Polymers and Composites Processing

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An Introduction to Composite Materials

1. General Introduction

- **1.1. Types of Composite material**
  - Polymer matrix composites (PMC)
  - Metal matrix composites (MMC)
  - Ceramic matrix composites (CMC)

- The properties of particular interest
  - Stiffness (Young’s modulus), strength, toughness, density
  - Thermal properties : expansivity, thermal conductivity
  - Electrical properties : electrical conductivity, dielectric constant

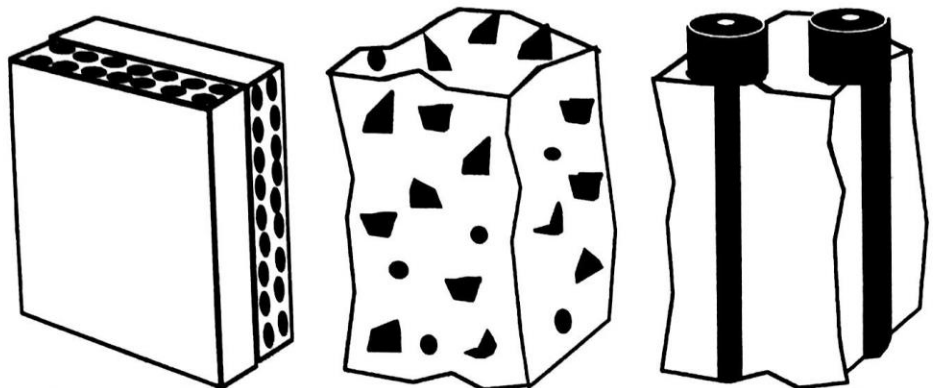
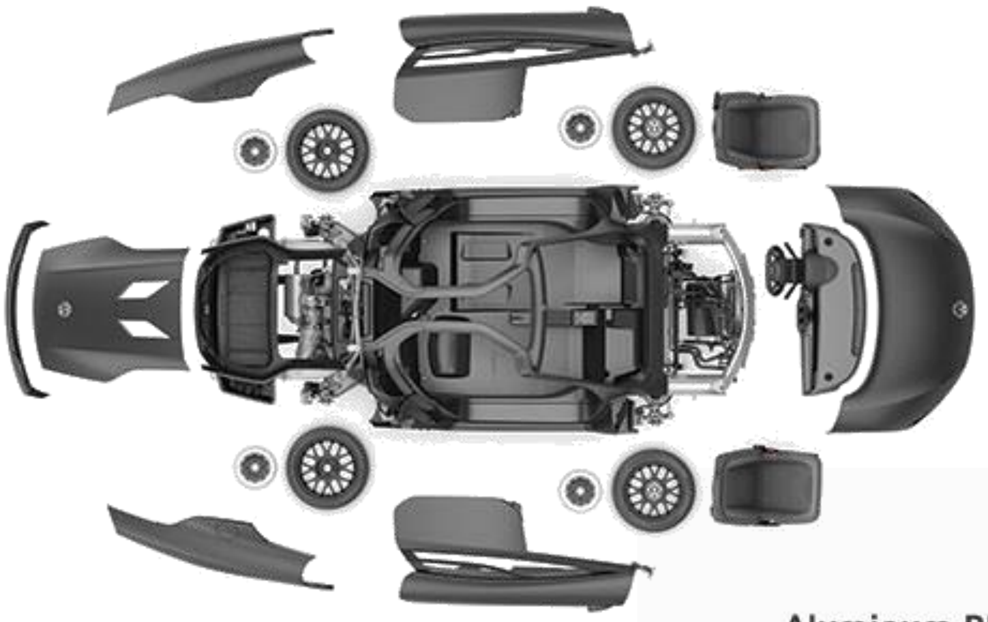
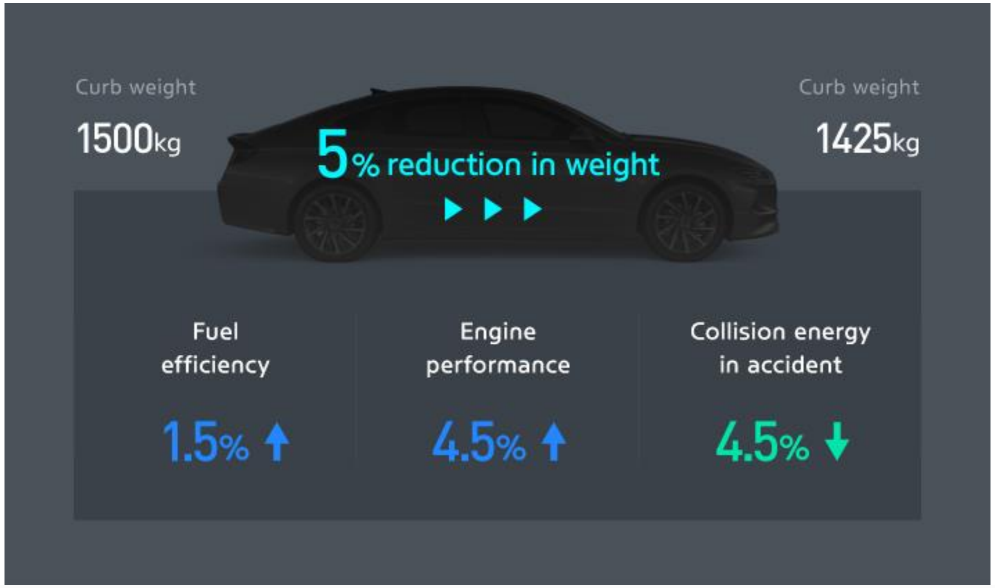


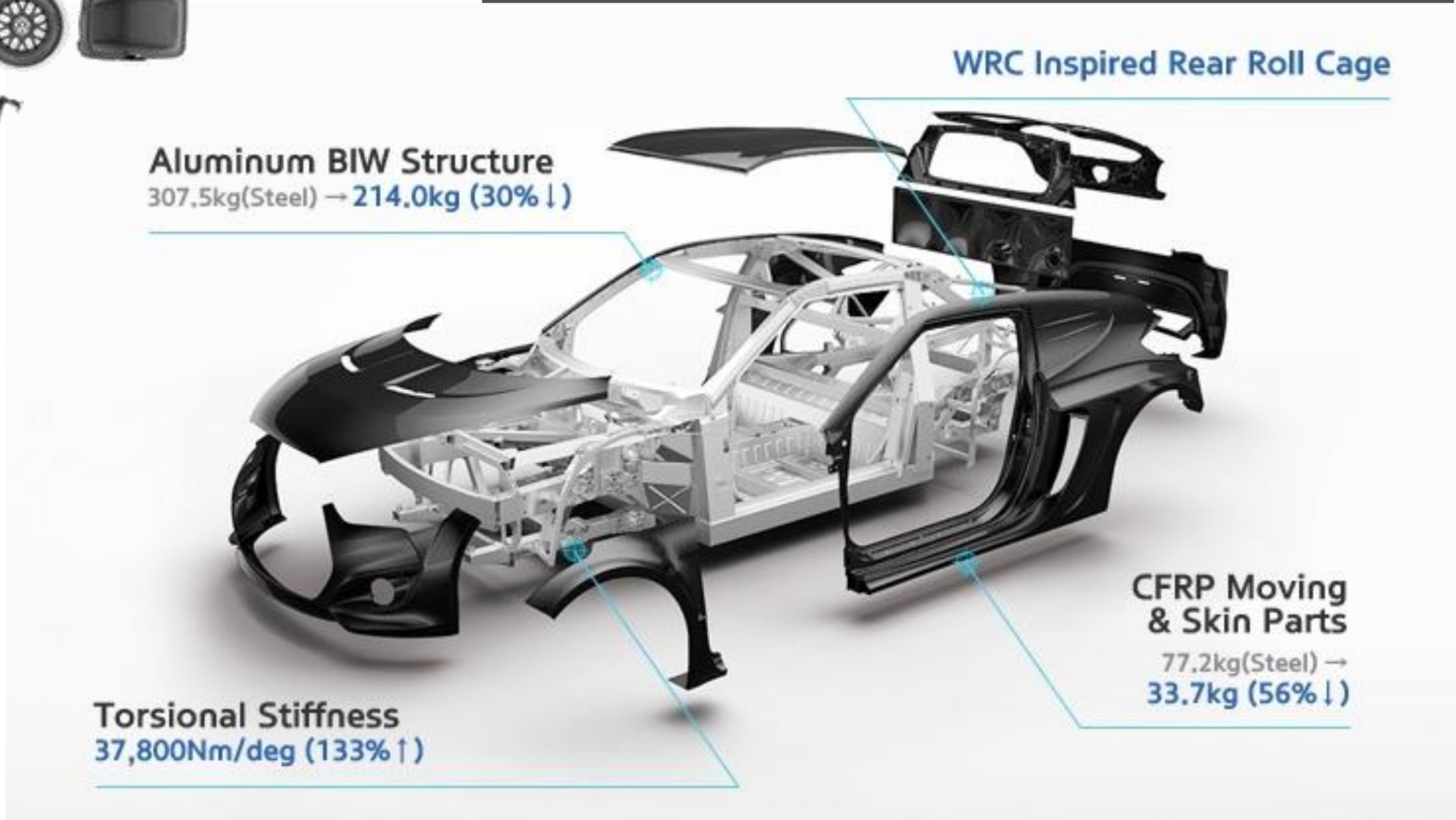
Fig. 1.1 Schematic depiction of representative polymer, metal and ceramic matrix composites.

