How natural rubber latex gels deform under stress: a study using a rheo-ultrasonic technic

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Context

Natural rubber latex (NRL), extracted from Hevea brasiliensis tree, coagulate by addition of an acid and formed a colloidal gel. Such gels present a strain hardening behavior if submitted to large amplitude oscillatory deformation. Whether this irreversible strain hardening occurs homogeneously in the entire material or propagate from the rotor to the stator is an open question. Here, we used conventional rheometry coupled with USV to study the displacements fields inside the gel during the strain hardening.

Materials & Methods

A prevulcanized NRL suspension (φ = 0.025) was acidified by addition of Glucono-δ-lactone. Oscillatory measurements were performed in a commercial rheometer (TA Instruments ARG2) using a PMMA Couette geometry (R1 = 25 mm, R2 = 23 mm, h = 60 mm) to allow USV measurements with a Ultrasonic 2D-scanner (20 sequences of 3000 pulses sent at 350Hz). Polystyrene particles (Dynoseed TS20µm) were added in the suspension (1%wt) as ultrasonic contrast agent, without any impact on rheological properties. The gel was formed in situ and its viscoelastic properties were monitored for 30 min at f = 1Hz and γ = 0.5%. Then the gel was submitted to increasing oscillatory deformation step by step.

Material response to an oscillatory strain

NRL gels present a irreversible strain hardening under large stress (or strain), and eventually fracture. This behavior is characterized by an increase of G’ and G’’ with the strain.

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Displacements fields inside the gel during the oscillations (stator: r=0 and rotor: r=2)

Velocity profile obtained from USV during the oscillations

Harmonics obtained from Fourier decomposition of the local velocity vs time

Three different velocity profile:

- Sinusoidal (for γ < 10%) : (1st harmonic alone)
- Triangular (for 10% < γ < 80%) : (1st - 3rd - 5th harmonics)
- Second harmonic (for γ > 80%) : (1st-2nd-3rd,... harmonics)

Conclusions

The macroscopic strain hardening behavior is associated with local rearrangements at a microscopic scale.

Local rearrangements are quasi-instantaneous upon strain increase, and homogeneously distributed in the sample.

At high strain deformations, while approaching rupture, the emergence of second harmonics in the signal might be the characteristic of preliminary disruption events.

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