EPFL

Charaterization of viscoelastic materials

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CCMX Advanced Materials Characterization,

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Goal of this course

- Introduction: what is viscoelasticity, where do we see it?
- Definitions
- Theory: simple models, Boltzmann superposition
- Measurement methods: creep, oscillatory mode, other methods with practical examples

Introduction

Viscoelastic materials: materials for which the relation between stress and strain depends on time.

Anelastic/linear viscoelastic solids: a subset of viscoelastic materials that recover fully after removal of a transient load.

Phenomena:

- Cases of constant stress, strain increases with t: creep
- Cases of constant strain, stress decreases with t: relaxation
- Cases where effective stiffness depends on loading rate
- In cyclic loading, the response is not in phase with the solicitation, leading to mechanical energy dissipation.
- Attenuation of acoustic waves
- Incomplete rebound, added friction…

Introduction

Example of viscoelastic materials:

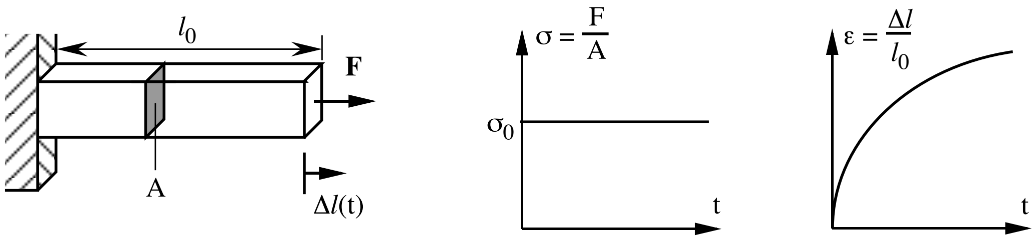
- Metals at high temperature, due to increased diffusion rate, or lead and solders that creep at low temperature
- Polymers, elastomers
- Wood
- Human tissues: ligaments, tendons, bone, skin
- Geological materials

Implications:

- Problems: long term performance of structural materials, change in shape or stress with time…
- Benefits: impact absorption, dampers, shoe soles, sealing plugs,..

Definitions

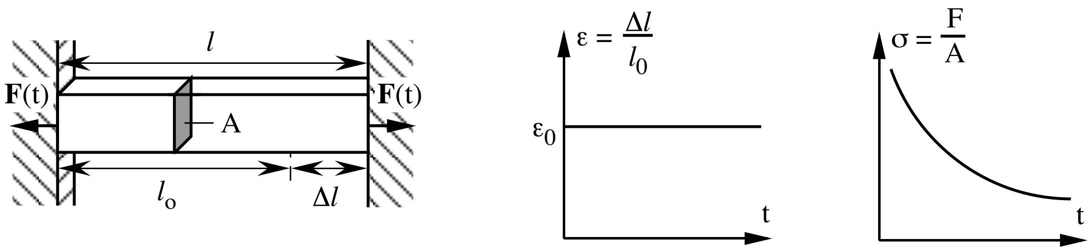
Creep



(a) (b) (c)

$$J(t) = \frac{\varepsilon(t)}{\sigma_0}$$

Relaxation

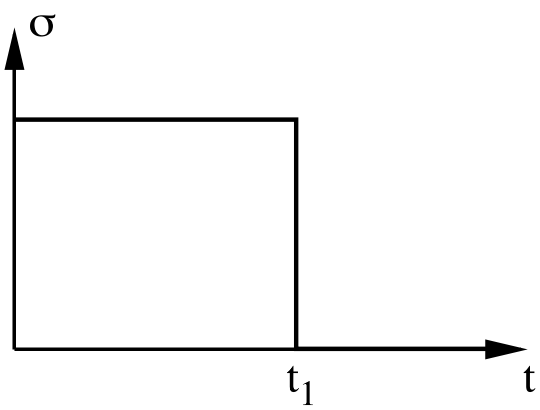


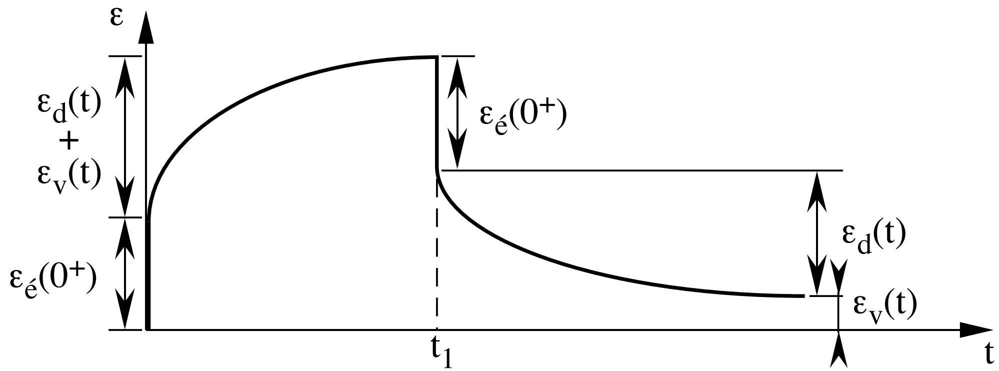
(a) (b) (c)

$$R(t) = \frac{\sigma(t)}{\varepsilon_0}$$

Definitions

Recovery: evolution after the stress (or strain) is removed. For a linear elastic case, the recovery tends to zero after a certain time.





A simple method for creep characterization

Material under constant load:

It is possible to extract a creep modulus, by dividing the stress by strain at time t, to find an experimental function to identify, i.e. a Creep modulus for a given stress level and time.

$E_c(t)=\sigma_0/\varepsilon(t)$

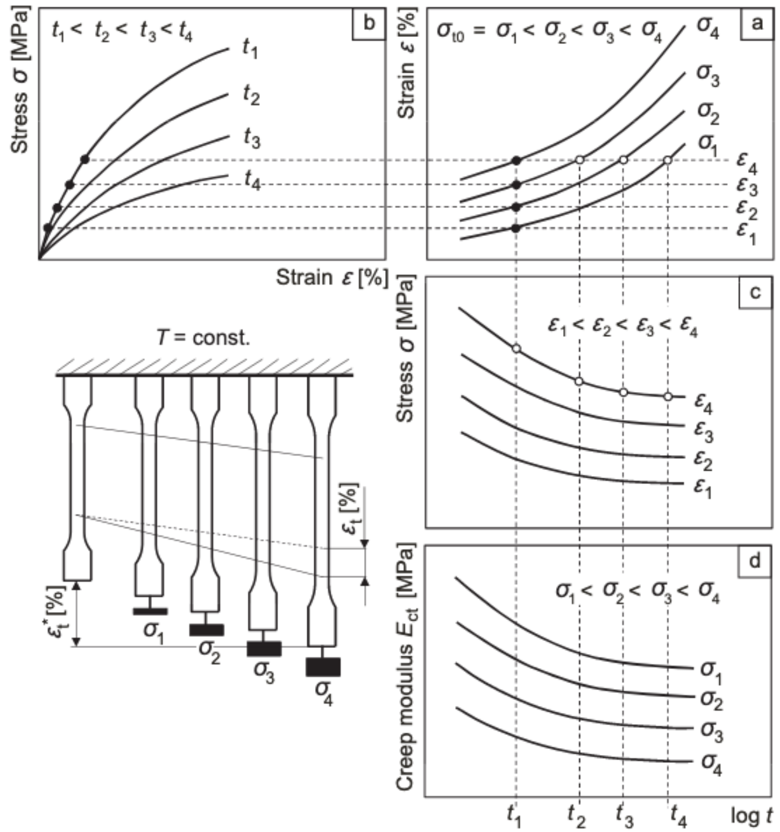


Fig. 4.133. Evaluation of tensile creep tests on polymers according to ISO 899-1.

Long-term loading – Compression Creep Test – Introduction, January 2014, DOI: 10.1007/978-3-642-55166-6_60

- In book: Polymer Solids and Polymer Melts–Mechanical and Thermomechanical Properties of Polymers Christian Bierögel, Wolfgang Grellmann

Example of tensile creep characterization for a Polyamide 6,6

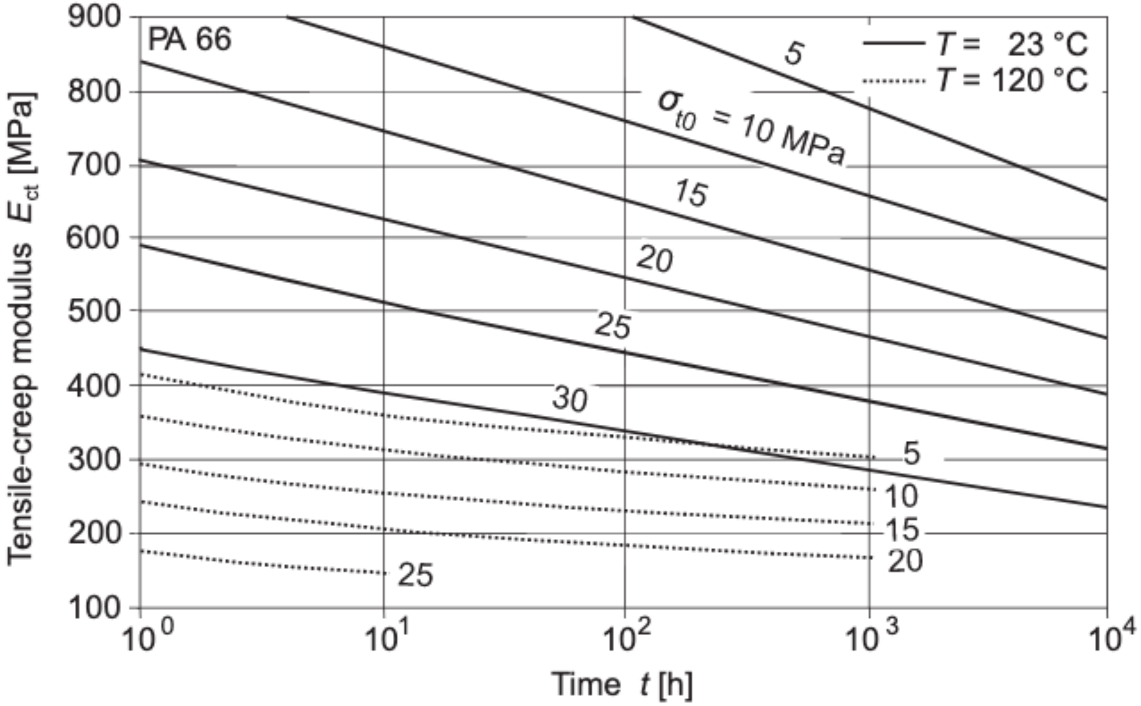


Fig. 4.138. Tensile-creep modulus of polyamide 66 at different temperatures and stress levels [12Els].

