 \tag{S13}$$

with $D = G + B + \frac{\lambda}{2}$, from equation S6.

According to Hamilton *et al.* (Hamilton, Ilinskii, et Zabolotskaya 2004), the A constant determines the nonlinear shear stress (/or behavior) while D is required to describe nonlinear distortion of shear waves in finite amplitudes.

Finally, Gennisson and colleagues (Gennisson et al. 2007) developed eq. S13 to retrieve the shear wave speed when the material is subjected to a stress. We expanded these equations to the 4th order:

$$\rho v_{21}^2 = \mu_0 - \sigma_{22}\left(\frac{A}{12\mu_0}\right) + \sigma_{22}^2(2\mu_0 + A + 3D)\frac{1}{9{\mu_0}^2} \quad \text{(S14)}$$

$$\rho v_{13}^2 = \mu_0 + \sigma_{22}\left(1 + \frac{A}{6\mu_0}\right) + \sigma_{22}^2\left(5\mu_0 + \frac{7}{4}A + 3D\right)\frac{1}{9{\mu_0}^2} \quad \text{(S15)}$$

$$\rho v_{12}^2 = \mu_0 - \sigma_{22}\left(1 + \frac{A}{12\mu_0}\right) + \sigma_{22}^2\left(5\mu_0 + \frac{7}{4}A + 3D\right)\frac{1}{9{\mu_0}^2} \quad \text{(S16)}$$

$$\rho v_{12}^2 = \mu_0 + \sigma_{22}\left(1 + \frac{A}{12\mu_0}\right) + \sigma_{22}^2\left(5\mu_0 + \frac{7}{4}A + 3D\right)\frac{1}{9{\mu_0}^2} \quad \text{(S17)}$$

The first index corresponds to the direction of the shear displacement induced by radiation force (direction of polarization of the shear wave) while the second index corresponds to the axis of propagation of the shear wave (see Fig. 2). One can notice when σ = 0, at rest, we retrieve the direct dependence of μ on the shear wave velocity.

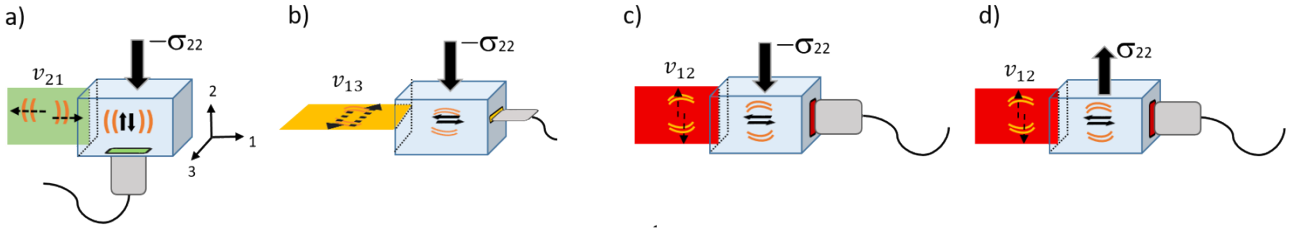



Figure T1. The three possible configurations to generate shear wave in the material. Acoustic radiation pressure is used to create the shear wave and ultrafast imaging to detect the propagation of polarized shear waves in the phantom under uniaxial stress $\sigma_{22}$. The three resulting shear waves are designated as a) 21 or23, b) 13 or 31, c) 12 or 32.

Note that if we consider a tensile test with a stress applied in the direction 2, $\sigma_{22}$,with faces in axis 1 and 3 free to deform, we can write the right Cauchy-Green deformation tensor, according to the incompressibility assumption:

$$C = \begin{pmatrix} \lambda_1^2 & 0 & 0 \\ 0 & \lambda_2^2 & 0 \\ 0 & 0 & \lambda_3^2 \end{pmatrix} = \begin{pmatrix} \frac{1}{\lambda} & 0 & 0 \\ 0 & \lambda^2 & 0 \\ 0 & 0 & \frac{1}{\lambda} \end{pmatrix} \quad \text{(S18)}$$

with $\lambda = \frac{L}{L_0}$, with $L_0$, the initial length and L, the length at stretch state.