CFRP automotive



http://www.compositesworld.com





https://news.hyundaimotorgroup.com/

Table 1.1 *Overview of properties exhibited by different classes of material*

Type of material (example)	Density ρ ($Mg\ m^{-3}$)	Young's modulus E (GPa)	Tensile strength σ (MPa)	Fracture toughness K_c (MPa $\sqrt{m}$)	Thermal conductivity K ($W\ m^{-1}\ K^{-1}$)	Thermal expansivity α ($10^{-6}\ K^{-1}$)
Thermosetting resin (epoxy)	1.25	3.5	50	0.5	0.3	60
Engineering thermoplastic (nylon)	1.1	2.5	80	4	0.2	80
Rubber (polyurethane)	1.2	0.01	20	0.1	0.2	200
Metal (mild steel)	7.8	208	400	140	60	17
Construction ceramic (concrete)	2.4	40	20	0.2	2	12
Engineering ceramic (alumina)	3.9	380	500	4	25	8
Wood (spruce) (load // grain)	0.6	16	80	6	0.5	3
(load ⊥ grain)	0.6	1	2	0.5	0.3	10
General PMC (in-plane) (chopped strand mat)	1.8	20	300	40	8	20
Adv. PMC (APC-2) (load // fibres)	1.6	200	1500	40	200	0
(load ⊥ fibres)	1.6	3	50	5	40	30
MMC (A1–20%SiC_p)	2.8	90	500	15	140	18

- # 1.2. Design of composite materials

▪ Property maps

- A broad view of the property combinations obtainable from different composite systems

- A convenient method of comparing the property combinations offered by potential matrices and reinforcements with those of alternative conventional materials

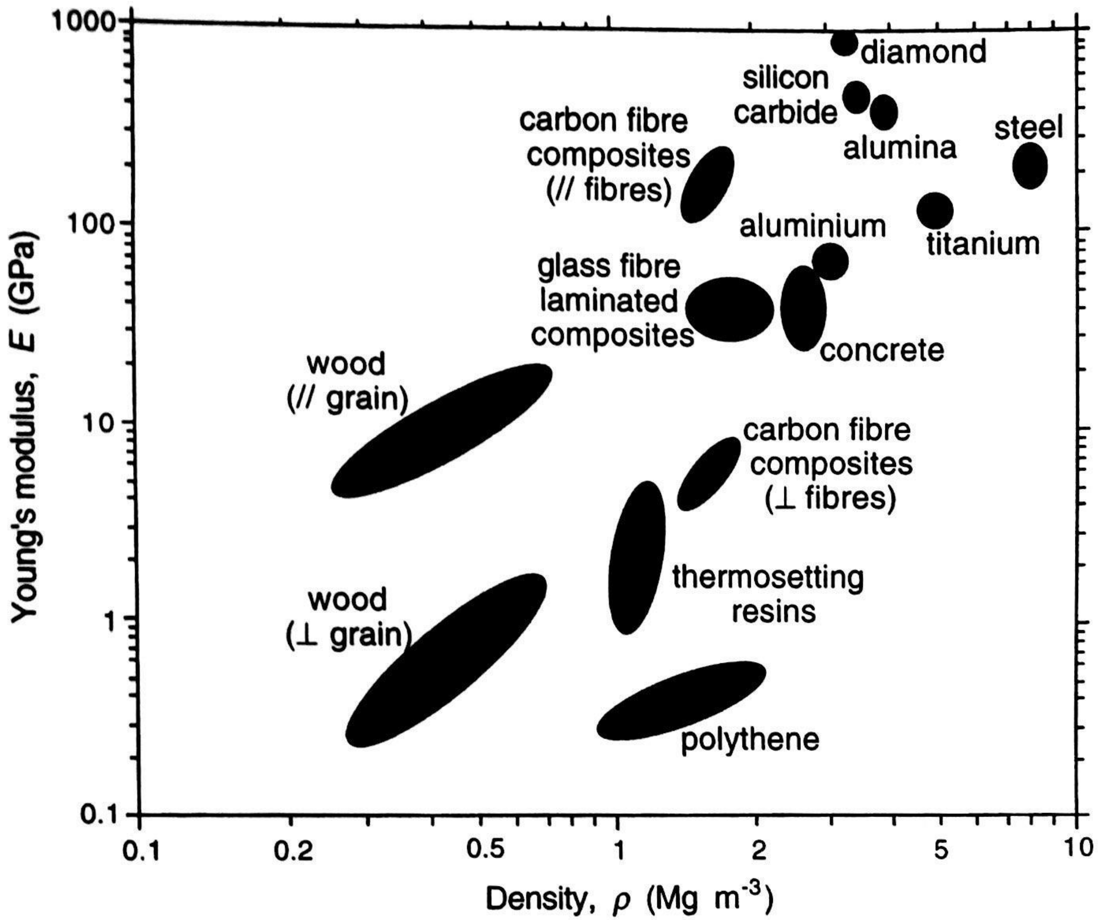


Fig. 1.2 Data for some engineering materials in the form of areas on a map of Young's modulus E against density ρ.

▪ Merit index (*M*)

- Attractive matrix/reinforcement combinations for the performance required in the form of a specified combination of properties
- Lower bound, upper bound of the composite properties for a given volume fraction of reinforcement
- Shaded areas are bounded by upper and lower bounds predicted for the composite systems
- diagonal dotted lines represent E/ρ, E/ρ^2, E/ρ^3 → the smallest density (e.g. carbon fiber composite) would show the highest increase of Young's modulus

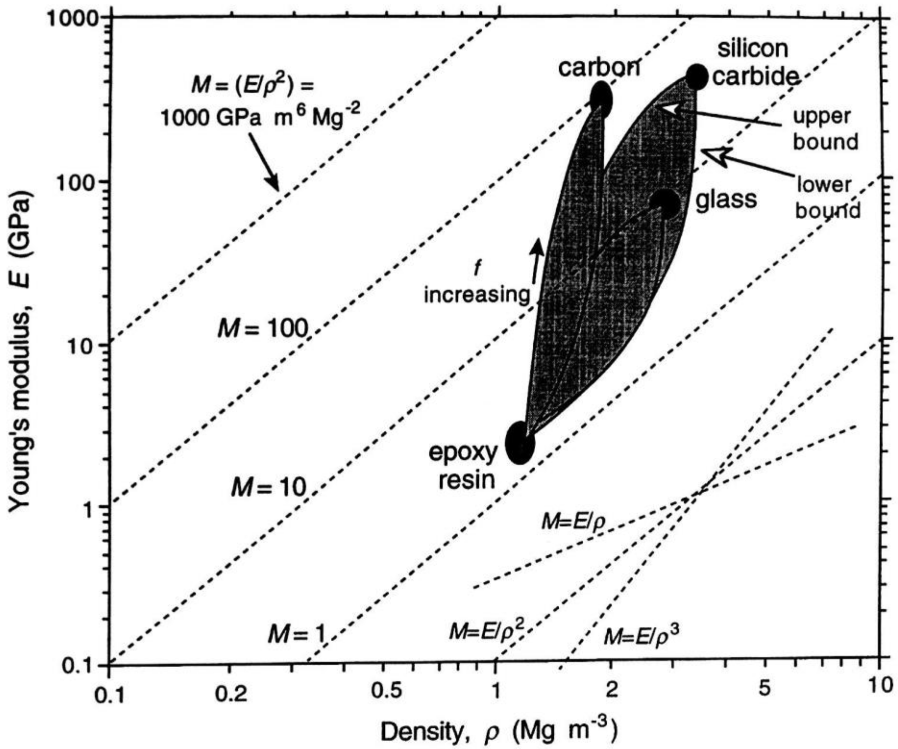


Fig. 1.3 Predicted map of Young's modulus E against density ρ for composites of glass, carbon or silicon carbide fibres in a matrix of epoxy resin. The shaded areas are bounded by the axial and transverse values of E predicted for the composite systems. The diagonal dotted lines represent constant values of three merit indices (E/ρ, E/ρ^2 and E/ρ^3). For the E/ρ^2 case, several lines are shown corresponding to different values of the ratio.

- **1.3. The concept of load transfer**

 - Load sharing between the matrix and the reinforcing materials
 - At equilibrium, the volume-averaged loads on matrix and fibers are expressed as

$$(1-f)\bar{\sigma}_m + f\bar{\sigma}_f = \sigma_A$$

$\bar{\sigma}_m$, $\bar{\sigma}_f$: volume-averaged matrix and fiber stresses in a composite under an external applied stress σ_A

f : volume fraction of reinforcements (fibers)

 - A certain proportion of that load will be carried by the fiber and the remainder by the matrix
 - The reinforcement carries a high proportion of the externally applied load because the reinforcement is usually stronger as well as stiffer than the matrix

2. Fiber architecture

- **2.1. General considerations**

  - **2.1.1. Volume fraction and weight fraction**

- Most calculations on composite materials are based on the volume fractions
- When calculating the density of the composite, weight fractions are used
- The appropriate conversion equation is :

fiber

$$f = \frac{\frac{w_f}{\rho_f}}{\frac{w_f}{\rho_f} + \frac{w_m}{\rho_m}}$$

matrix

Fiber volume fraction

$$w_f = \frac{f\rho_f}{f\rho_f + (1-f)\rho_m}$$

Fiber weight fraction

- **2.1.2. Fiber packing arrangements**

- In a unidirectional lamina, all the fibers are aligned parallel to each other
- Ideally, the fibers can be considered to be arranged on a hexagonal or square lattice
- For the ideal arrangements, the volume fraction of fibers f is related to the fiber radius r :

$$f = \frac{\pi}{2\sqrt{3}}\left(\frac{r}{R}\right)^2$$ hexagonal

$$f = \frac{\pi}{4}\left(\frac{r}{R}\right)^2$$ square

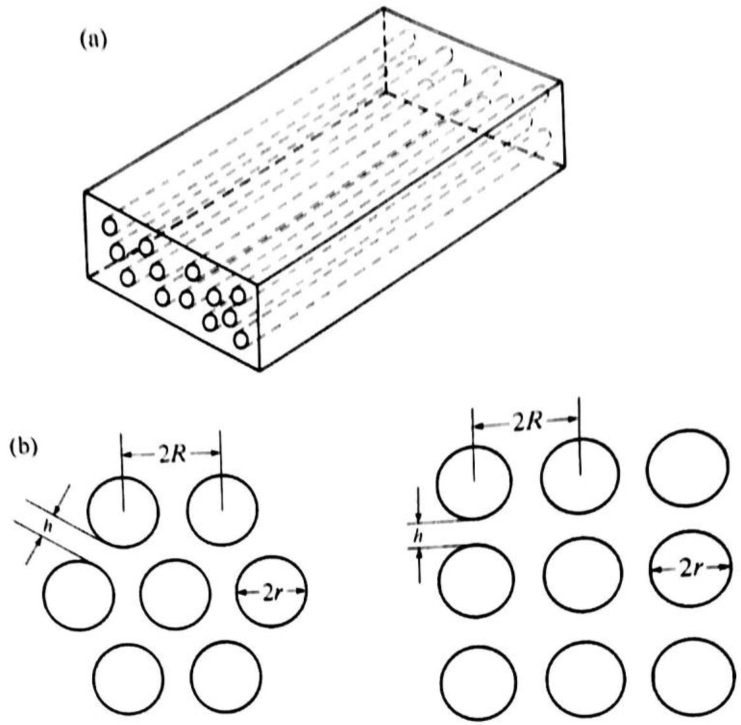


Fig. 3.1 (a) Unidirectional lamina. (b) Hexagonal and square packing of unidirectional fibres.