Long-term loading – Compression Creep Test – Introduction, January 2014, DOI: 10.1007/978-3-642-55166-6_60

- In book: Polymer Solids and Polymer Melts–Mechanical and Thermomechanical Properties of Polymers Christian Bierögel, Wolfgang Grellmann

Theoretical constitutive equations

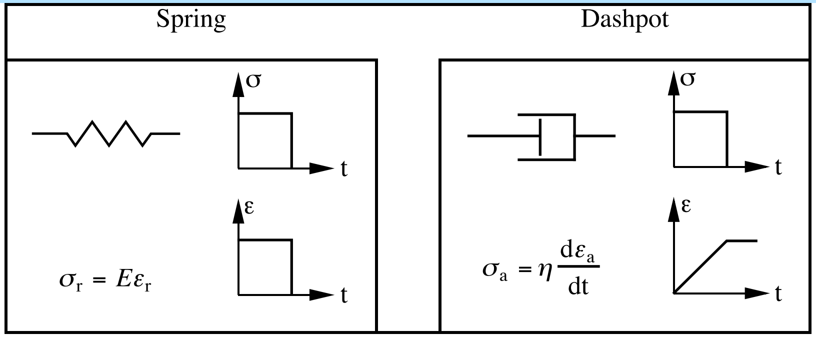
Need to predict and model the viscoelastic behavior: for small deformations, linear viscoelasticity describes well the behavior, with total recovery.

Ideally, we need to have a constitutive equation describing the relation between stress and strain, for the form $P\sigma = Q\varepsilon$

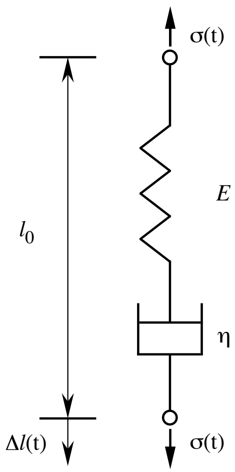
$$P = \sum_{i=0}^{p} a_i \frac{\partial^i}{\partial t^i} \qquad Q = \sum_{i=0}^{q} b_i \frac{\partial^i}{\partial t^i}$$

$$a_0\sigma + a_1\frac{d\sigma}{dt} + a_2\frac{d^2\sigma}{dt^2} + \ldots = b_0\varepsilon + b_1\frac{d\varepsilon}{dt} + b_2\frac{d^2\varepsilon}{dt^2} + \ldots$$

Mechanistic models

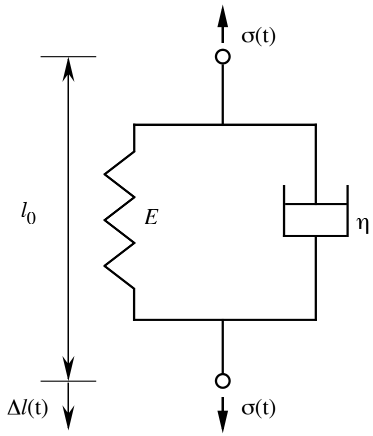


Maxwell model



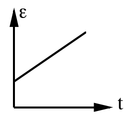
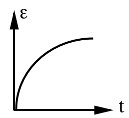
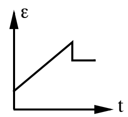
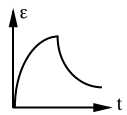
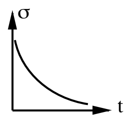
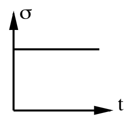
$$\frac{d\varepsilon}{dt} = \frac{1}{E}\frac{d\sigma}{dt} + \frac{1}{\eta}\sigma$$

Kelvin model



$$\sigma = E\varepsilon_r + \eta\frac{d\varepsilon_a}{dt}$$

Mechanistic models

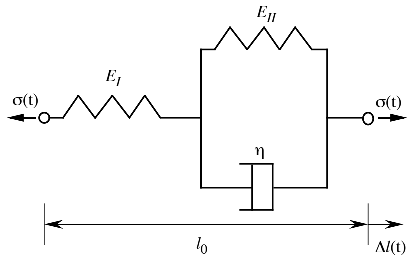
	Maxwell	Kelvin
Creep	$\varepsilon(t) = \frac{\sigma_0}{E}\left(1 + \frac{t}{\tau_\sigma}\right)$  Infinite creep Ok for fluids	$\varepsilon(t) = \frac{\sigma_0}{E}\left[1 - \exp\left(-\frac{t}{\tau_\sigma}\right)\right]$  Finite Creep OK
Recovery	$\varepsilon(t) = \text{constant}$  Dashpot stays deformed Not OK	$\varepsilon(t) = \varepsilon_0 \exp\left(-\frac{t}{\tau_\sigma}\right)$  OK
Relaxation	$\sigma(t) = \sigma_0 \exp\left(-\frac{t}{\tau_\varepsilon}\right)$  Ok, but relaxes to 0 when t is large. OK	$\sigma(t) = \text{constant}$  Elastic element Not OK

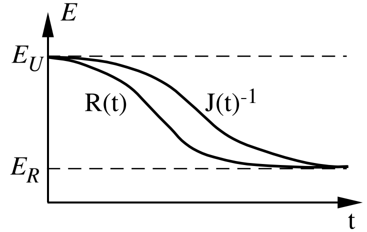
Mechanistic models

Standard Linear Solid Model

$$\sigma + \tau_\varepsilon \dot{\sigma} = E_R \varepsilon + E_I \tau_\varepsilon \dot{\varepsilon}$$

$$\tau_\varepsilon = \frac{\eta}{E_I + E_{II}} \text{ [s] and } E_R = \frac{E_I E_{II}}{E_I + E_{II}} \text{ [Pa]}$$





Creep

$$\varepsilon(t) = \frac{\sigma}{E_I} + \frac{\sigma}{E_{II}}\left[1 - \exp\left(-\frac{t}{\tau_\sigma}\right)\right]$$

$$\tau_\sigma = \frac{\eta}{E_{II}}$$

Compliance function:

$$J(t) = \frac{1}{E_I} + \frac{1}{E_{II}}\left[1 - exp\left(-\frac{t}{\tau_\sigma}\right)\right] = \frac{1}{E_R} + J_v(t)$$

Relaxation

$$\sigma(t) = \varepsilon \frac{E_I E_{II}}{E_I + E_{II}}\left[1 + \frac{E_I}{E_{II}} \exp\left(-\frac{t}{\tau_\varepsilon}\right)\right] = R(t) \cdot \varepsilon$$

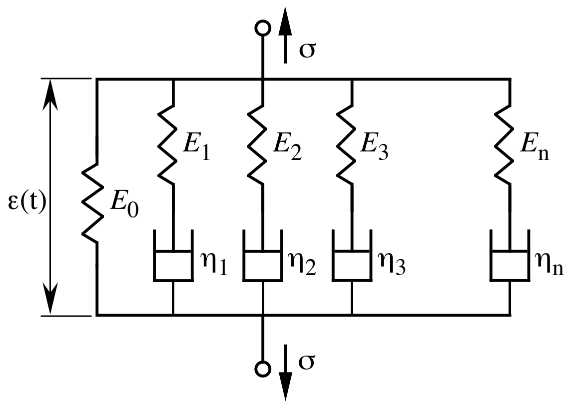
$$\tau_\varepsilon = \frac{\eta}{E_I + E_{II}}$$

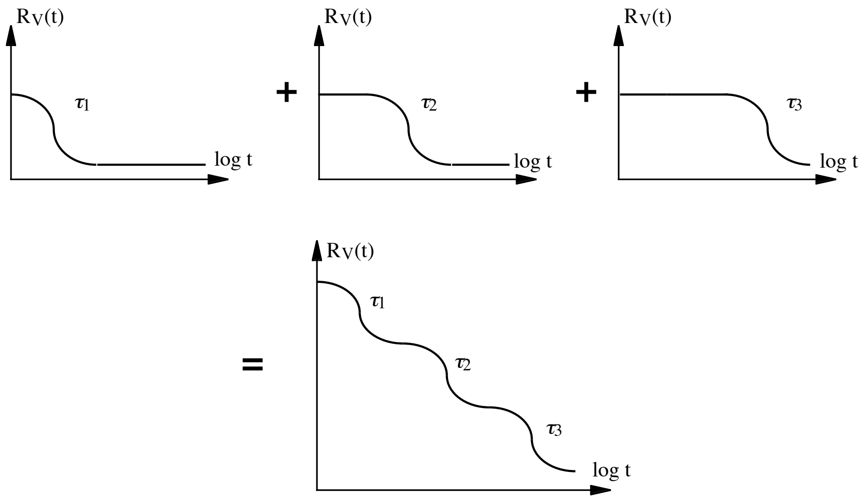
Relaxation function:

$$R(t) = \frac{E_I E_{II}}{E_I + E_{II}}\left[1 + \frac{E_I}{E_{II}} exp\left(-\frac{t}{\tau_\varepsilon}\right)\right] = E_R + R_v(t)$$

Mechanistic models

Any combination of Maxwell or Kelvin elements is also possible

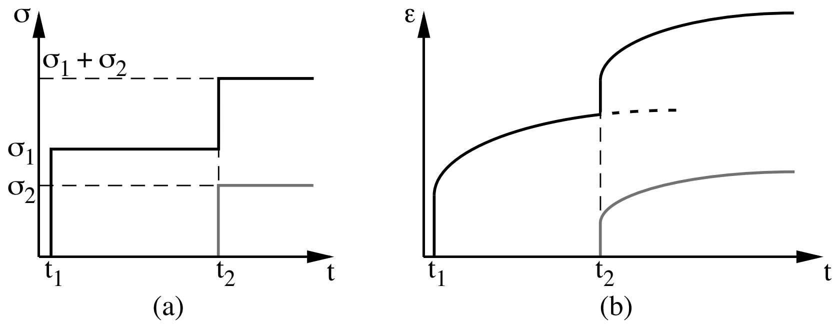




$$\sigma = \sigma_0 \sum_{i=0}^{n} C_i \exp\left(-\frac{t}{\tau_i}\right)$$

Last bit on theory: Boltzmann superposition

Valid for linear viscoelastic materials, small deformations: the effect of a compound cause is the sum of the effects of the individual causes