It comes that the Green-Lagrange strain tensor invariants in tension can be written as:

$$\begin{aligned} I_1 &= tr(e) = \frac{1}{2}\left(\lambda^2 + \frac{2}{\lambda} - 3\right) \\ I_2 &= tr(e^2) = \frac{1}{4}\left[(\lambda^2 - 1)^2 + 2\left(\frac{1}{\lambda} - 1\right)^2\right] \\ I_3 &= tr(e^3) = \frac{1}{8}\left[(\lambda^2 - 1)^3 + 2\left(\frac{1}{\lambda} - 1\right)^3\right] \end{aligned} \quad \text{(S19)}$$

From eq. S6, it comes that the strain energy density function can be written in tension as:

$$W = \mu\frac{1}{4}\left[(\lambda^2 - 1)^2 + 2\left(\frac{1}{\lambda} - 1\right)^2\right] + \frac{A}{24}\left[(\lambda^2 - 1)^3 + 2\left(\frac{1}{\lambda} - 1\right)^3\right] + \frac{D}{16}\left[(\lambda^2 - 1)^2 + 2\left(\frac{1}{\lambda} - 1\right)^2\right]^2 \quad \text{(S20)}$$

From the strain energy density function and using eq. S2 ($\sigma_{22} = \partial W/\partial\lambda$), we can express the stress as a function of $\lambda$:

$$\sigma_{22} = \mu\left[\lambda(\lambda^2 - 1) - \frac{1}{\lambda^2}\left(\frac{1}{\lambda} - 1\right)\right] + \frac{A}{4}\left[\lambda(\lambda^2 - 1)^2 - \frac{1}{\lambda^2}\left(\frac{1}{\lambda}\right.\right. \quad \text{(S21)}$$

For the most elastic incompressible material we have (swollen elastic gel at equilibrium), the stress calculated from the invariants matches well with the experimental true stress given by Instron.