- The maximum value of f will occur when the fibers are touching, i.e. $r = R$

→ $f_{max} = 0.907$ for a hexagonal array, $f_{max} = 0.785$ for a square array

- The separation of the fibers, h, varies with f:

$$h = 2\left[\left(\frac{\pi}{2f\sqrt{3}}\right)^{1/2} - 1\right]r \quad \text{hexagonal} \qquad\qquad h = 2\left[\left(\frac{\pi}{4f}\right)^{1/2} - 1\right]r \quad \text{square}$$

- At higher values of f (~0.7), the spacing h becomes very small
- The presence of the fiber modifies the surrounding matrix
- The matrix is often highly constrained between closely spaced fibers
- A highly uniform distribution of fibers is required if physical contact between the fibers is to be avoided

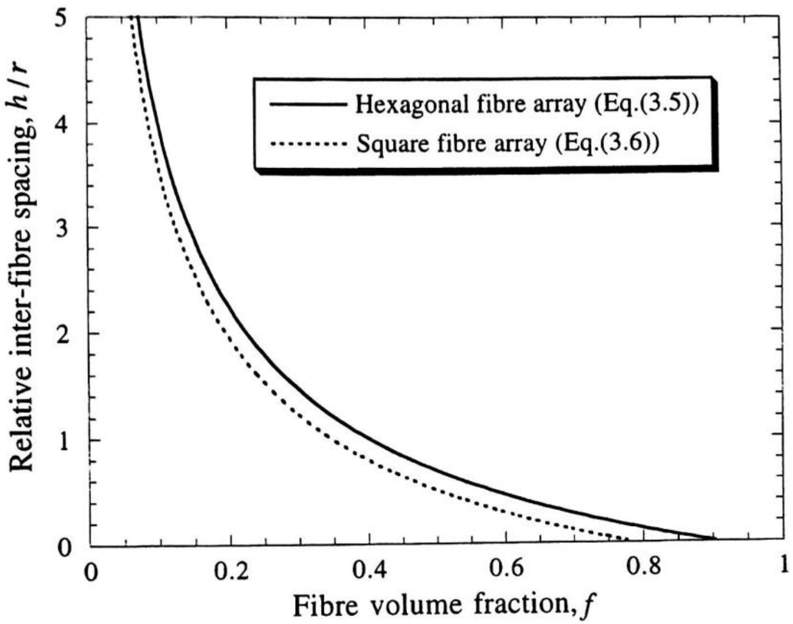


Fig. 3.2 Effect of fibre volume fraction f on the spacing between fibres.

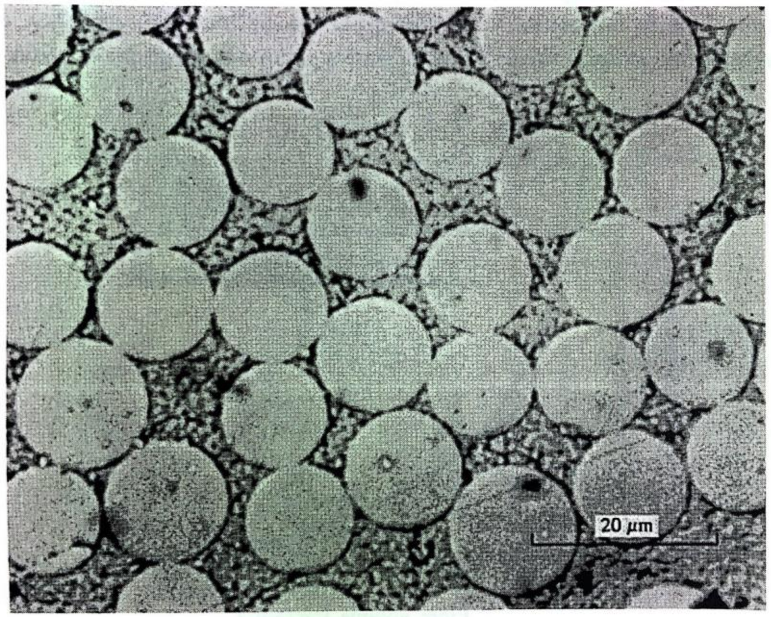


Fig. 3.3 Optical micrograph of a section cut at right angles to fibres in unidirectional laminae of glass fibre/polyester resin.

- **2.2. Long fibers**
  - **2.2.1 Laminates**

- High-performance polymer components usually consist of layers or *laminae* stacked in a pre-determined arrangement
- A unidirectional laminae is often called a ***ply*** and a stack of laminae is called a ***laminate***

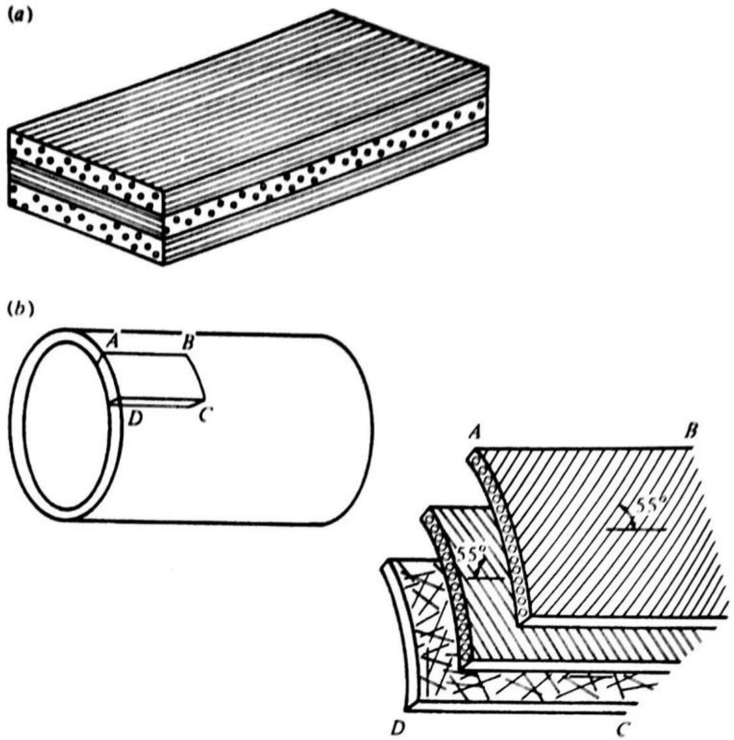


Fig. 3.4 (a) Flat laminate with unidirectional laminae at 90° to each other. (b) Cylindrical laminate with one layer of chopped-strand mat and two unidirectional laminae.

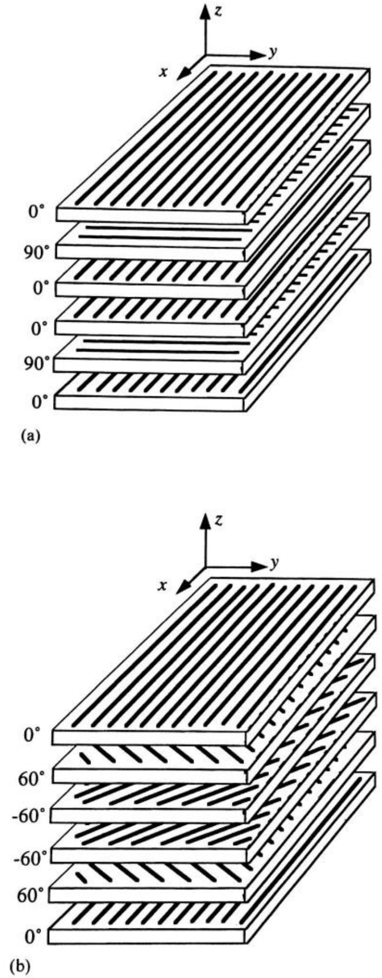


Fig. 3.5 Arrangement of plies in (a) a crossply laminate and (b) an angle-ply laminate sandwiched between 0° plies.

• 2.2.2 Woven, braided and knitted fiber arrays

- Textile processes to produce a variety of geometrical forms : *weaving, braiding, knitting*
- Woven structures lead to pockets of matrix at the cross-over points, and the maximum fiber content for woven roving composites is less than for fully aligned materials

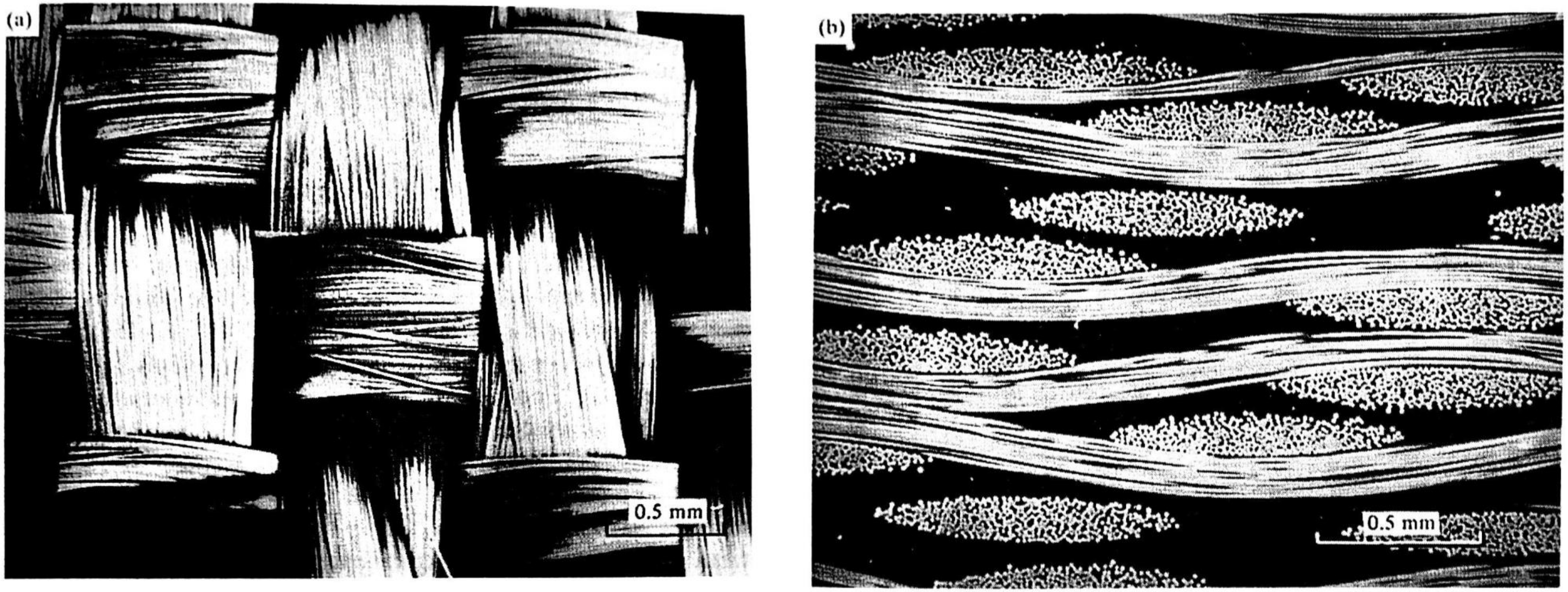


Fig. 3.6 (a) SEM micrograph of a woven roving before infiltration with resin. (b) Photomicrograph of a polished section through a woven roving laminate parallel to one set of fibres.