$\varepsilon(t)=\sigma_1 J(t - t_1)$ for $t_1 < t < t_2$

$\varepsilon(t)=\sigma_1 J(t - t_1) + \sigma_2 J(t - t_2)$ for $t>t_2$

For very small increments:

$$\varepsilon(t) = \int_{-\infty}^{t} J(t-\tau)d\sigma(\tau) = \int_{-\infty}^{t} J(t-\tau)\frac{\partial \sigma(\tau)}{\partial \tau}d\tau$$

Similarly for stress:

$$\sigma(t) = \int_{-\infty}^{t} R(t-\tau)\frac{\partial \varepsilon(\tau)}{\partial \tau}d\tau$$

Summary for the models

For any solicitation in stress, strain is found as:

$$\varepsilon(t) = \frac{\sigma(t)}{E} + \int_{-\infty}^{t} J_V(t-\tau)\frac{\partial\sigma}{\partial\tau}d\tau$$

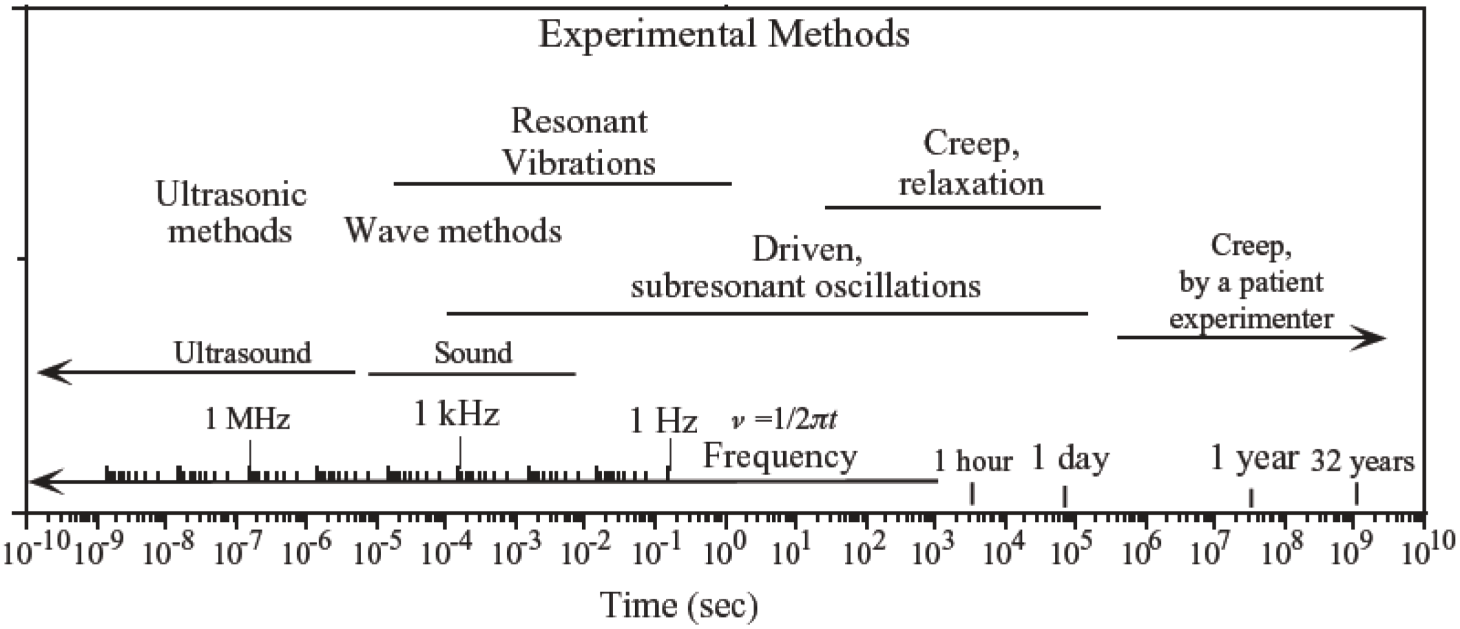
For any solicitation in strain, stress is found as:

$$\sigma(t) = \varepsilon(t)\mathrm{E}_R + \int_{-\infty}^{t} R_v(t-\tau)\frac{\partial\varepsilon}{\partial\tau}d\tau$$

So, for prediction of behavior, we need to identify the functions J and R characterizing the material, then we can take care of all types of material history.

Characterization methods

Mechanical tests are mostly used: force or torque is applied, and strain is measured versus t. The tests are either static, or dynamic with oscillatory stress or strain. To explore wide ranges of time or frequency, some tricks have been devised (time temperature equivalence), or several methods are used.



From Lakes, 2009

Creep, relaxation

- Constant stress applied with calibrated dead weights, levers or hydraulic systems for stiff materials (concrete). Specimens of biological origin or some polymers need to be preconditioned by cycling first to obtain reproducible results.
- Issues: long time needed to cover several decades (11 days for 5 decades), and caution to ensure proper gripping for uniform stress
- Possible to measure nonlinear viscoelasticity: creep at various stress levels, followed by recovery.
- In case of Quasi Linear viscoelasticity, J(t, σ)=J(t) f(σ)
- Strain measurement is performed using LVDT, extensometer or strain gauges

$$\epsilon(t) = \int_0^t J(t-\tau, \sigma(\tau)) \frac{d\sigma}{d\tau} d\tau,$$

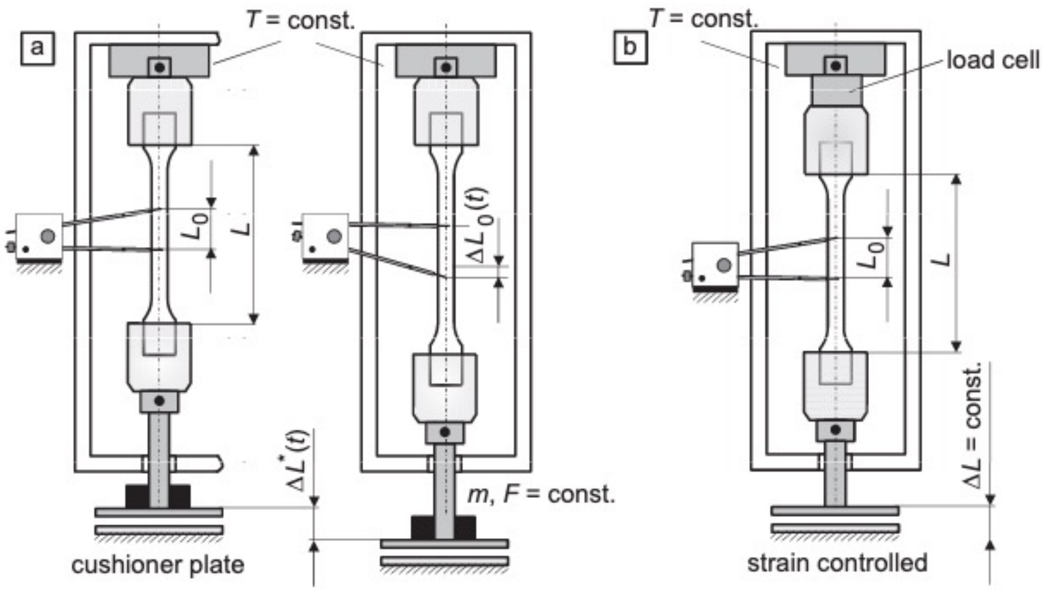
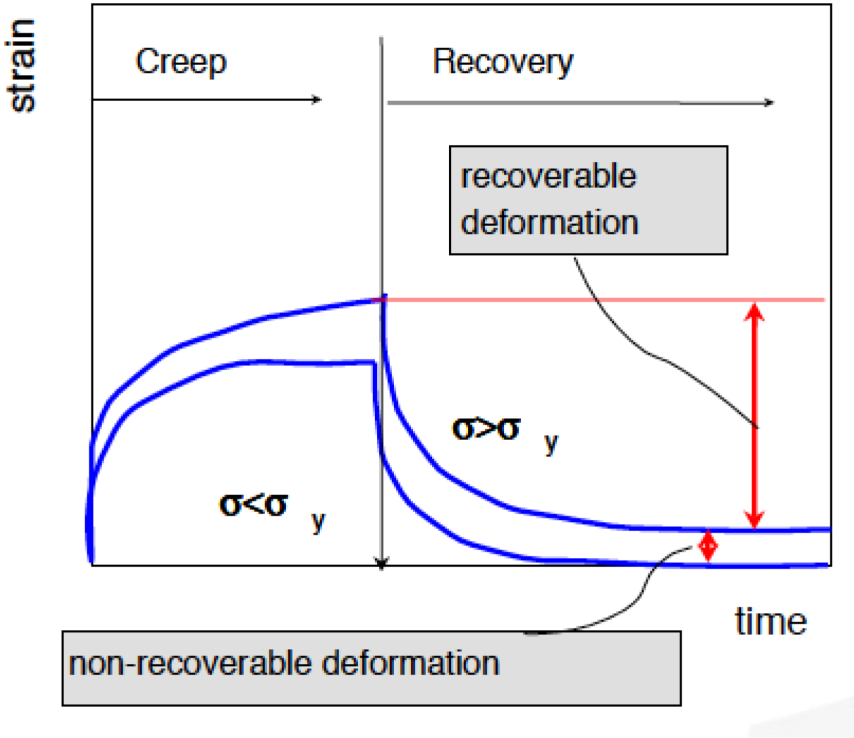


Fig. 4.132. Principle of tensile creep test (a) and stress relaxation test (b) under uniaxial loading.

Creep/Recovery experiments

Above a certain level of stress, the material is irreversibly deformed ("plastic deformation") and the recovery is not full.