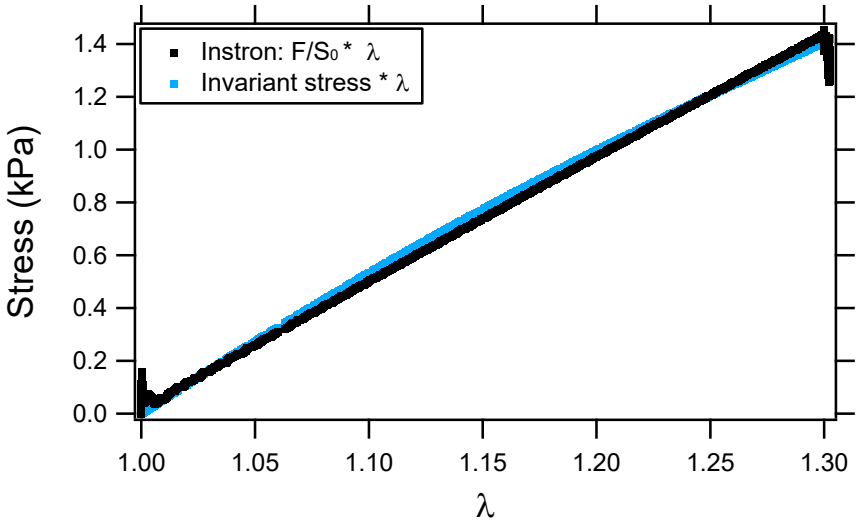
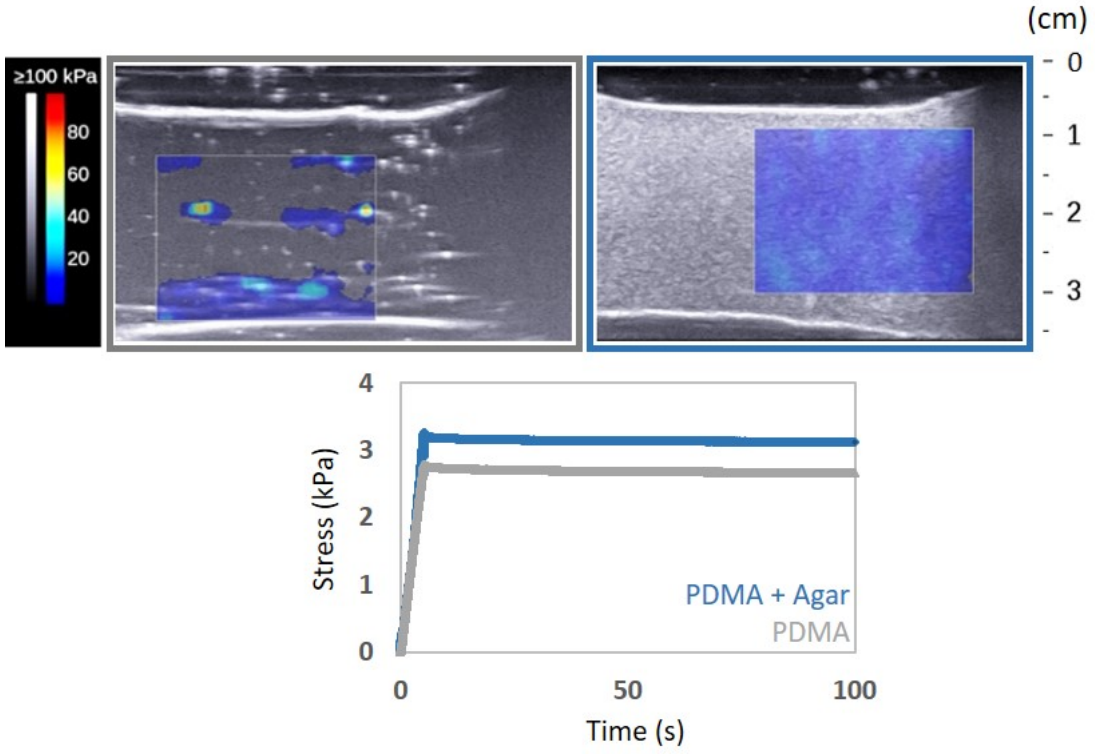
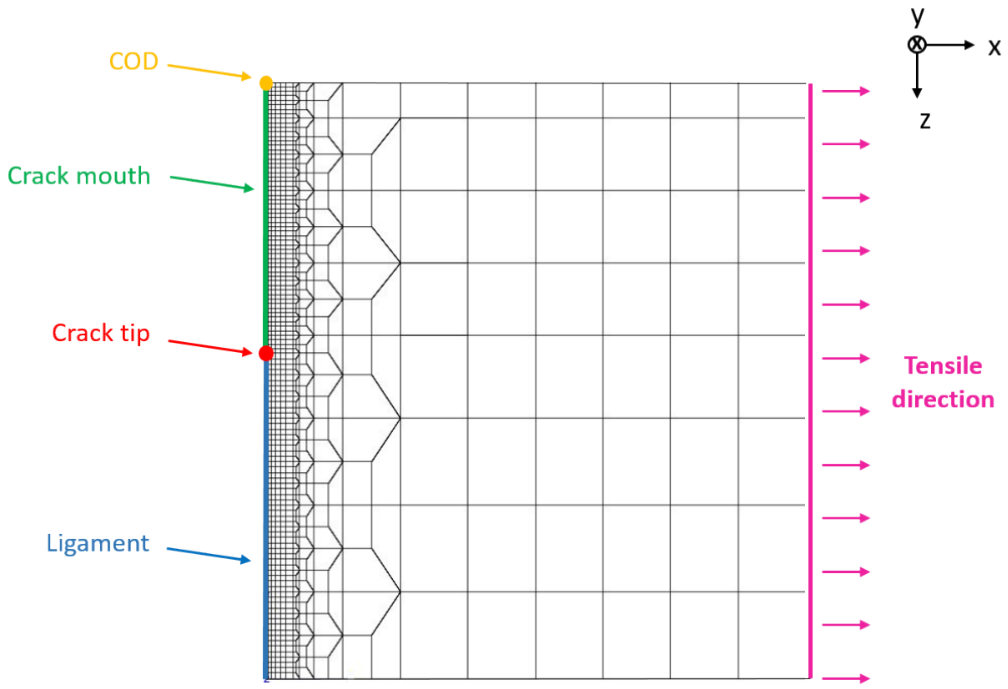



Figure T2. Experimental stress and stress calculate from the strain energy density function as a function of $\lambda = 1.3$. The model fits well for $\lambda = 1.3$.