- **2.3. Short fibers**
  - **2.3.1 Fiber orientation distributions in three dimensions**

- Fiber orientation distributions are more difficult to measure and characterize when the fibers do not lie parallel to a single plane
- The simplest method is based on the assumption that the fibers are straight cylinders of circular section
- Projected length, l_p, of the fibers are used to calculate the angle β

$$\beta = \tan^{-1}\left(\frac{t}{l_p}\right)$$

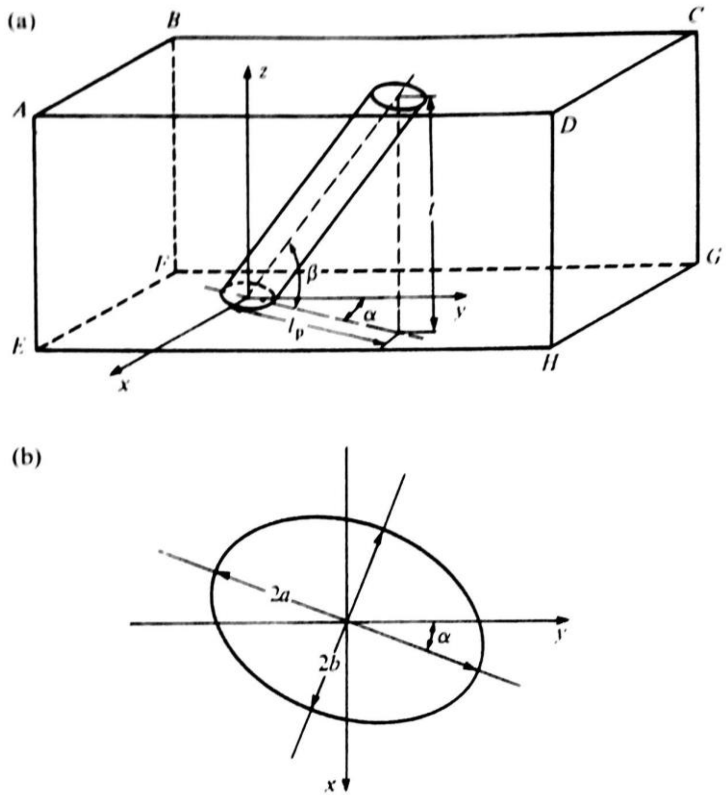


Fig. 3.9 Determination of fibre orientation in a thin section. Orientation defined (a) by angles α and β; and (b) by shape and orientation of fibre cross-section.

- **2.3.2 Fiber length distributions**

 - Processing operations such as extrusion can cause extensive fiber fracture with strong effects on mechanical properties

 - The ***number average*** fiber length :

$$L_N = \frac{\sum N_i L_i}{\sum N_i}$$

N_i : the number of fibers of length L_i

 - The ***weight (or volume) average*** fiber length :

$$L_w = \frac{\sum W_i L_i}{\sum W_i}$$

W_i : the weight of fibers of length L_i

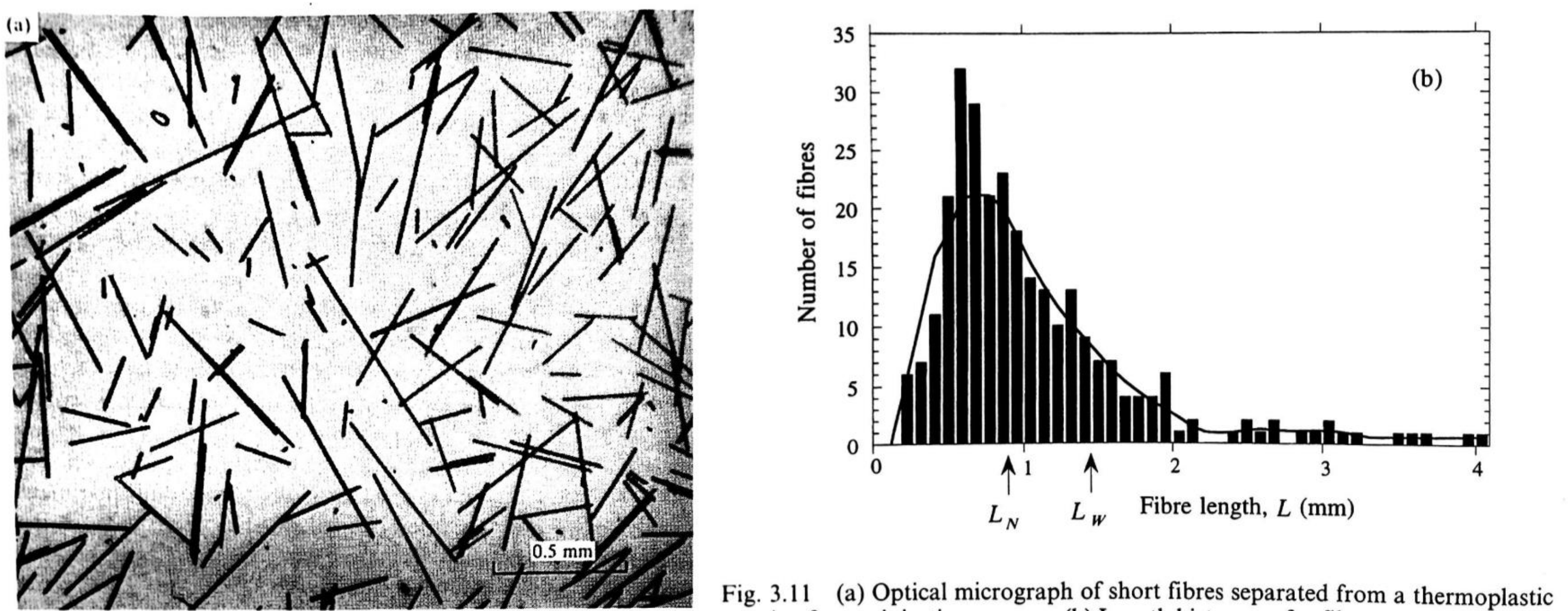


Fig. 3.11 (a) Optical micrograph of short fibres separated from a thermoplastic matrix after an injection process. (b) Length histogram for fibres extracted from a thermoset injection moulding. (Pennington 1979).

- **2.4. Fiber orientation during processing**

 - Changes in fiber orientation often occur during the processing of short-fiber composite composites
 - The polymer melt undergoes both elongational or extensional flow
 - The viscosity of the matrix affects the final orientation distribution

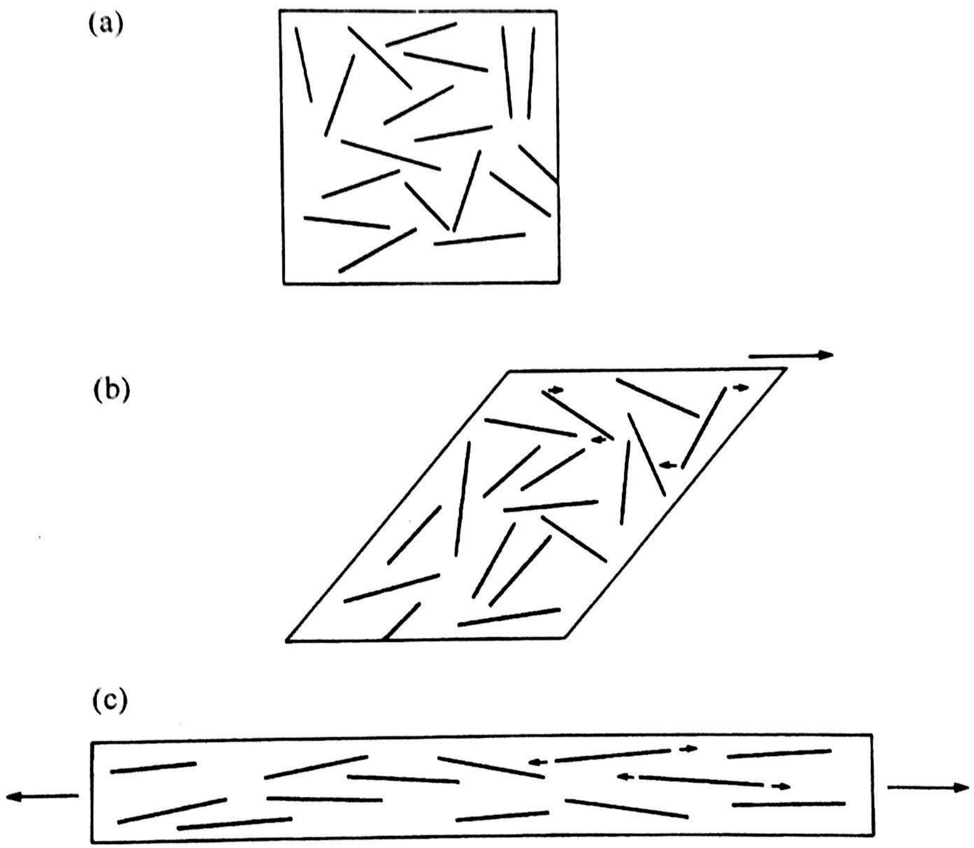


Fig. 3.14 Schematic representation of the changes in fibre orientation occurring during flow. (a) Initial random distribution, (b) rotation during shear flow and (c) alignment during elongational flow.

3. Elastic deformation of long-fiber composites

- **3.1. Axial stiffness**

  - The composite materials have aligned long-fibers in the matrix

  - The long-fiber composites can be treated as if parallel slaps of the two materials bonded together with relative thicknesses in proportion to the volume fractions of matrix and fibers

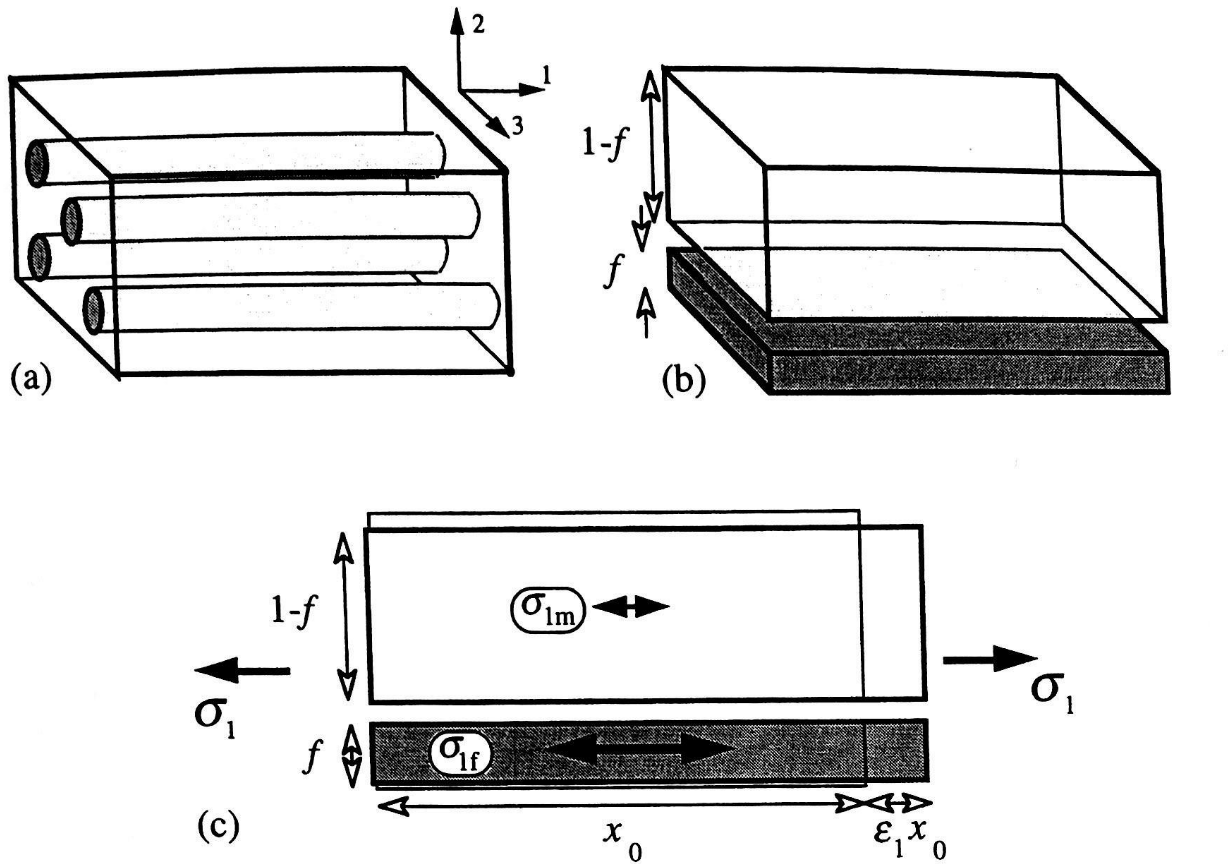


Fig. 4.1 Schematic illustration of (a) a composite containing a volume fraction f of aligned, continuous fibres, and (b) a representation of this as bonded slabs of matrix and fibre material. (c) On applying a stress σ_1 parallel to the fibre axis, the two slabs experience the same axial strain ϵ_1.