Succesive creep/recovery tests are needed to determine the threshold stress

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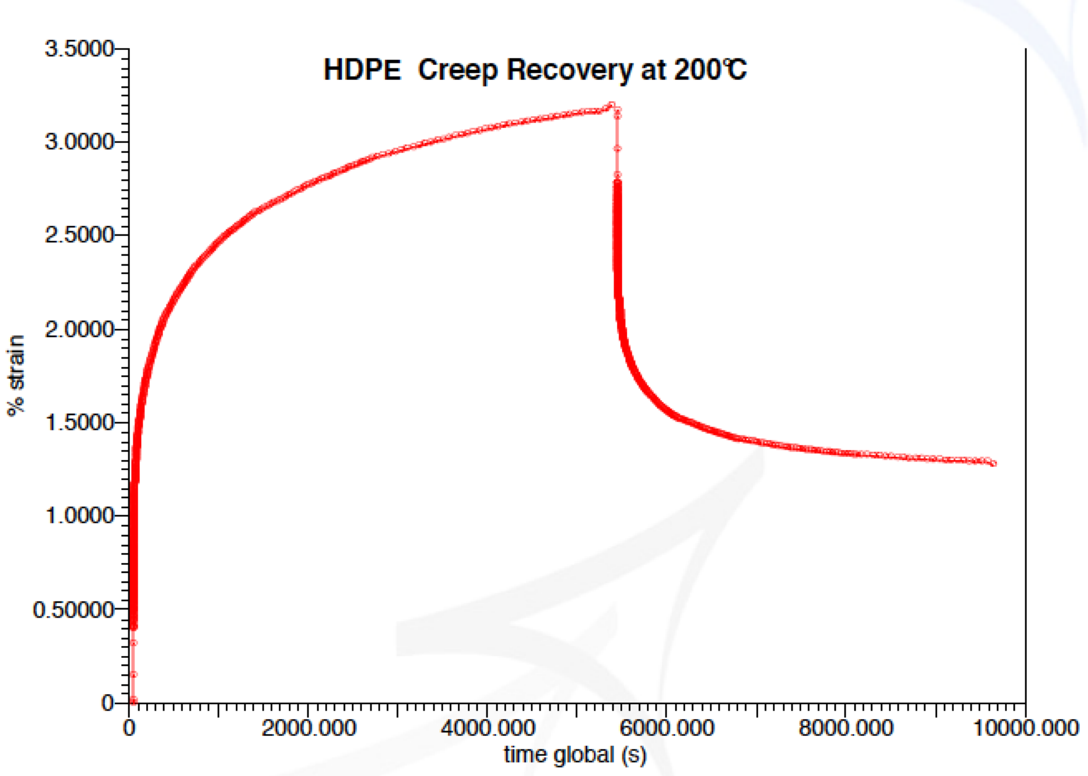
Creep/Recovery experiments

EXAMPLE: HDPE



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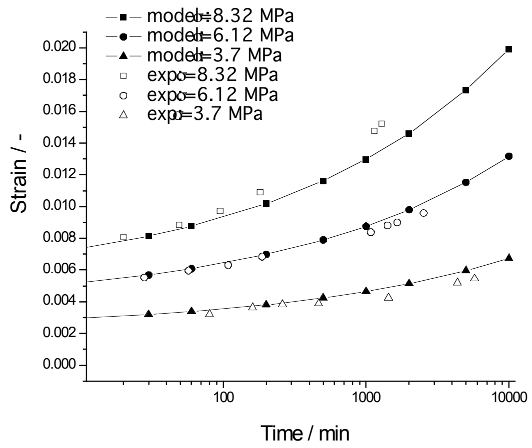
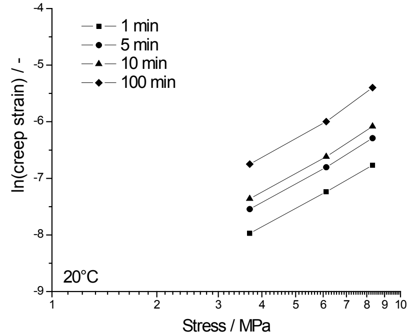
Creep at room temperature

Example: Creep of Polypropylene at room temperature



$$\dot{\varepsilon}_{cr} = A\sigma^n t^m$$

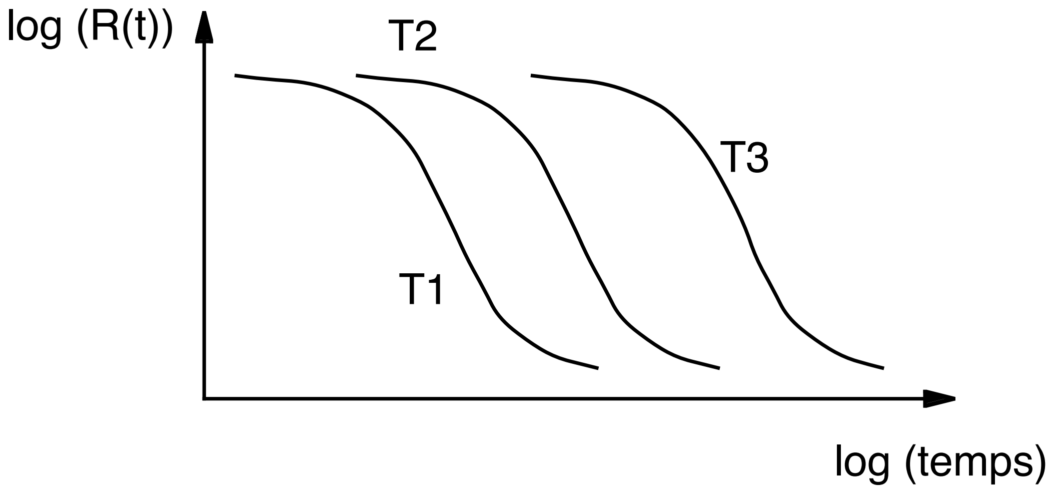


Linear viscoelastic

$\dot{\varepsilon}$ is the creep strain rate, A, m and n are constants, σ is the stress, t is the time. At room temperature, A=1.367 10^{-5}, m=0.72 and n=1.48.

Issues with creep/recovery measurements

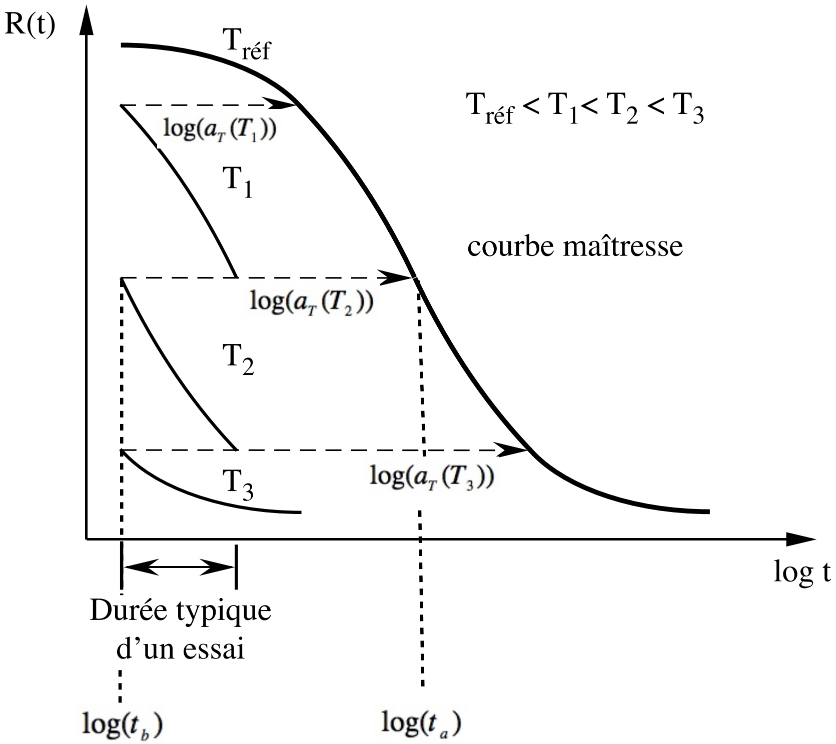
- Find the right stress, if too high, no creep but plastic flow, if too low, the strain may be too low to measure and the experiments takes too long.
- A trick that works for materials for which viscoelasticity is thermally activated (i.e. many materials) is to perform tests at various temperatures and then perform at time-temperature superposition.



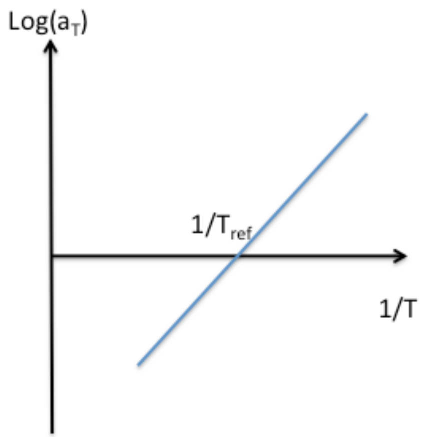
Time-temperature superposition

Defining a_T the shift factor, we can then identify the R(t) function for several temperatures.

$$R(t,T) = R(t/a_T, T_{ref})$$



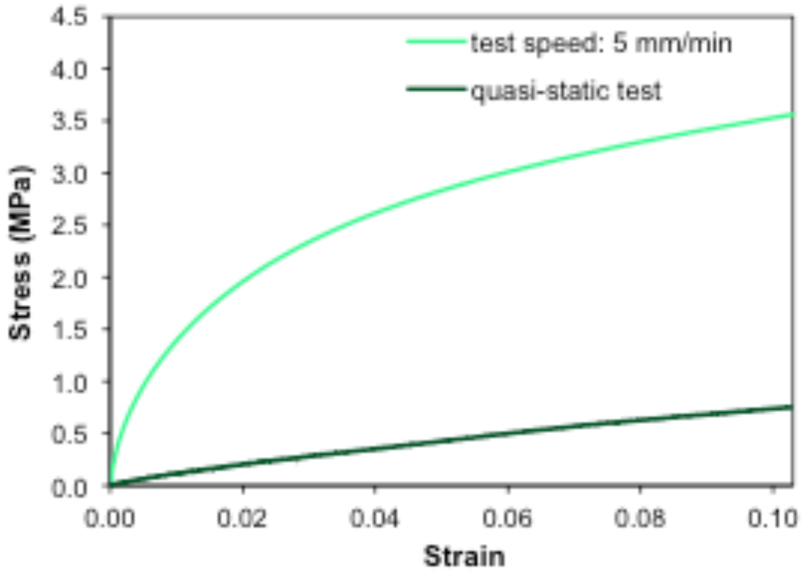
$$a_T(T_2 \rightarrow T_{ref}) = t_b / t_a$$



Static tests at various testing speed

With a simple tensile testing or compression testing equipment, it is possible to test materials at various speed to assess the effect of strain rate on the apparent properties.

Example of a low modulus epoxy resin, stress strain curve as a function of testing speed.