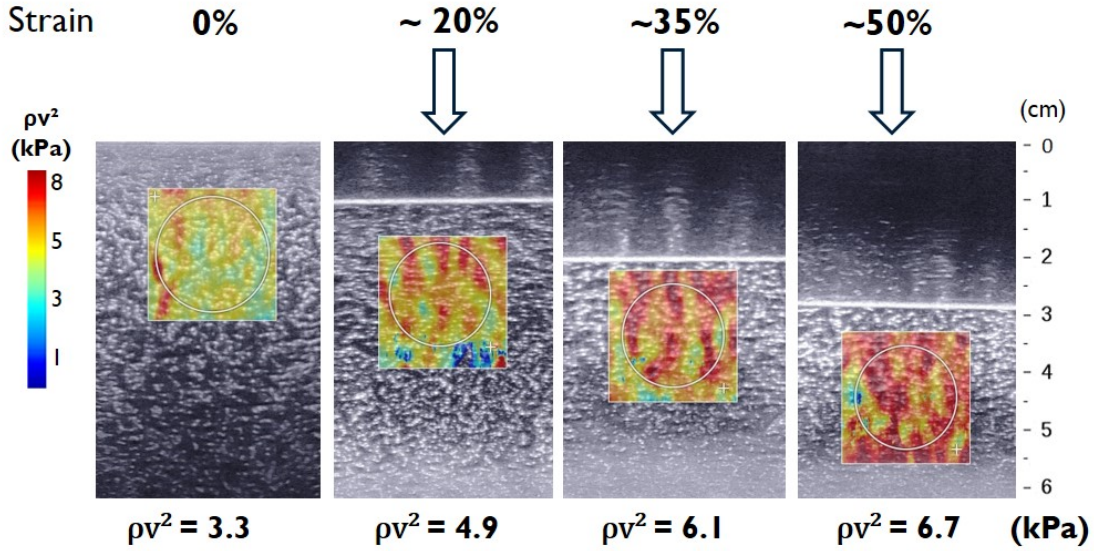
# Supplementary Figures



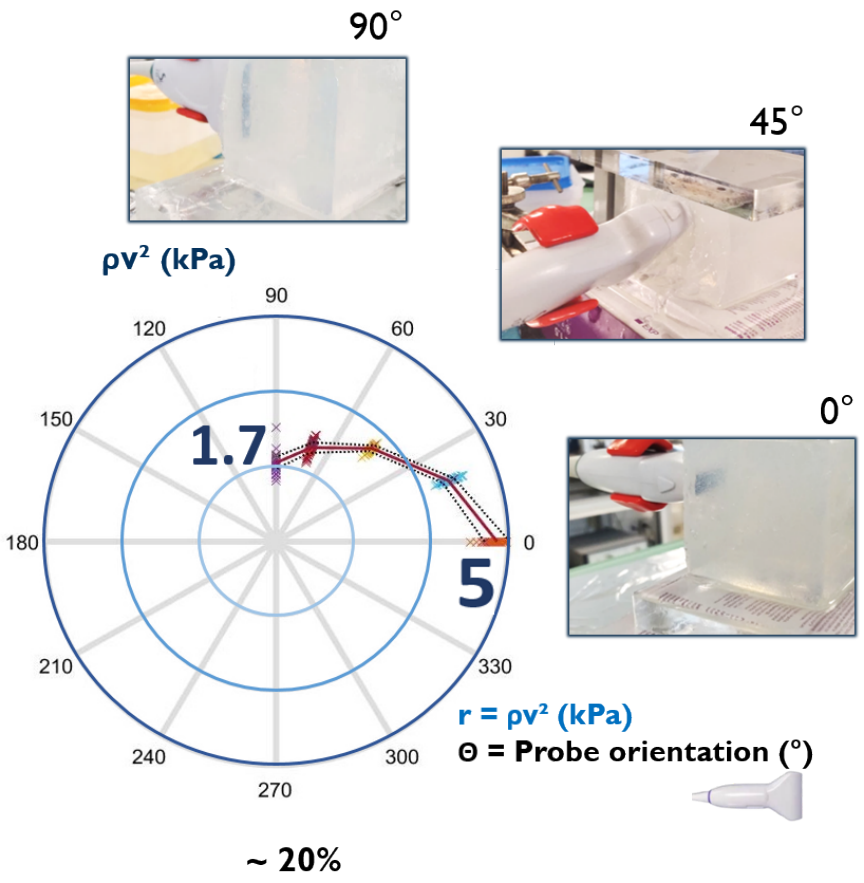

**Supplementary Figure S1**. Importance of agar addition in elastic gel. Left: Elastic gel without agar. Right: Elastic gel with 1 wt.% of agar. Agar increases echogenicity, the signal is better, homogeneous. Color bar represents the Young's modulus (kPa). Bottom: Tensile test until 30% strain. Agar does not change the mechanics.