- ***Equal strain*** condition : ϵ_1 is equal to matrix and fibers
- No interfacial sliding condition : perfect bonding between matrix and fibers

$$\epsilon_1 = \epsilon_{1f} = \frac{\sigma_{1f}}{E_f} = \epsilon_m = \frac{\sigma_{1m}}{E_m}$$

- Fiber are much stiffer than the matrix : $E_f \gg E_m$

→ fillers is subject to much higher stresses than the matrix : $\sigma_{1f} \gg \sigma_{1m}$

→ The overall stress σ_1 is expressed with two contributions

$$\sigma_1 = (1-f)\sigma_{1m} + f\sigma_{1f}$$

→ The Young's modulus of the composite can be :

$$E_1 = \frac{\sigma_1}{\epsilon_1} = \frac{\left[(1-f)\sigma_{1m} + f\sigma_{1f}\right]}{\left(\sigma_{1f}/E_f\right)} = E_f\left[\frac{(1-f)\sigma_{1m}}{\sigma_{1f}} + f\right] = \boxed{(1-f)E_m + fE_f}$$

Rule of mixture

- Rule of mixture is valid for the composites with fibers which are long enough for the equal strain assumption

- **3.2. Transverse stiffness**

- Prediction of the transverse stiffness of a composite from the elastic properties is far more difficult than the axial value

- Slab model is again assumed as a conventional approach

- ***Equal stress*** condition (commonly used to describe transverse properties) are used

$$\sigma_2 = \sigma_{2f} = \epsilon_{2f}E_f = \sigma_{2m} = \epsilon_{2m}E_m$$

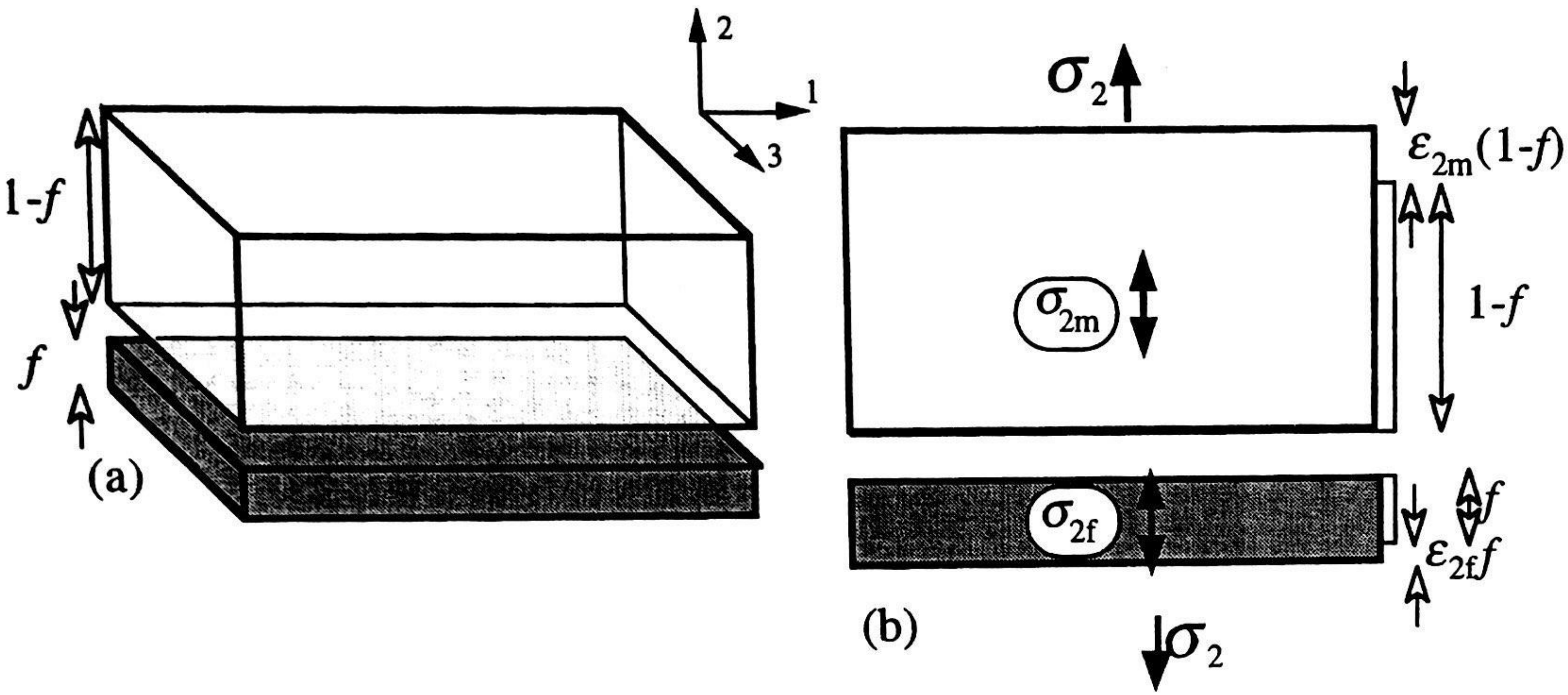


Fig. 4.2 Schematic showing (a) the slab model and (b) the 'equal stress' assumption during transverse stressing.

- The overall net strain is :

$$\epsilon_2 = f\epsilon_{2f} + (1-f)\epsilon_{2m}$$

- The composite modulus is :

$$E_2 = \frac{\sigma_2}{\epsilon_2} = \frac{\sigma_{2f}}{\left[f\epsilon_{2f} + (1-f)\epsilon_{2m}\right]} = \boxed{\left[\frac{f}{E_f} + \frac{(1-f)}{E_m}\right]^{-1}}$$

Called “Reuss model” (equal stress treatment)

- Reuss model is simple and convenient, but gives poor approximation for E_2.

→ Why? The regions of the matrix in parallel with fibers are constrained to have the **same (low) strain as the fibers** → The matrix carries **a low stress than a real case**.

LFT composites : 삼양사

https://www.youtube.com/watch?v=q78Hy7JQIV8

LFT composites : automotive

https://www.youtube.com/watch?v=uH-sXrf4gJM

https://www.youtube.com/watch?v=h9oByiPsYF8

- Strain field visualization :

- A square mesh represents unstrained material, which becomes distorted on loading the composites
- This strain is distributed inhomogeneously within the matrix
- This inhomogeneity with sharp concentrations of stress in certain locations is very significant in terms of the onset of non-elastic behavior
- Inhomogeneity also results from the **interfacial de-bonding, matrix plastic deformation** and **micro-cracking**

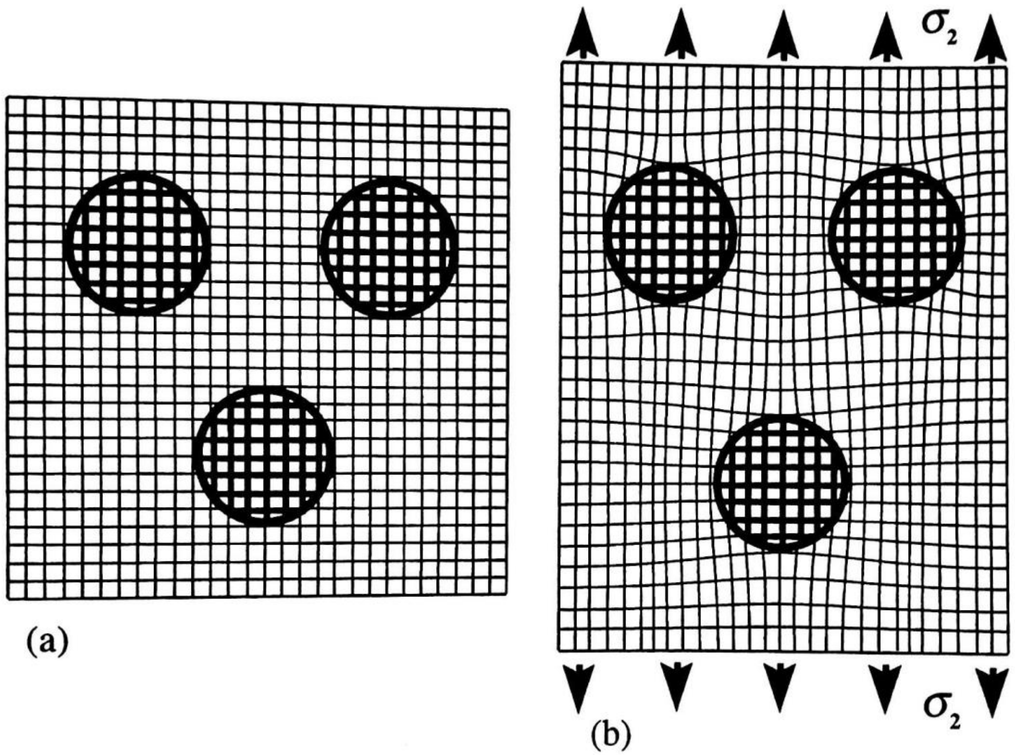


Fig. 4.3 Schematic illustration of the strain field in a composite transverse to an hexagonal array of parallel fibres which are much stiffer than the matrix. (a) Unstrained and (b) on application of a transverse stress in the vertical direction.