Epoxy system	T_g , (°C)	E-modulus (MPa)	Strength (MPa)	Strain at failure (%)	E modulus (MPa) quasistatic
	13	202 ± 10	14 ± 1	106 ± 4	9

Oscillatory measurements

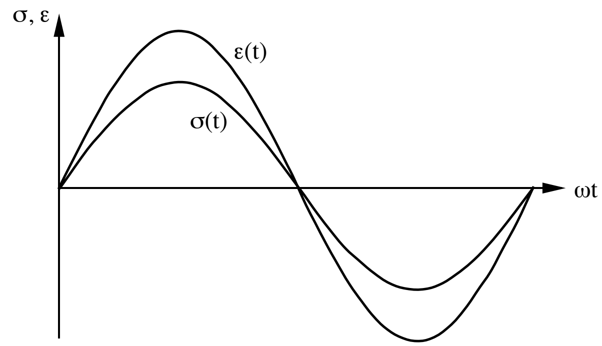
- Sub resonant dynamic method: imposed oscillatory stress or strain to a sample, and the resulting strain or stress is measured.
- This technique is very often used for polymers, composites, any material where the phase angle is rather large.
- It is very practical as several parameters can be adjusted: stress or strain amplitude, frequency, temperature.
- Solicitations can be in tensile mode, in bending mode, in shear depending on the apparatus.

From TA instruments slides

Oscillatory measurements-principle

Elastic material:

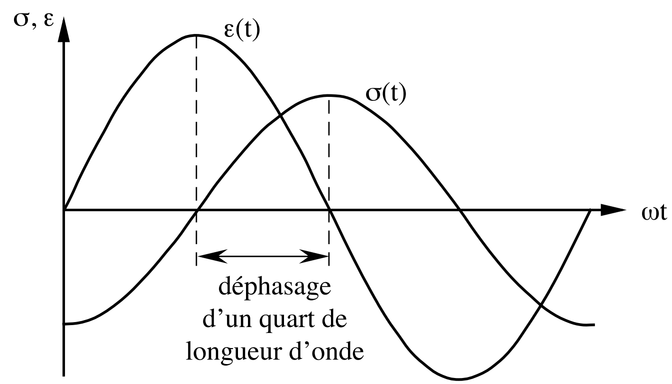
$\varepsilon = \varepsilon_0 \sin(\omega t)$ $\sigma = \sigma_0 \sin(\omega t)$



Viscous material:

$\varepsilon = \varepsilon_0 \sin(\omega t)$ $\sigma = \sigma_0 \sin(\omega t + \pi/2)$

because $\sigma = \eta\, d\varepsilon/dt$



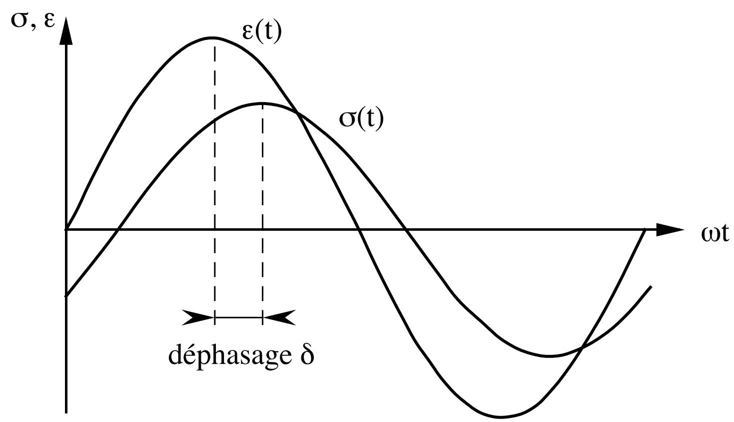
Oscillatory measurements-principle

Viscoelastic material:

$\varepsilon = \varepsilon_0 \sin(\omega t)$ $\sigma = \sigma_0 \sin(\omega t+\delta)$

δ is the phase angle

$$\sigma = \sigma_0 \sin\omega t \cos\delta + \sigma_0 \cos\omega t \sin\delta$$



Definitions:

Storage modulus $$E' = \frac{\sigma_0}{\varepsilon_0}\cos\delta$$

Loss modulus $$E'' = \frac{\sigma_0}{\varepsilon_0}\sin\delta$$

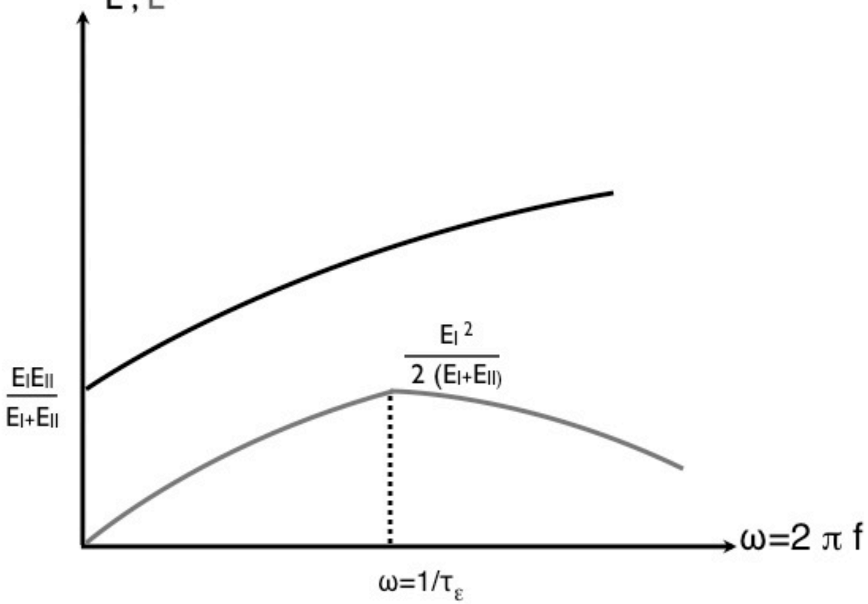
Then, phase angle $$\tan\delta = \frac{E''}{E'}$$

So, finally:

$$\sigma = \varepsilon_0 E' \sin(\omega\ t) + \varepsilon_0 E'' \cos(\omega\ t)$$

Moduli for a SLSM model material

E' increases with frequency, and E'' passes through a maximum when the angular velocity corresponds to the characteristic time of the system.



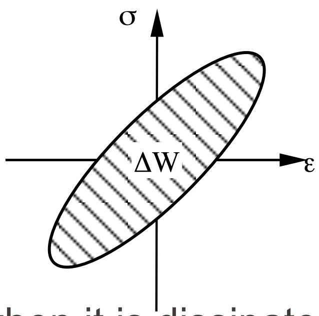
$$E' = \left(\frac{E_I E_{II}}{E_I + E_{II}}\right)\left[1 + \frac{E_I}{E_{II}} \frac{\omega^2 \tau_\varepsilon^2}{1 + \omega^2 \tau_\varepsilon^2}\right]$$

$$E'' = \left(\frac{E_I^2}{E_I + E_{II}}\right)\left[\frac{\omega \tau_\varepsilon}{1 + \omega^2 \tau_\varepsilon^2}\right]$$

Lissajous curves

Plotting the stress versus strain parametric curves, an ellipse is obtained.
The dissipated energy is:

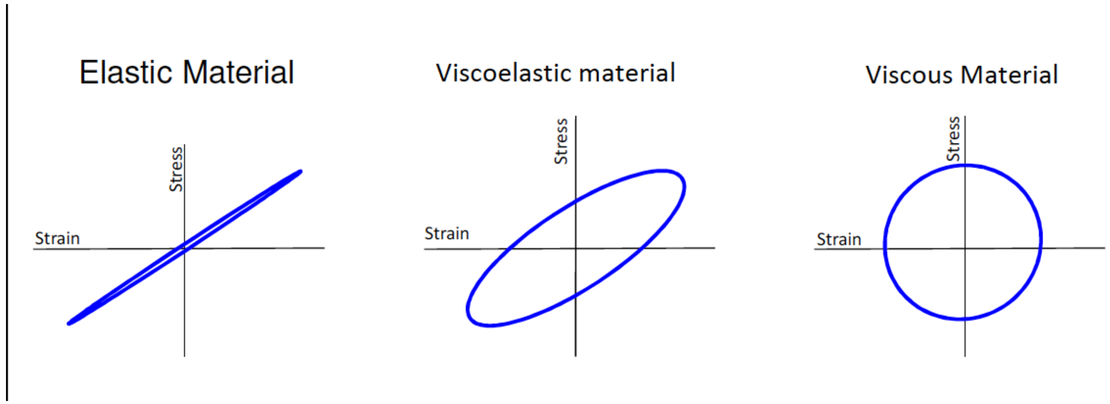
$$\Delta W = \int \sigma \, d\varepsilon = \int_0^{2\pi/\omega} \sigma \frac{\partial \varepsilon}{\partial t} dt$$



So, for a solicitation in strain, counting negative energy when it is dissipated:

$$\Delta W = -\pi E'' \varepsilon_0^2$$

In stress: $\Delta W = -\pi J'' \sigma_0^2$



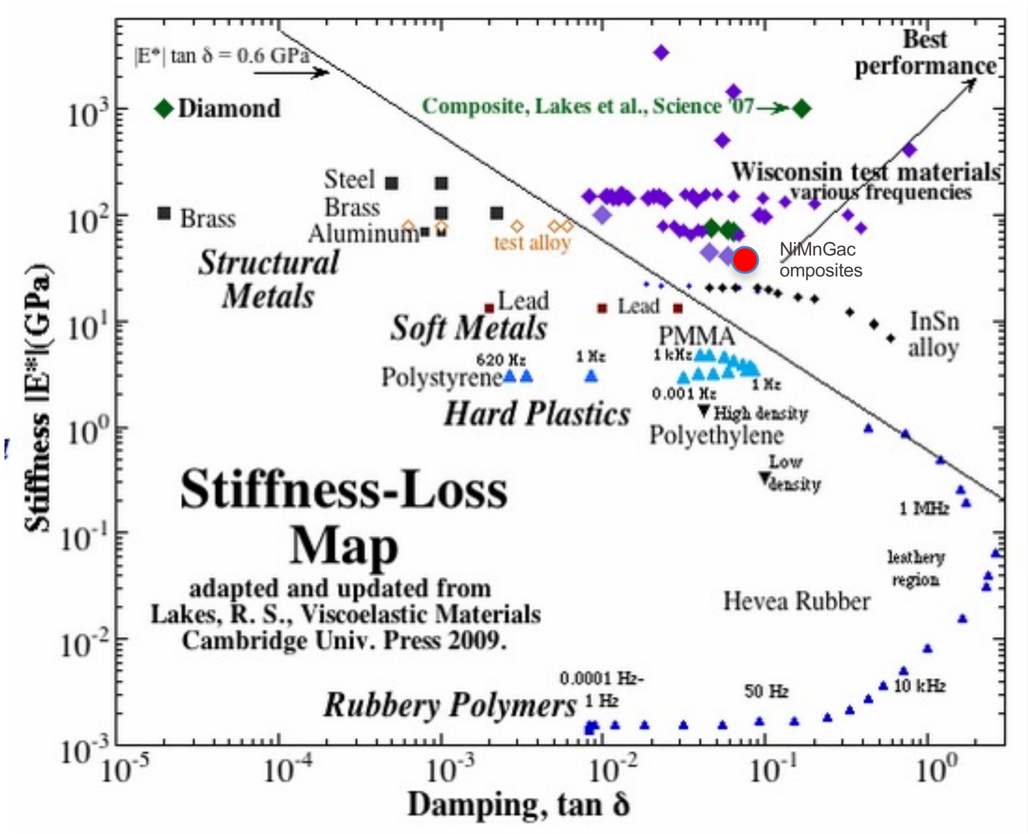
Summary

Parameter	Shear	Elongation	Units
Strain	$\gamma = \gamma_0 \sin(\omega t)$	$\varepsilon = \varepsilon_0 \sin(\omega t)$	---
Stress	$\sigma = \sigma_0 \sin(\omega t + \delta)$	$\tau = \tau_0 \sin(\omega t + \delta)$	Pa
Storage Modulus (Elasticity)	$G' = (\sigma_0/\gamma_0)\cos\delta$	$E' = (\tau_0/\varepsilon_0)\cos\delta$	Pa
Loss Modulus (Viscous Nature)	$G'' = (\sigma_0/\gamma_0)\sin\delta$	$E'' = (\tau_0/\varepsilon_0)\sin\delta$	Pa
Tan δ	G''/G'	E''/E'	---
Complex Modulus	$G^* = (G'^2+G''^2)^{0.5}$	$E^* = (E'^2+E''^2)^{0.5}$	Pa
Complex Viscosity	$\eta^* = G^*/\omega$	$\eta_E^* = E^*/\omega$	Pa-sec

Cox-Merz Rule for Linear Polymers: $\eta^*(\omega) = \eta(\dot{\gamma})$ @ $\dot{\gamma} = \omega$

From TA instruments slides

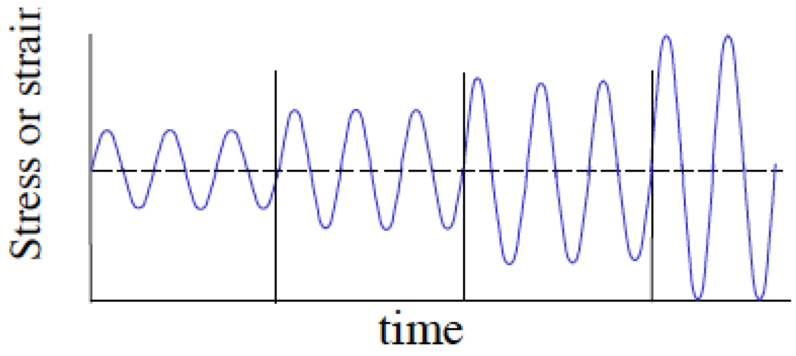
Stiffness loss maps for several materials

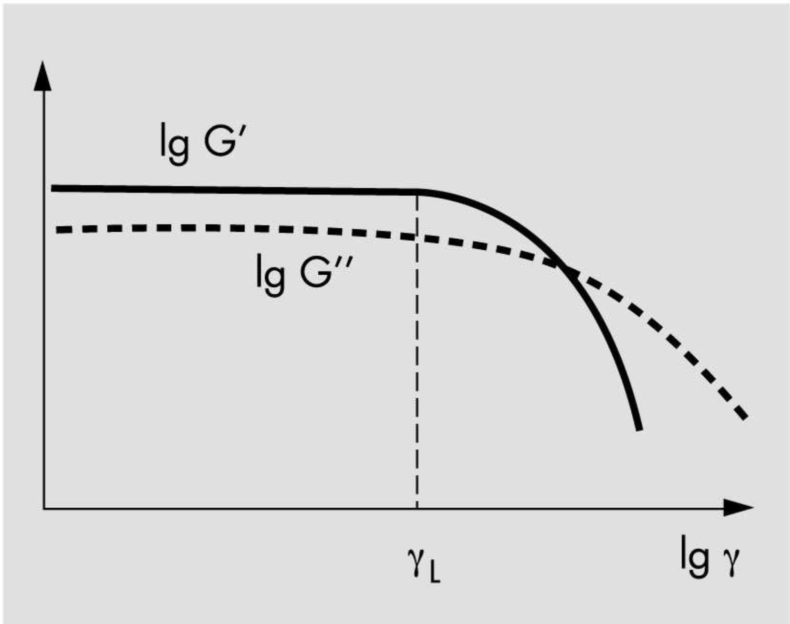


From Lakes, 2009

Practical oscillatory mesurements

Define the linear viscoelastic range, using a strain sweep:





This defines the range of amplitude we can use for the test.

From TA instruments slides

Practical oscillatory mesurements

Then perform the dynamic measurement, at a given temperature and/or frequency.

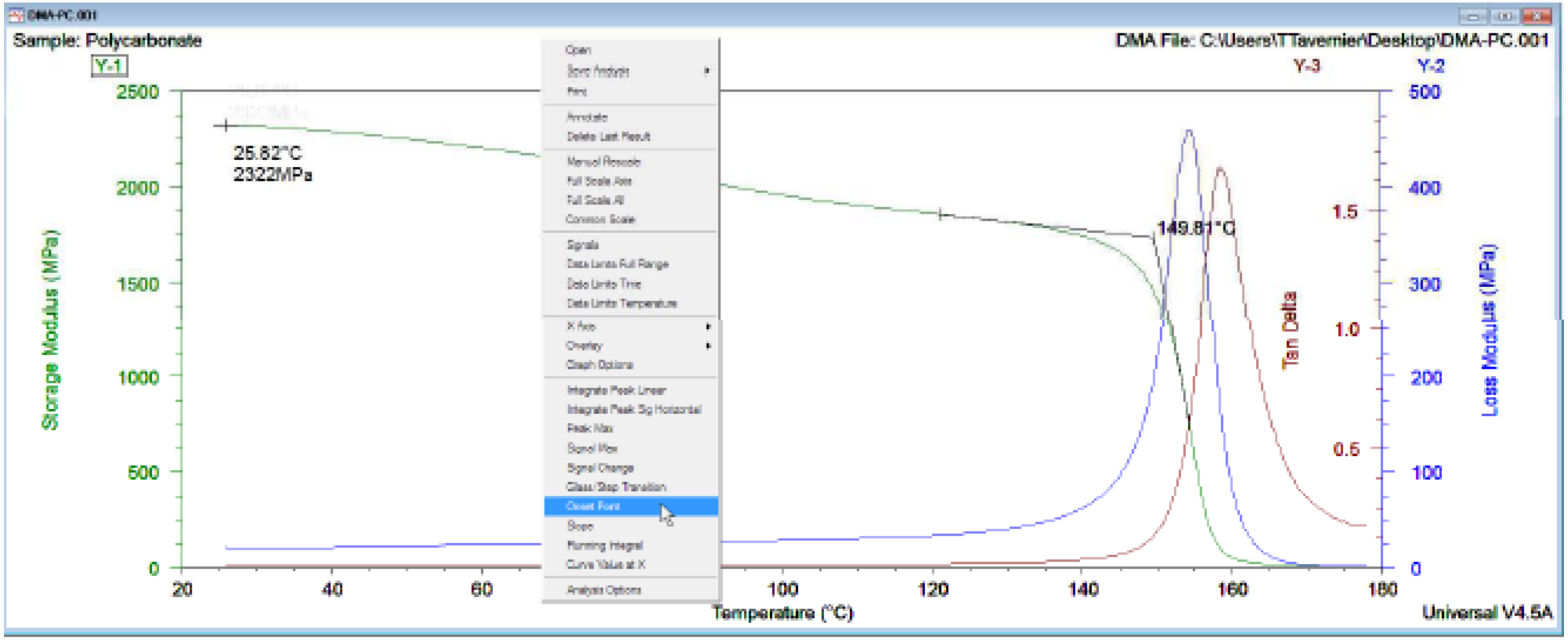
Example of equipment:

Dynamic Mechanical Analysis

From TA instruments slides

Practical measurements

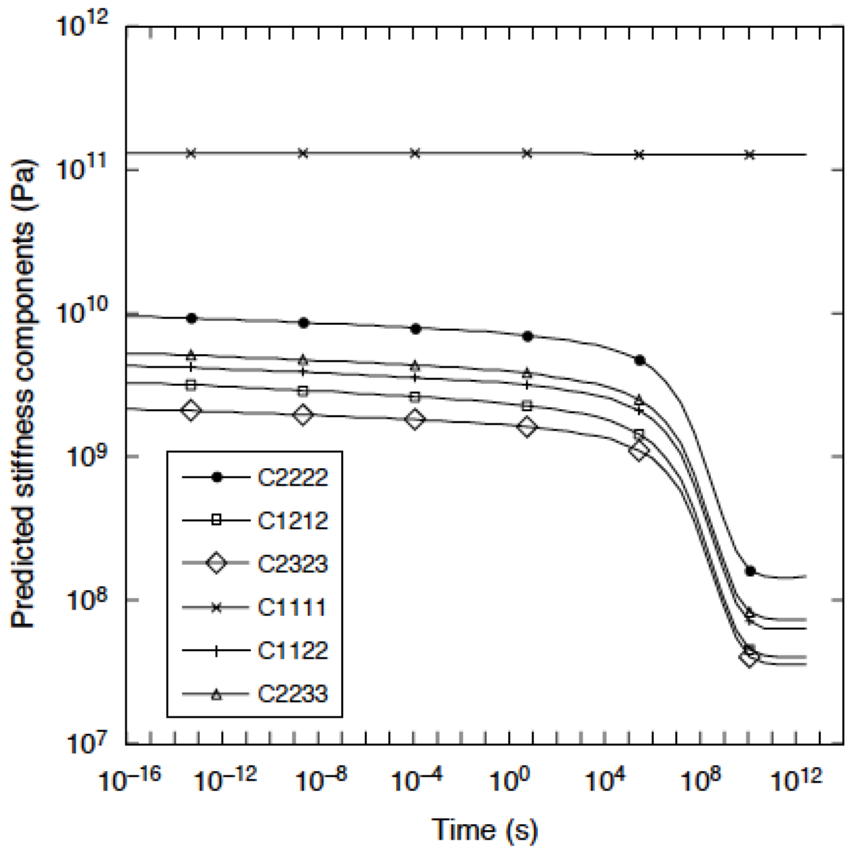
Example: Polycarbonate



Time temperature superposition
Carbon-epoxy UD composite

Composite moduli, as a function of time, obtained through a time temperature superposition