**Supplementary Figure S2.** Meshing of the SENT specimens indicating the crack tip, the ligament, and the node sets where boundary conditions and symmetry were applied. The ligament in blue solid line was blocked in the x-direction due to symmetry.



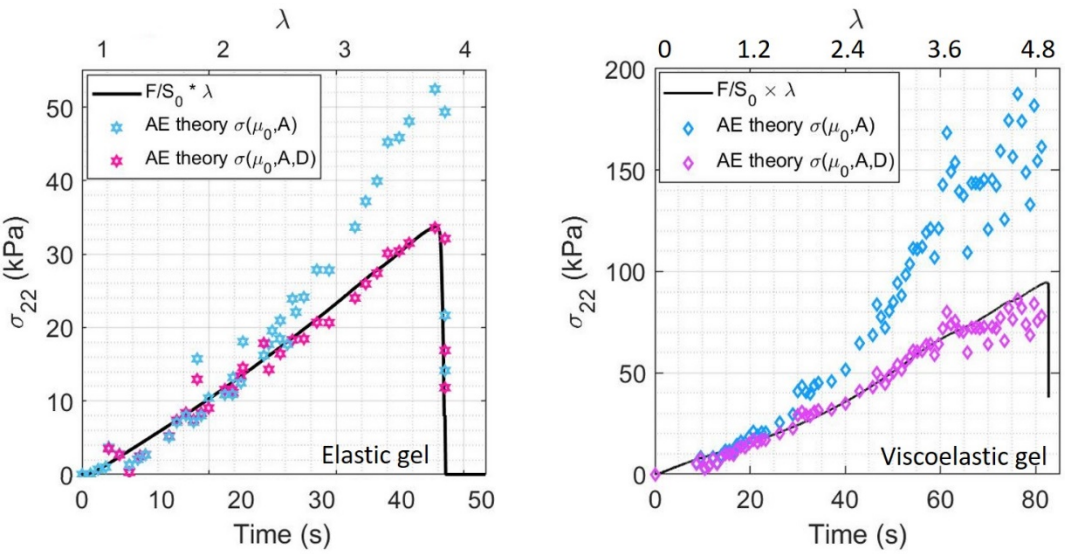

**Supplementary Figure S3.** Compression tests with different strain applied on the cubic gel. The shear wave velocity depends on the stress applied.



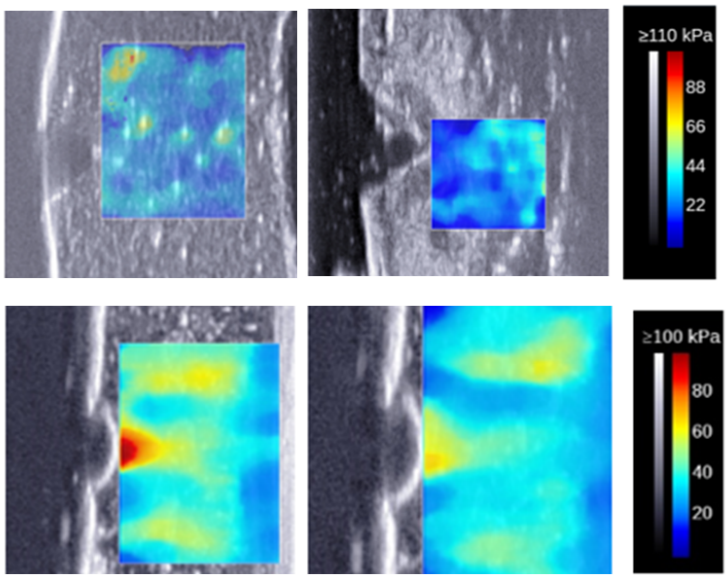

**Supplementary Figure S4.** Polar plot showing five different probe position during a compression test at a fixed 20% strain. Nonlinear effects are significant since the value of $\rho v^2$ is multiplied by 3 between 0 and 90°. Shear wave velocity depends on the probe orientation when the gel is under stress.