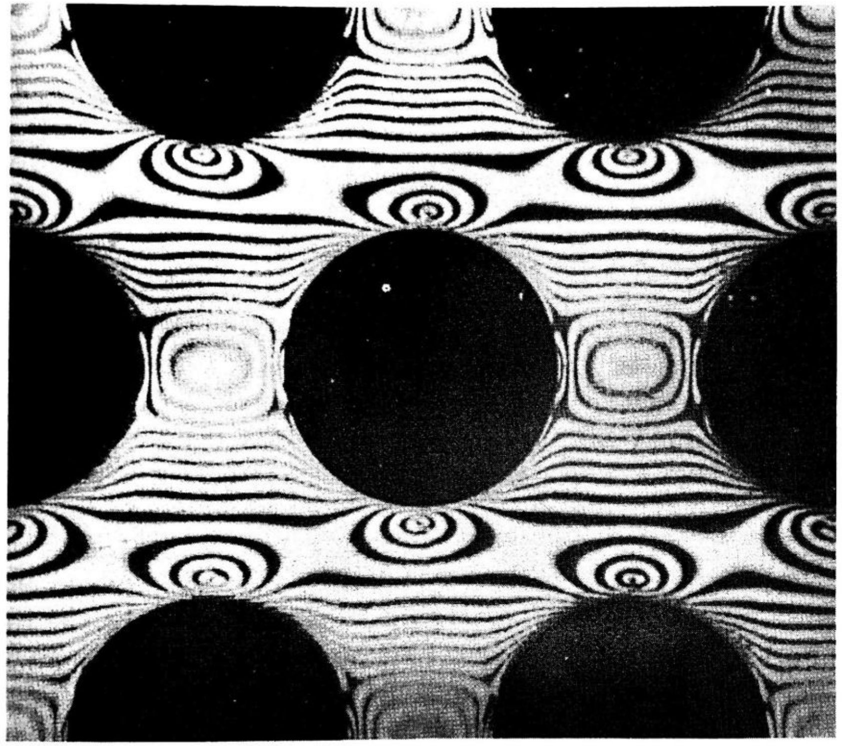


Fig. 4.4 A photoelastic image (Puck 1967), showing isochromatic (equal bi-refringence) fringes for a macromodel composite loaded in transverse tension (vertical direction).

- Halpin and Tsai model (1967) : Semi-empirical expression

  - The non-uniform distribution of stress and strain during transverse loading means that **the simple equal stress model (the slap model) is inadequate**

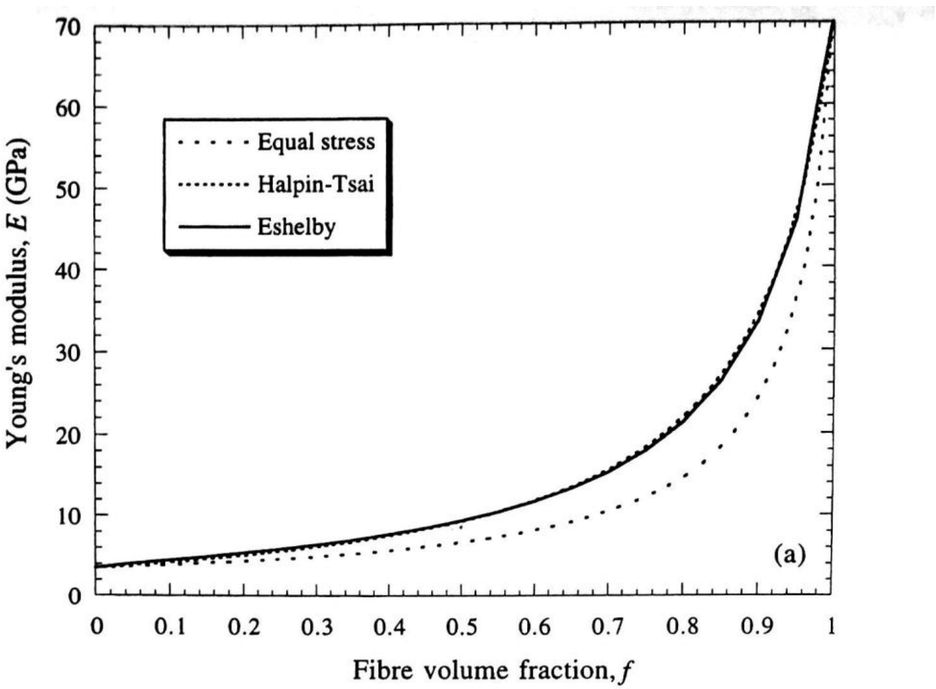
  - The slap model gives an underestimates of the E_2 (transverse) Young's modulus and gives an lower bound

  - **Enhanced fiber load bearing (Halpin and Tsai model)** relative to the equal stress assumption corrects the underestimated E_2

$$E_2 = \frac{E_m(1+\xi\eta f)}{(1-\eta f)} \qquad \text{where,} \quad \eta = \frac{\left(\frac{E_f}{E_m}-1\right)}{\frac{E_f}{E_m}+\xi}$$

Adjustable parameter

ξ ← often taken to have a value of around unity



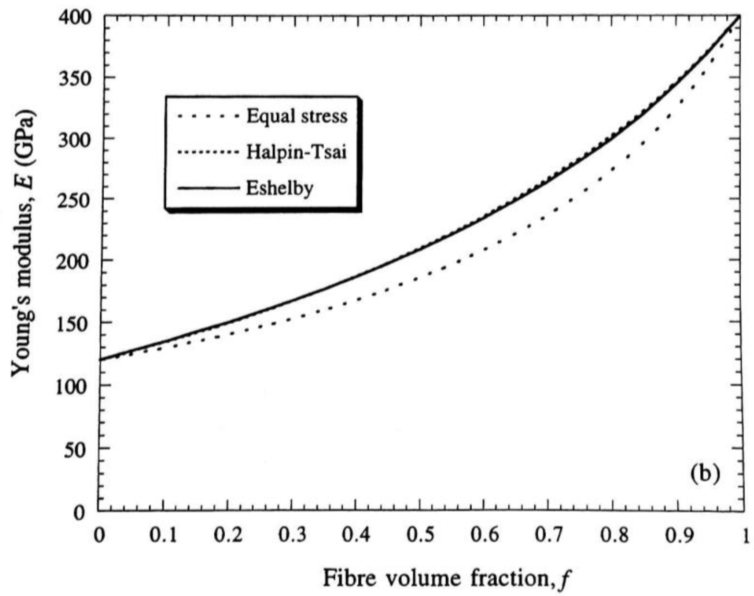


Fig. 4.6 Predicted dependence on fibre volume fraction of the transverse Young's moduli of continuous-fibre composites, according to the equal stress, Eqn (4.6), Halpin–Tsai, Eqn (4.7) and Eshelby models for (a) glass fibres in epoxy and (b) silicon carbide fibres in titanium.

- **3.3. Shear stiffness**

  - The shear moduli of composites (G_{12}) can be predicted using the slab model
  - The transverse stiffness is :

$$\tau_{12} = \tau_{12f} = \gamma_{12f} G_f = \tau_{12m} = \gamma_{12m} G_m$$

γ_{12f}, γ_{12m} : individual shear strains in the fiber and matrix

γ_{12} : total strain in the composite

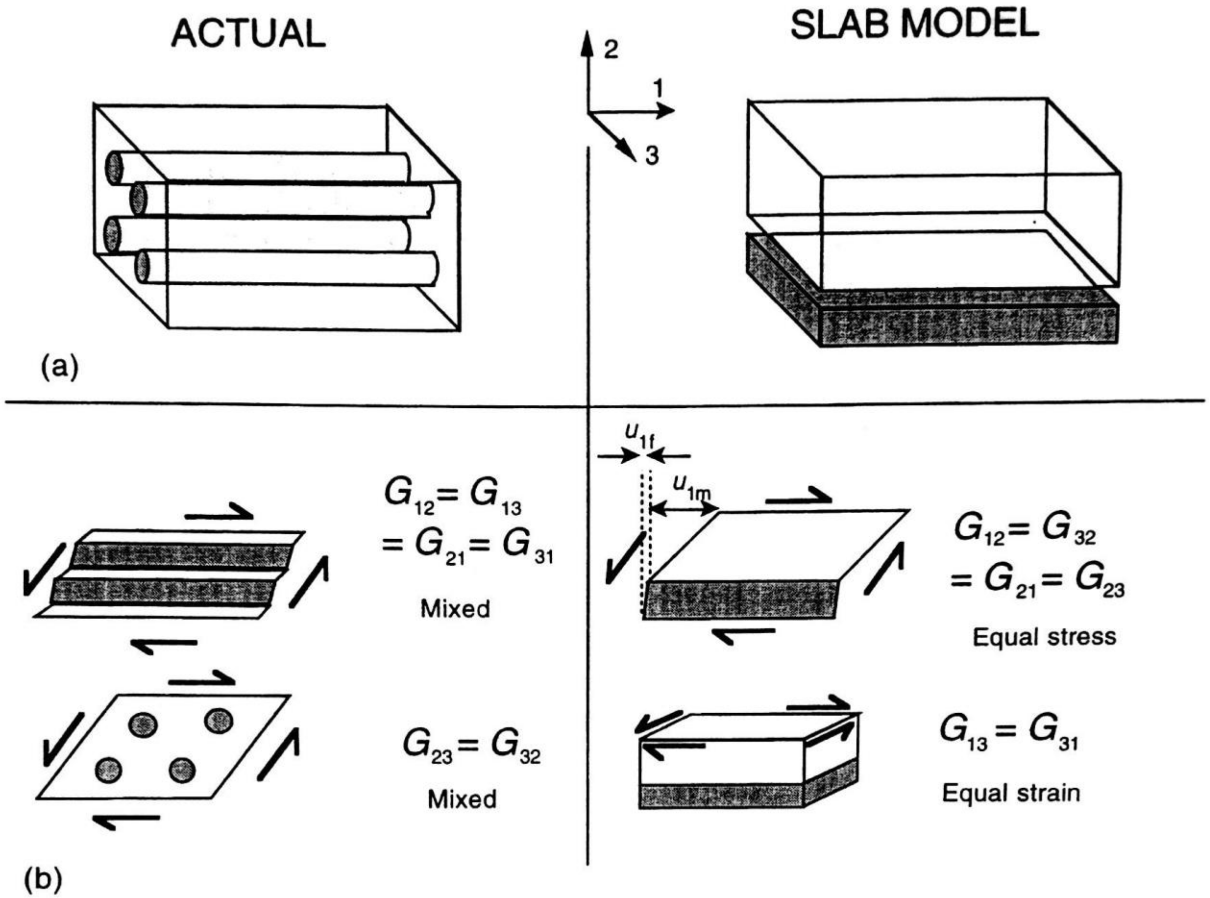


Fig. 4.7 Schematic illustration of how the shear moduli are defined for a real fibre composite and for the slab model representation, indicating how stress and strain partition between the two constituents in each case.