$$a_T = 2 \times 10^{18} \cdot e^{-0.33767T}$$

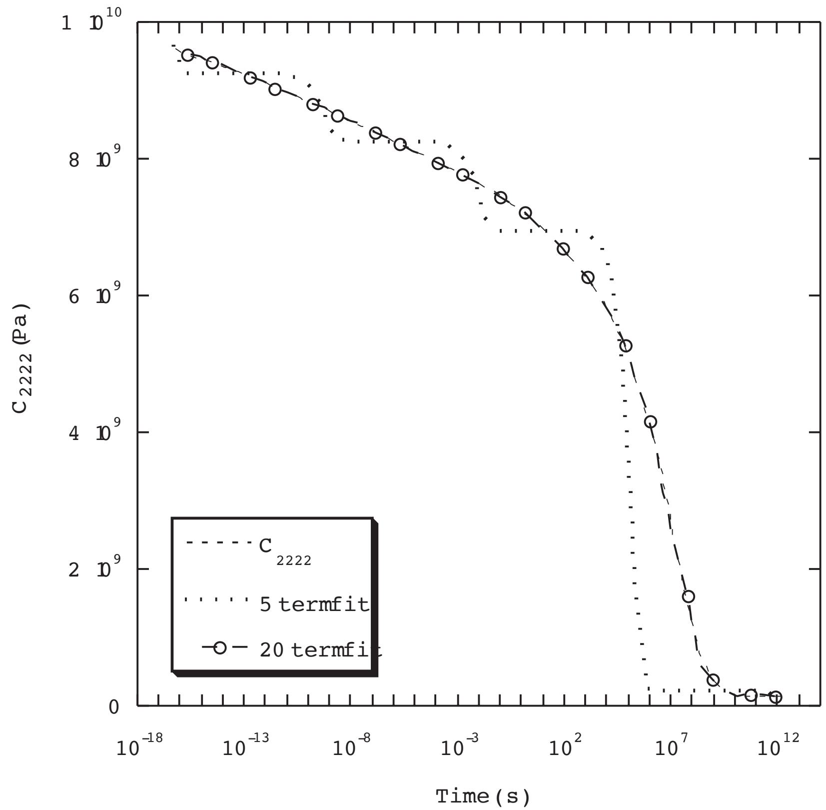


From Clifford, Comp A 2005

TTS Carbon-epoxy UD composite

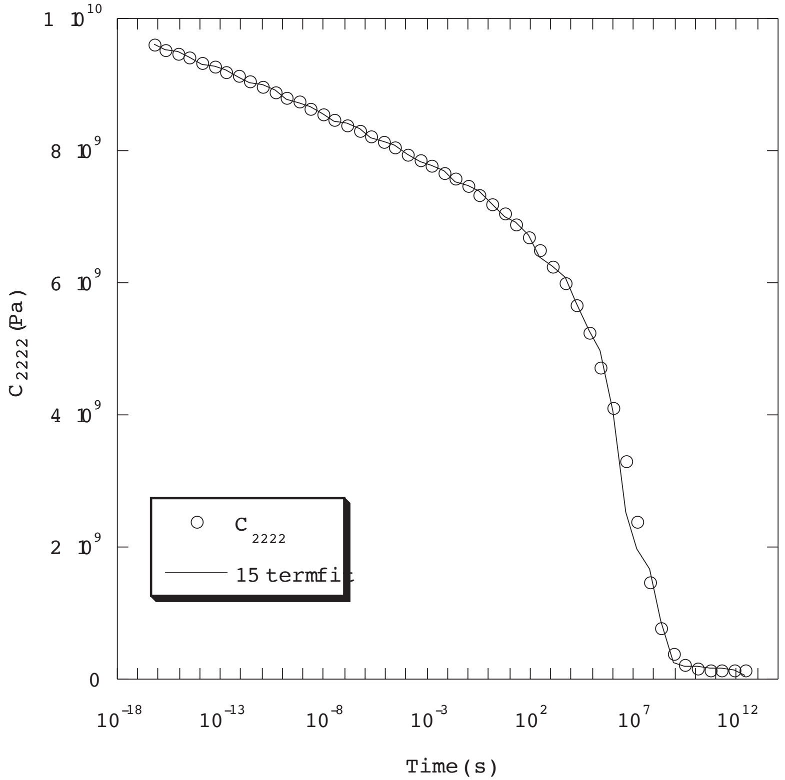
Fitting of the curve, example of transverse modulus C2222, with a series of Maxwell models

$$C_{ij}(t) = C_{ij0} + \sum_{\omega=1}^{N} C_{ij\omega} \exp\left(-\frac{t}{\lambda_{ij\omega}}\right)$$



TTS Carbon-epoxy UD composite

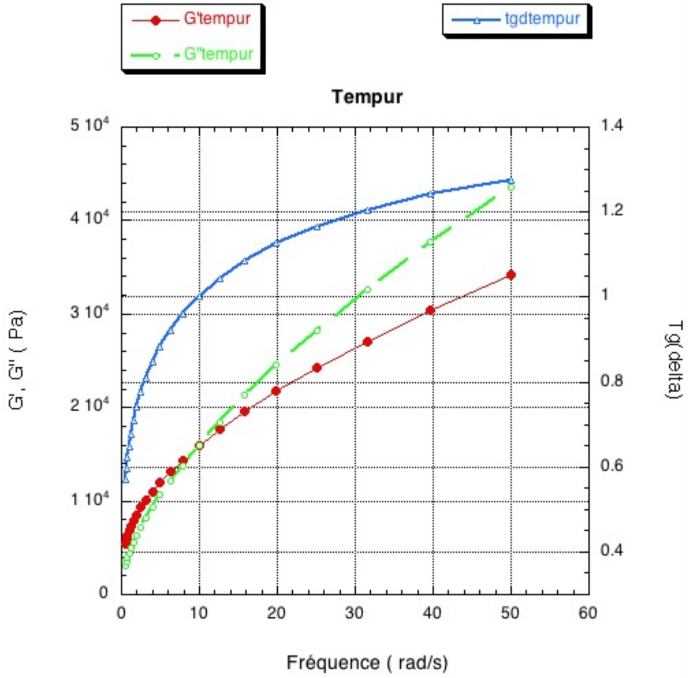
With 15 terms in the fitting, good agreement is obtained. This model can then be introduced in a Finite Element Model

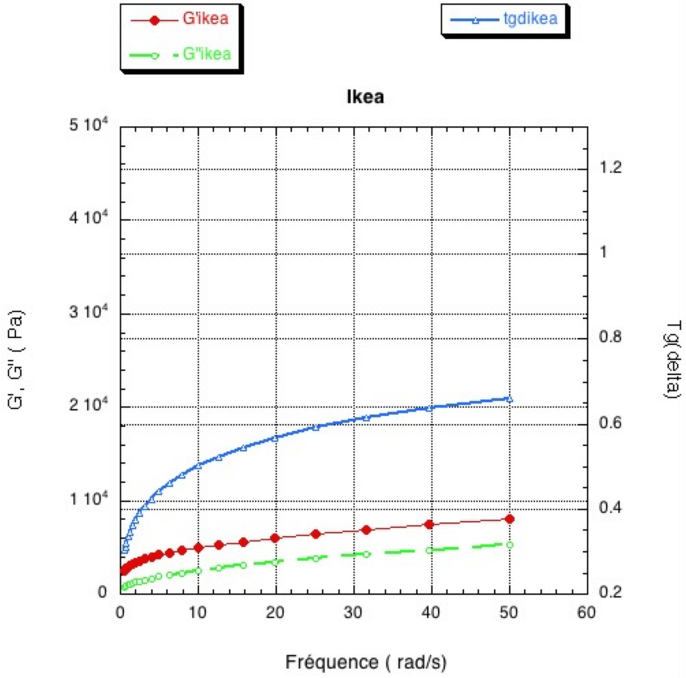


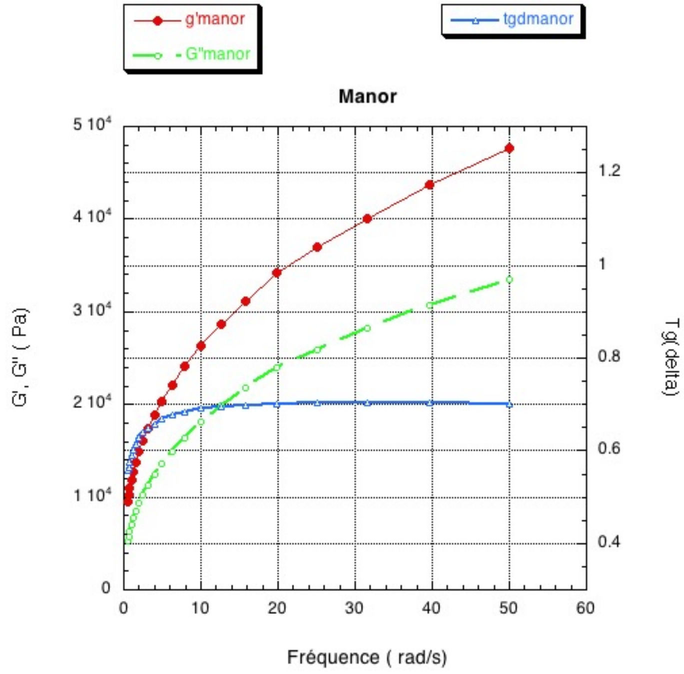
EPFL

Other example: Which is the best pillow?

Viscoelastic pillows from Tempur, Ikea and Manor.
G', G", Tg δ as a fct of frequency.
What do we observe?



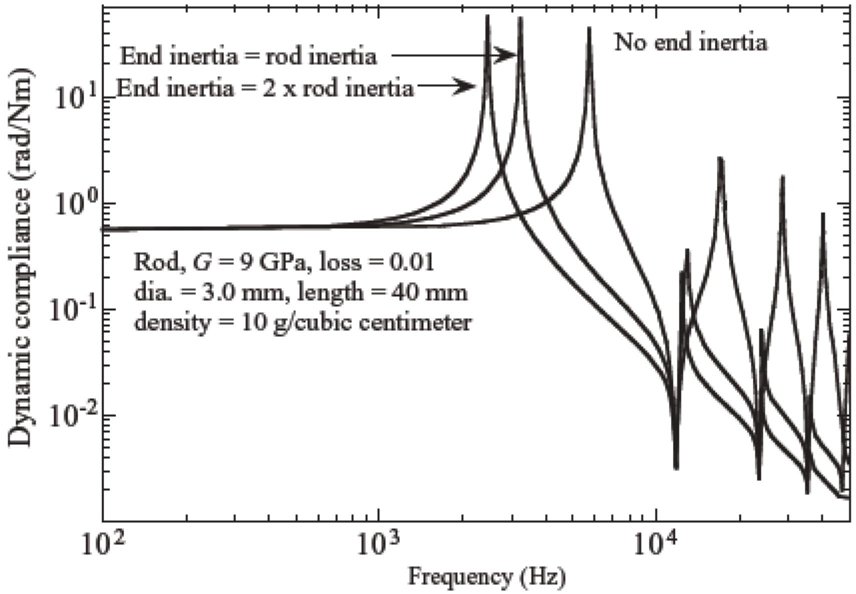




Other dynamic measurement techniques

Resonance methods: appropriate for low loss materials, tgδ<10^{-3}. Instruments can be a torsion pendulum, or magnetic devices that vibrate the sample, generally fixed as one or both ends.