**Supplementary Table S1.** Landau coefficients $\mu_0$, A and D from compression and tensile tests for elastic (0.5 vol.% SiO2) gel.

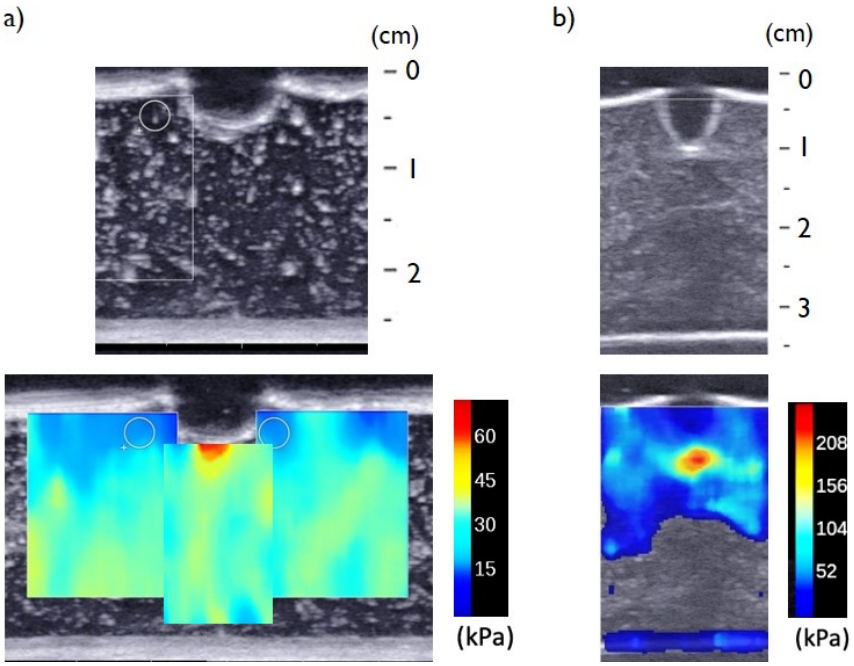
| (kPa) | at rest $\mu_0$ | Propagation index 12 A / D | Propagation index 21 A / D | Propagation index 13 A / D | Mean value |
|---|---|---|---|---|---|
| Compression | 4.2 ± 0.1 | -21.4 / 4.7 | -16.4 / 2.2 | -19.0 / 5.5 | -18.9 / 4.1 |
| Tension | 4.0 ± 0.2 | -18.9 / 7.3 | - | - | -18.9 / 7.3 |