- The shear moduli of composites (G_{12}) can be predicted using the slab model
- The transverse stiffness is :

$$\gamma_{12} = \frac{(u_{1f} + u_{1m})}{f + (1-f)} = f\gamma_{12f} + (1-f)\gamma_{12m}$$

Equal stress assumption: $\tau_{12f} = \tau_{12m}$

$$\therefore G_{12} = \frac{\tau_{12}}{\gamma_{12}} = \frac{\tau_{12f}}{f\gamma_{12f} + (1-f)\gamma_{12m}} = \left[\frac{f}{G_f} + \frac{(1-f)\gamma_{12m}}{\tau_{12f}}\right]^{-1} = \left[\frac{f}{G_f} + \frac{(1-f)}{G_m}\right]^{-1}$$

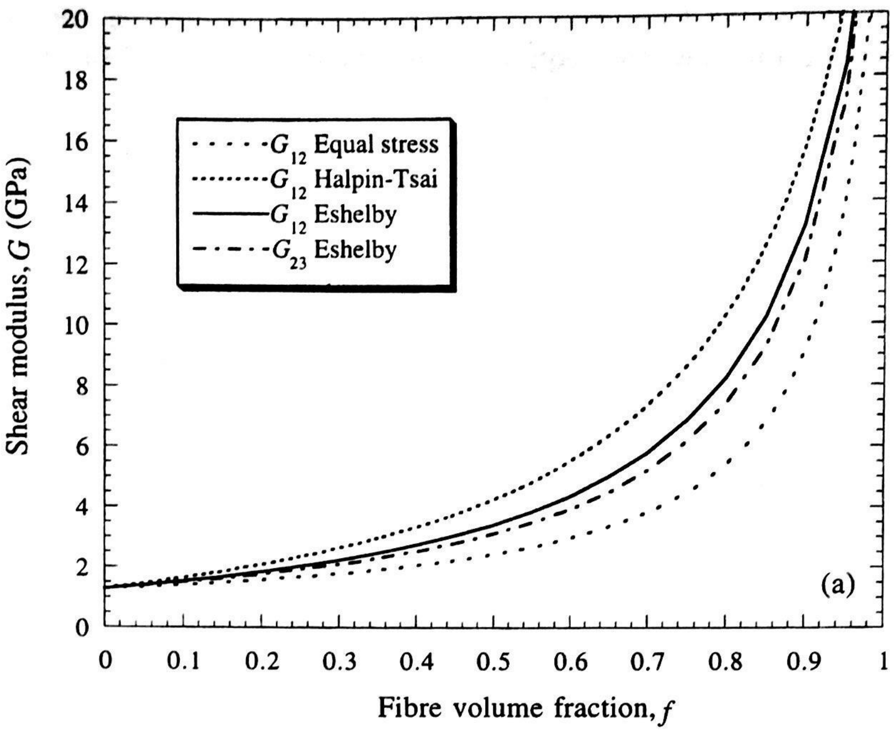
- As already mentioned, equal stress assumption gives very poor results on G_{12} too!
- Halpin and Tsai model :

$$G_{12} = \frac{G_m(1+\xi\eta f)}{(1-\eta f)}$$

where, $\eta = \dfrac{\left(\frac{E_f}{E_m} - 1\right)}{\frac{E_f}{E_m} + \xi}$

Adjustable parameter

often taken to have a value of around unity



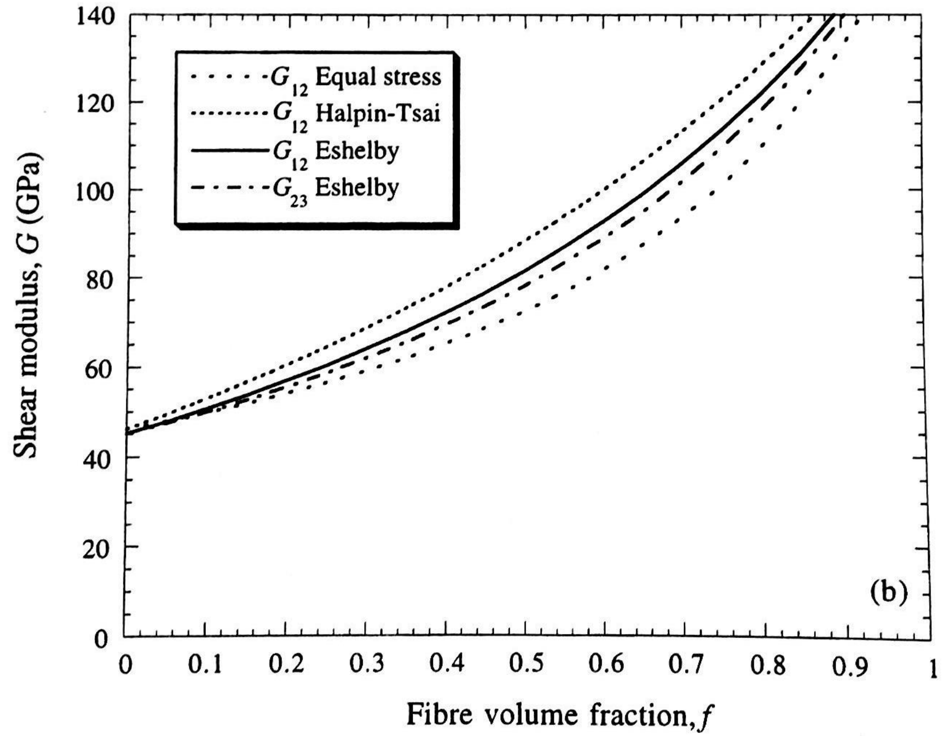


Fig. 4.8 Predicted dependence on fibre volume fraction of the shear moduli of continuous-fibre composites, according to the equal stress equation for G_{12}, Eqn (4.8), the Halpin–Tsai expression for G_{12}, Eqn (4.10) and the Eshelby model (G_{12} and G_{23}). Data are shown for (a) glass fibres in epoxy and (b) silicon carbide fibres in titanium.

- Halpin and Tsai model gives a fairly good approximation to the axial shear modulus

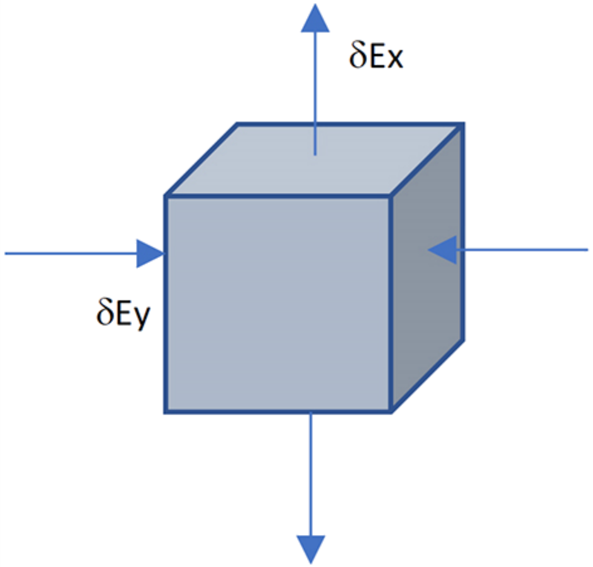
• 3.4. Poisson contraction effects

- The Poisson's ratio ν_{ij} describes the contraction in the *j*-direction on applying a stress in the *i*-direction.

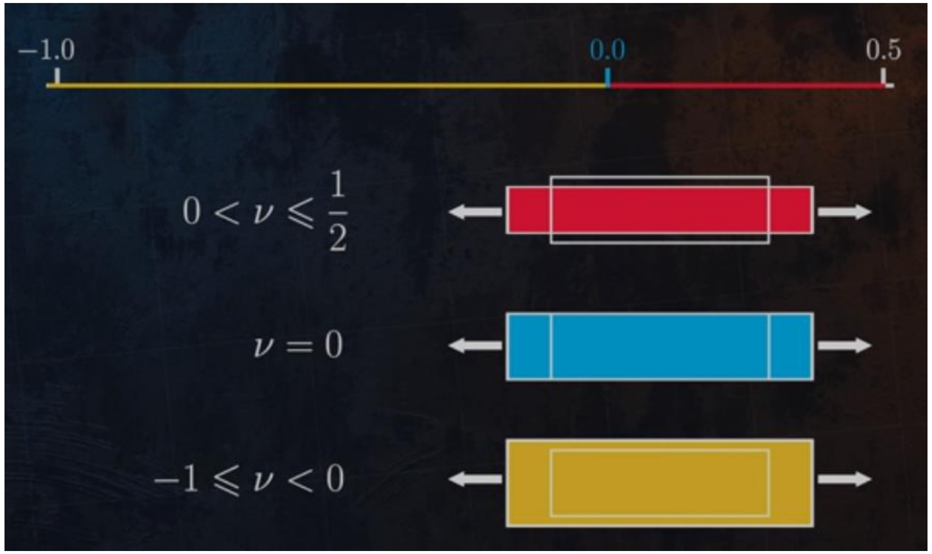
- The Poisson's ratio :

$$\nu_{ij} = -\frac{\epsilon_j}{\epsilon_i}$$

Material	Poisson's ratio
rubber	0.4999[9]
gold	0.42–0.44
saturated clay	0.40–0.49
magnesium	0.252–0.289
titanium	0.265–0.34
copper	0.33
aluminium-alloy	0.32
clay	0.30–0.45
stainless steel	0.30–0.31
steel	0.27–0.30
cast iron	0.21–0.26
sand	0.20–0.455
concrete	0.1–0.2
glass	0.18–0.3
metallic glasses	0.276–0.409[16]
foam	0.10–0.50
cork	0.0



Curiosity : Negative Poisson Ratio (NPR) = (Auxetic) materials exist?



Ref) wikipedia

Material	Plane of symmetry	ν_{xy}	ν_{yx}	ν_{yz}	ν_{zy}	ν_{zx}	ν_{xz}
Nomex honeycomb core	x-y, ribbon in x direction	0.49	0.69	0.01	2.75	3.88	0.01
glass fiber-epoxy resin	x-y	0.29	0.32	0.06	0.06	0.32	

- Understanding Poission's ratio

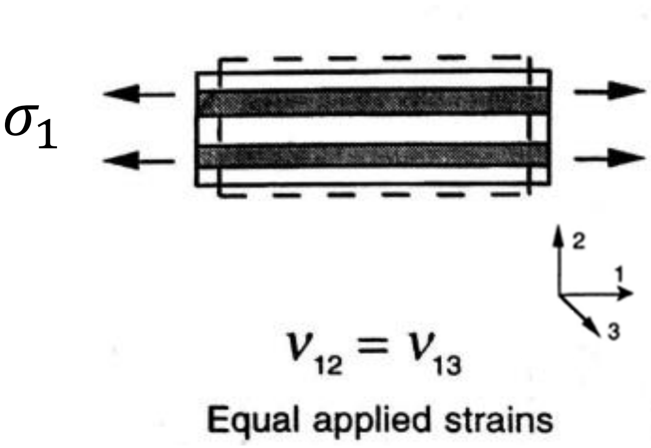
https://www.youtube.com/watch?v=tuOlM3P7ygA

- Negative Poission's ratio

https://www.youtube.com/watch?v=oLU-k7LhtZI

- For an aligned fiber composite, **three different Poisson's ratios** :

1)



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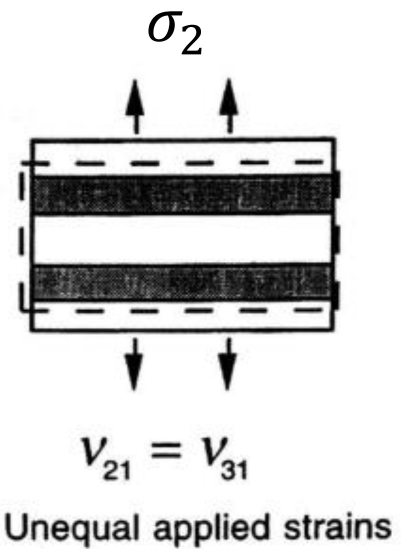