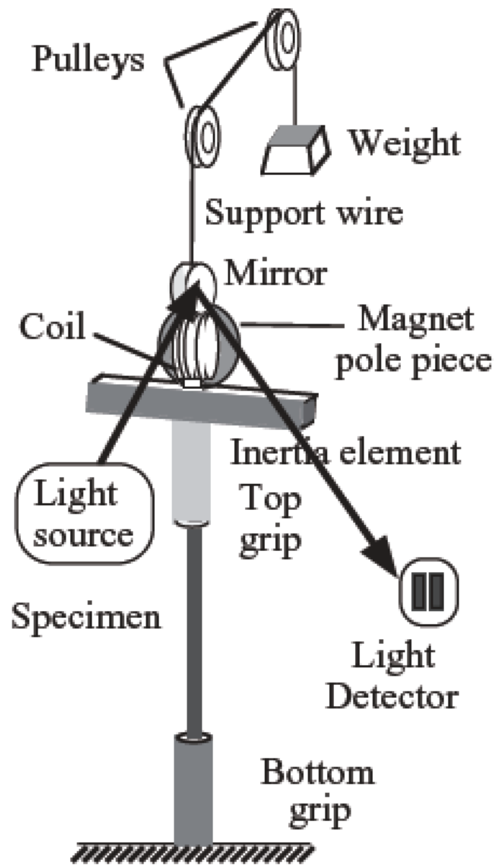
Inertia can become a problem.



From Lakes 2013

Other dynamic measurement techniques

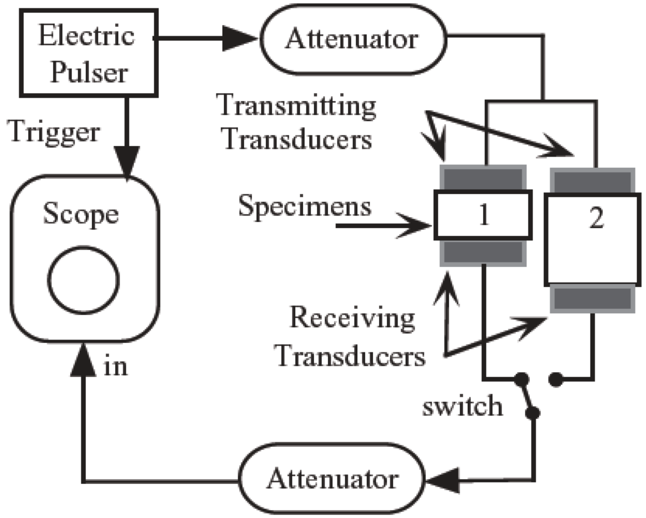
Example of a driven torsion pendulum.
Torque on the coil is pole piece generated by the action of a magnetic field supplied by a permanent magnet (one pole piece shown)



From Lakes, 2013

Other measurement techniques

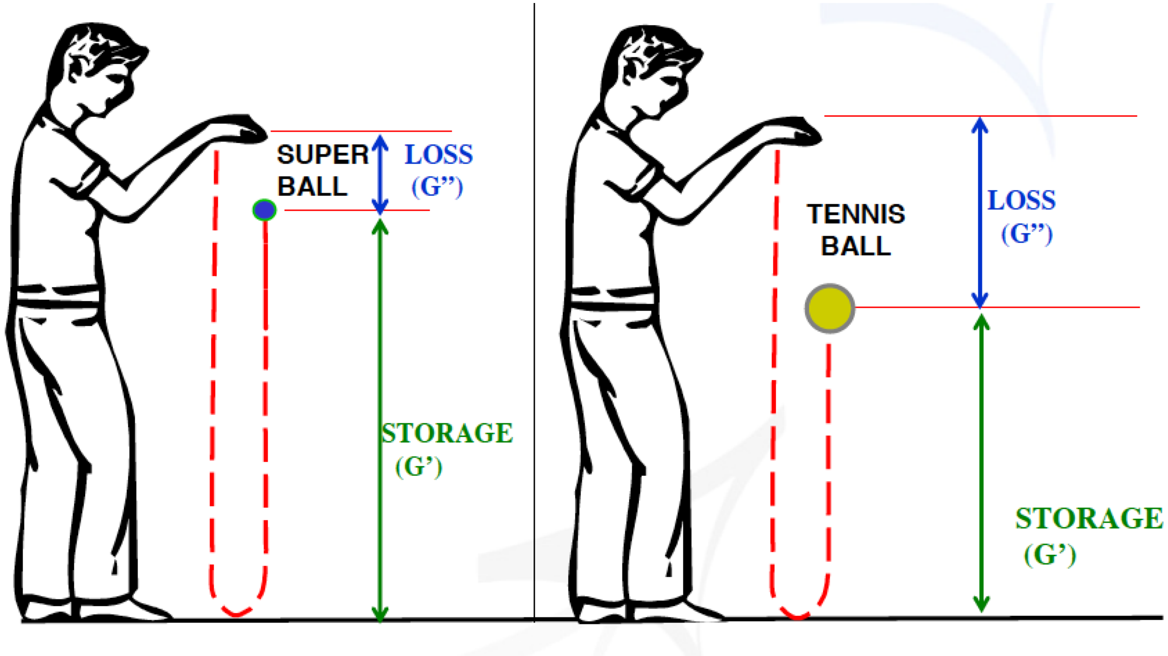
- Indentation testing can be used, for creep or dynamic testing, at small scales or for surface coatings.
- Attenuation of ultrasonic waves can also be used



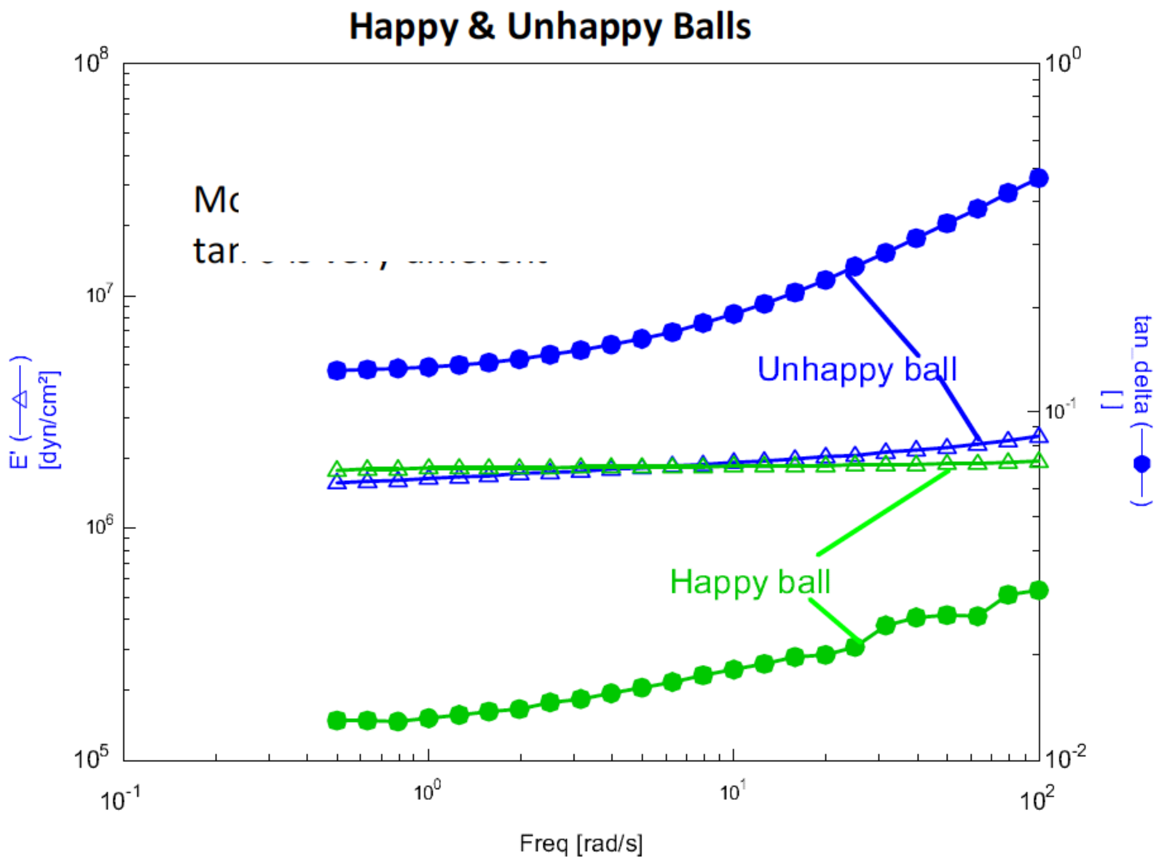
A simple dynamic measurement technique

Rebound test, drop at H1, rebounds to H0, approximate relation:

$$\tan\delta \cong \frac{1}{\pi}\ln\left(\frac{H_0}{H_1}\right)$$



Which ball has the best rebound?



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Conclusion

- Viscoelastic materials exhibit a time dependent behaviour
- Methods to characterize this behavior are simple static experiments, or dynamic subresonant or resonant methods.
- To obtain the behavior of the material over several decades of time or frequency, the equivalence between time and temperature is generally used.
- Take home message: when dealing with polymers or soft matter, do not trust one value of modulus, but always consider the time frame of the testing method as well.
- See you this afternoon at the DMA.

"Mr. Osborne, may I be excused? My brain is full."

Some References

- Viscoelasticity of engineering materials, Y.M. Haddad, Chapman & Hall, 1996.

- Viscoelastic solids, R. S. Lakes, CRC press, 2009 (available as e-book as well, 2013 edition).

- Ferry, J. D., Viscoelastic Properties of Polymers

- Ward, I. M. and Hadley, D. W., Mechanical Properties of Solid Polymers