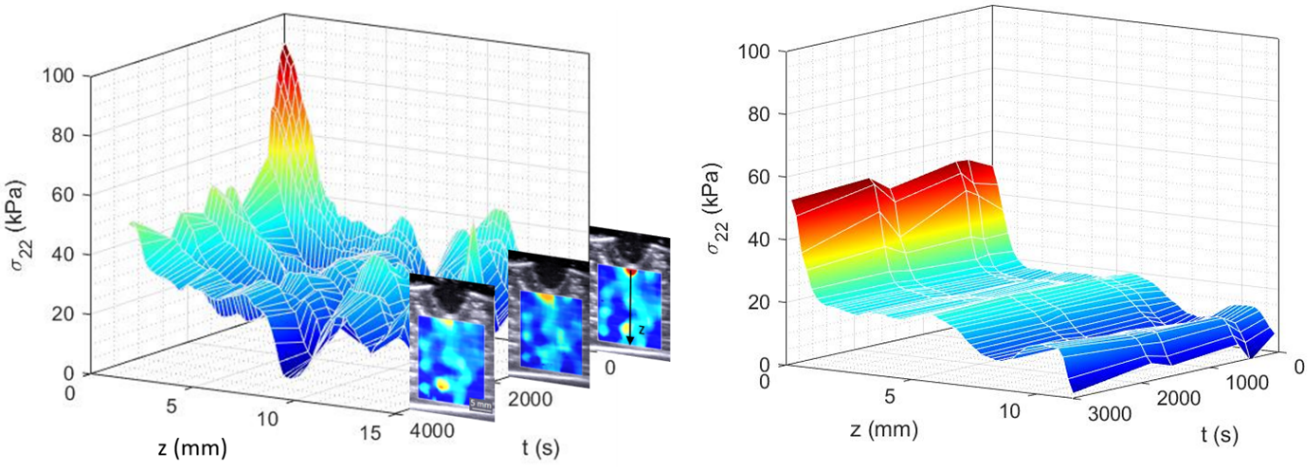

**Supplementary Figure S5.** Tensile test until fracture for elastic (0.5 vol.% SiO2) (left)/viscoelastic (10 vol.% SiO2) (right) gel. AE theory has been used to retrieve the macroscopical stress obtained from the mechanical tensile tool. It appears that the third order is no more valid when a stress of σ ≈ 20 kPa is reached.



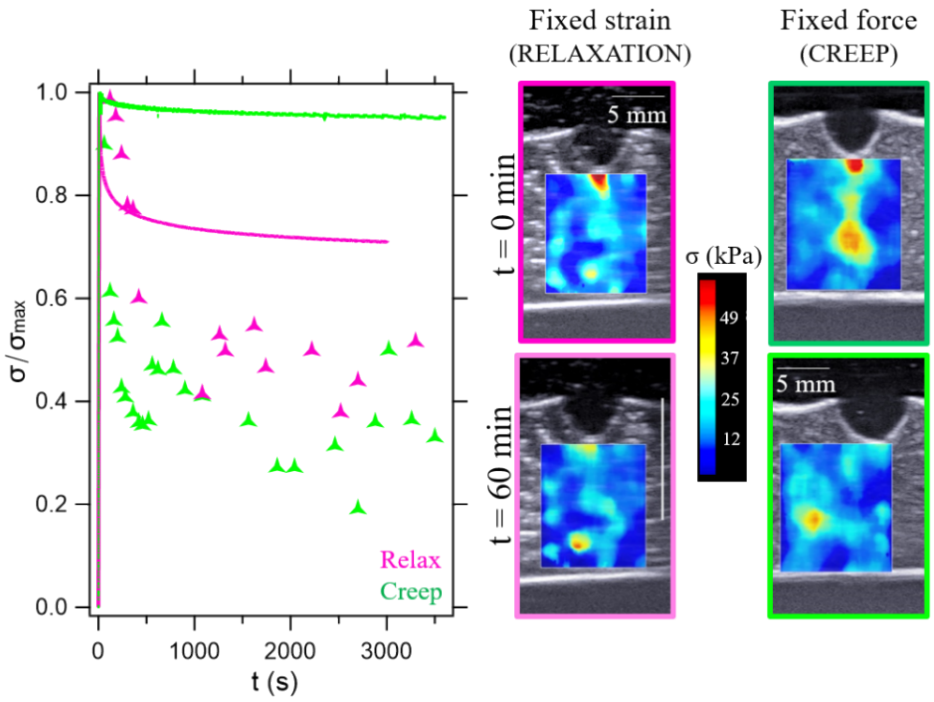

**Supplementary Figure S6**. Viscoelastic gel (with NP). Top: No stress is applied; the gel is cut to reproduce an open notch, i.e. V-shape. The map is homogeneous anywhere in the gel.
Bottom left: Singularity at t = 0. Bottom right: Singularity at t = 45 min. The stress amplification disappears proving that a physical phenomenon happens, i.e. network rearrangement at NP surface. Color bar is a scale for the Young's Modulus.




**Supplementary Figure S7.** Delimitation of the area of interest. The singularity appears at the crack tip but not around the crack tip, at the crack tip lips. Color bar is a scale for the Young's Modulus. a) Gel with 10 vol.% SiO2. b) Gel with 15 vol.% SiO2. Sometimes, for unknown reasons, it appears that there is a lack of signal inside the gel.




**Supplementary Figure S8.** 3D representation of the crack tip of a hybrid gel (left) et elastic gel (right). For the elastic gel, the stress is constant in time, there is no stress reorganization. In hybrid gel, stress is dissipated inside the gel, the singularity is erased, and fracture is avoided.




**Supplementary Figure S9.** Relative macroscopic (line) and local (triangle) stress variation during a relaxation and creep test in a 10 vol.% silica NPs.