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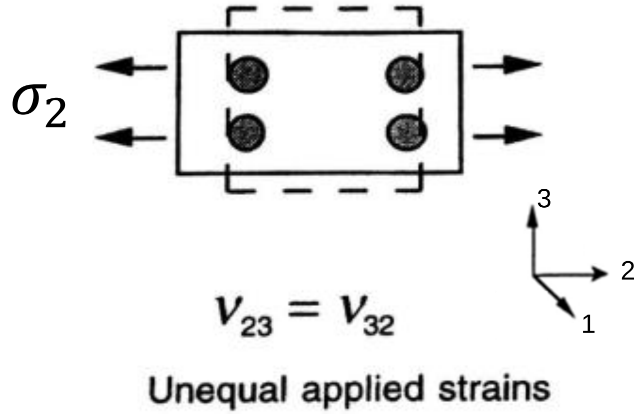


- For orthotropic materials in a general case:

$$\frac{\nu_{12}}{E_1} = \frac{\nu_{21}}{E_2}$$

$$G_{23} = \frac{E_2}{2(1+\nu_{23})}$$

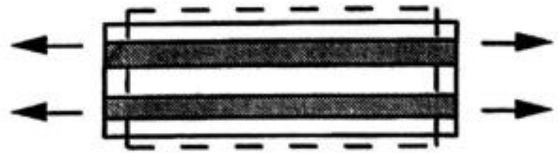
- For a slab model with equal strain assumption:

$$\epsilon_{2f} = -\nu_f \epsilon_{1f} = -\nu_f \frac{\sigma_{1f}}{E_f}$$

$$\epsilon_{2m} = -\nu_m \epsilon_{1m} = -\nu_m \frac{\sigma_{1m}}{E_m}$$

1)

σ_1



$\nu_{12} = \nu_{13}$

Equal applied strains

So that

Rule of mixture due to the assumption of Equal strain : f x E_f + (1-f) x E_m

$$\epsilon_2 = -\left[f\nu_f \underbrace{\frac{\sigma_{1f}}{E_f}}_{\epsilon_1} + (1-f)\nu_m \underbrace{\frac{\sigma_{1m}}{E_m}}_{\epsilon_1} \right] = -\left[f\nu_f\epsilon_1 + (1-f)\nu_m\epsilon_1 \right]$$

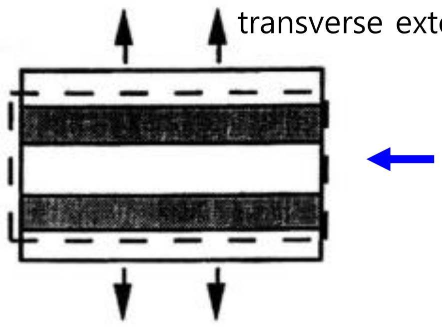
$$\boxed{\therefore \nu_{12} = -\frac{\epsilon_2}{\epsilon_1} = f\nu_f + (1-f)\nu_m}$$

"The result also follows the rule of mixture"

: Equal strain assumption is accurate for axial stressing of the composites.

- The ratio of the axial contraction to the transverse extension on stressing transversely is obtained from the reciprocal relationship :

2)



$$\frac{\nu_{12}}{E_1} = \frac{\nu_{21}}{E_2}$$

$$\boxed{\nu_{21} = [f\nu_f + (1-f)\nu_m]\frac{E_2}{E_1}}$$

> 1

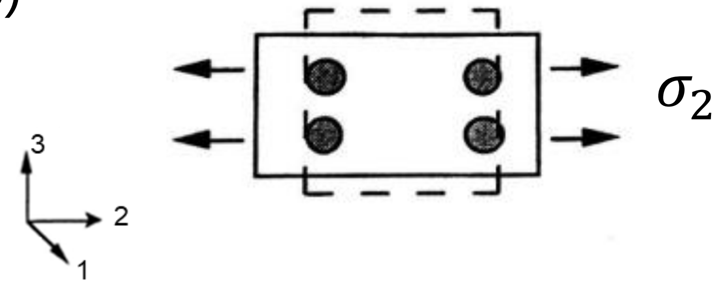
$\boxed{\nu_{21} < \nu_{12}.}$ Why?

On stressing transversely, the fibers offer strong resistance to axial contraction!

(Can be double-checked with $E_2 < E_1$)

- In the other transverse direction, ν_{23} is expected to be very high.

3)



$\nu_{23} = \nu_{32}$

Unequal applied strains

- ν_{23} can be obtained by considering the overall volume change :

$$\Delta = \epsilon_1 + \epsilon_2 + \epsilon_3 = \frac{\sigma_H}{K}$$

σ_H : applied hydrostatic stress

K : bulk modulus of the composite

σ_2 is only applied here, so that

$$\sigma_H = \frac{\sigma_1 + \sigma_2 + \sigma_3}{3} = \frac{\sigma_2}{3}$$

$$\epsilon_3 = \frac{\sigma_2}{3K} - \epsilon_1 - \epsilon_2$$

This directly leads to

$$\nu_{23} = -\frac{\epsilon_3}{\epsilon_2} = -\frac{\sigma_2}{3K\epsilon_2} + \frac{\epsilon_1}{\epsilon_2} + 1$$

$$\boxed{\therefore\ \nu_{23} = -\frac{E_2}{3K} - \nu_{21} + 1}$$

The accuracy of ν_{23} is largely dependent on E_2

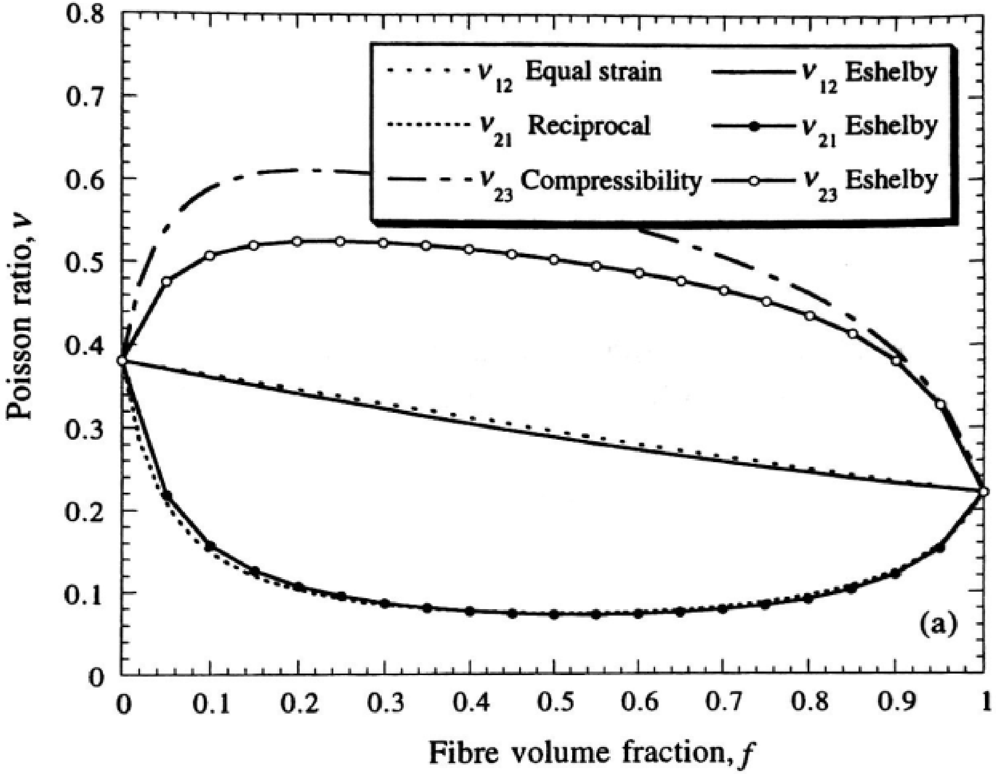
- The bulk modulus of the composite can be estimated by an equal stress assumption, which should be quite accurate in this case : (why? Due to bulk property assumption)

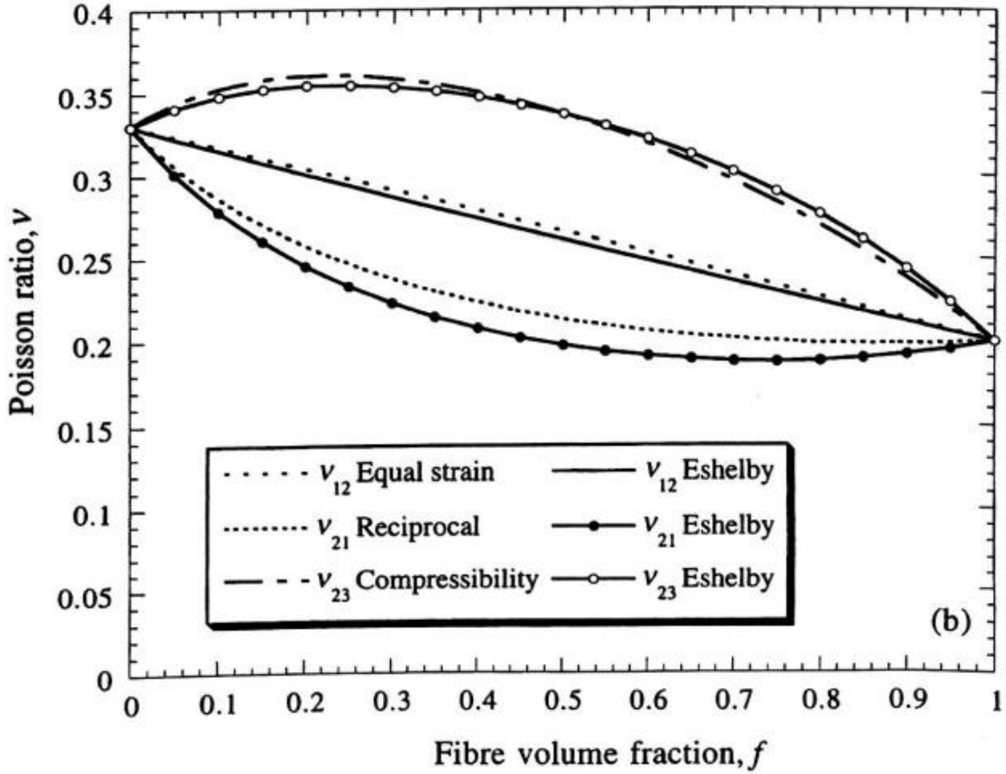
$$\sigma_H = \Delta_f K_f = \Delta_m K_m \quad \text{and} \quad \Delta = f\Delta_f + (1-f)\Delta_m$$

So,

$$\boxed{K = \frac{\sigma_H}{\Delta} = \left[\frac{f}{K_f} + \frac{(1-f)}{K_m}\right]^{-1}}$$

Equal strain and reciprocal relation quite predict well by comparing with Eshelby model





So far we have learned,

- Polymer & Composites processing technique
- A review of Navier-Stokes equation for polymer flow
- Fiber reinforced polymer composites (FRP) theory

Polymers Composites → Polymer Rheology → Recent industry

- Battery packing, Battery materials, Battery slurry coating
- Multifunctional nanocomposites
- Strong fibers, fiber composites
- Biomedical applications