

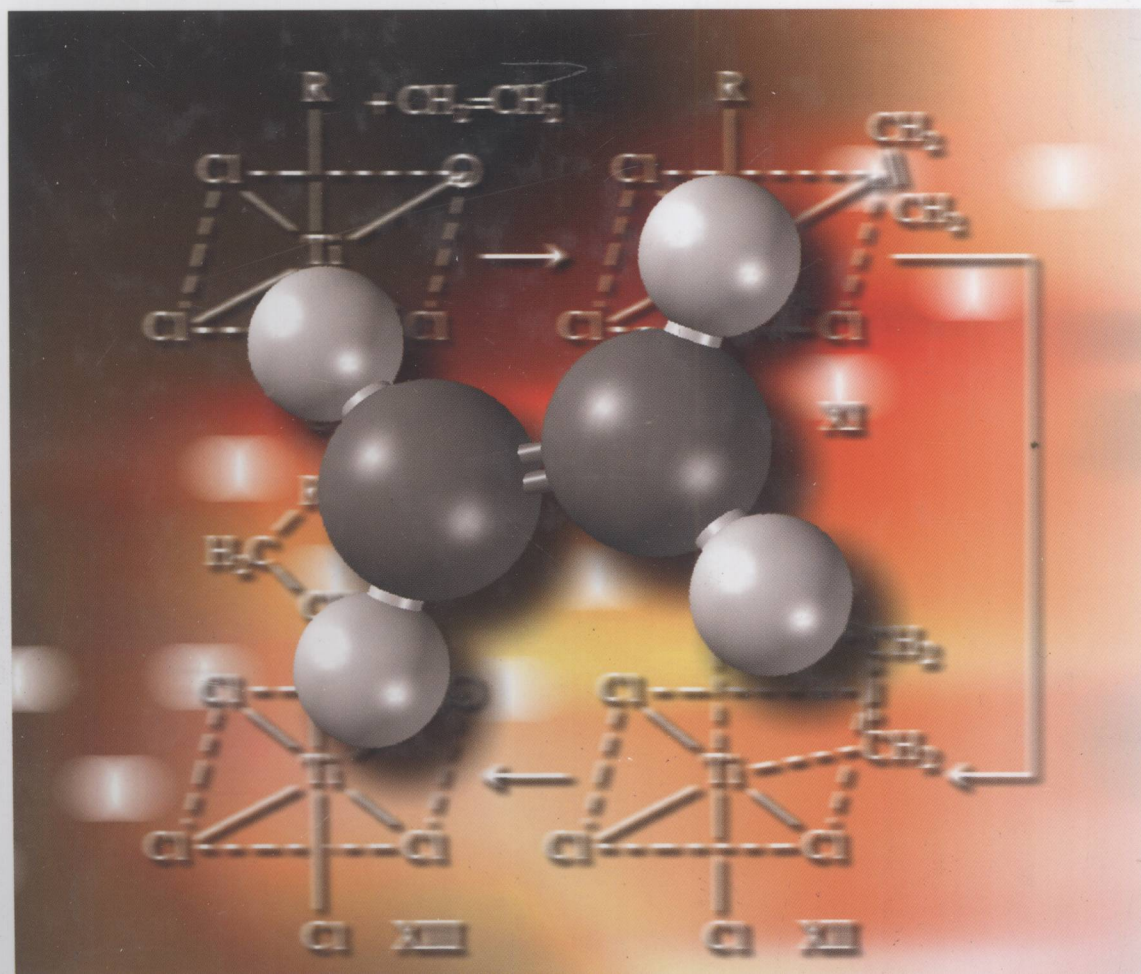
Hans-Georg Elias

 WILEY-VCH

# Macromolecules

**Volume 1:**

Chemical Structures and Syntheses



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*Hans-Georg Elias*

# Macromolecules

Volume 1: Chemical Structures and Syntheses



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- Volume 2 Industrial Polymers and Syntheses
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- Volume 4 Processing and Applications of  
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Didici in mathematicis ingenio,  
in natura experimentis,  
in legibus divinis humanisque auctoritate,  
in historia testimoniis nitendum esse.

Gottfried Wilhelm Leibniz  
(1646-1716)

(I learned that  
in mathematics one depends on inspiration,  
in science on experimental evidence,  
in the study of divine and human law on authority, and  
in historical research on authenticated sources.)

## Preface

Macromolecular science has grown substantially since the first German edition of this book was written (1962-1970). The German editions ballooned from the single volume of the first edition (1971) with 856 pages to the four volumes of the sixth edition (1999-2002) with a total of 2564 pages. This English edition is based on the latest (sixth) German edition. It is not, however, a cover-to-cover translation, but has been completely checked, modified, corrected, and updated where necessary. More than 25 % of the figures of volume I are new and so are many tables and derivations.

Like its German predecessor, the English-language edition comprises four volumes. Volume I is concerned with the fundamentals of chemical structure and synthesis of polymers, Volume II with individual polymers and their industrial syntheses, Volume III with fundamentals of physical structures and properties, and Volume IV with processing and applications of polymers as plastics, fibers, elastomers, thickeners, etc.

Chapter I of Volume I introduces the field by discussing basic terms and the arguments and counter-arguments from which the science of macromolecules arose. It is followed by four chapters that outline the constitution (Ch. 2), configuration (Ch. 4), and conformation (Ch. 5) of macromolecules, and the most common methods for the characterization of macromolecules (Chapter 3) that are used most frequently in the synthesis of polymers.

The remaining chapters are concerned with general aspects of polymerization, including statistics and polymerization of chiral monomers (Ch. 6), polymerization equilibria (Ch. 7), ionic polymerizations (Ch. 8), coordination polymerizations (Ch. 9), free-radical polymerizations (Ch. 10), polymerizations by radiation or in ordered states (Ch. 11), copolymerizations (Ch. 12), polycondensations and polyadditions (Ch. 13), biological polymerizations (Ch. 14), reactions of macromolecules (Ch. 15), and molecular engineering for the preparation of new polymer architectures (Ch. 16). The Appendix lists SI units and their prefixes, IUPAC symbols for physical quantities, multiplicative prefixes for numerals used in systematic names, definitions of concentrations, and names of ratios of physical quantities. Most chapters contain historical notes which list the ground-breaking early papers in the field. All chapters are followed by extensive lists of books and reviews, including some older ones that contain information which is rarely found in newer books on the same subject.

I am indebted to my good friends and former colleagues at Michigan Molecular Institute, Professors Petar R. Dvornic and Steven E. Keinath, who read and checked the final draft of all chapters and made many helpful suggestions. The remaining factual and judgmental errors are, of course, all mine.

Midland, Michigan  
Summer 2005

Hans-Georg Elias

# List of Symbols

Symbols for physical quantities follow the recommendations of the International Union of Pure and Applied Chemistry (IUPAC), symbols for physical units those of the International Standardization Organization (ISO). Exceptions are indicated.

I.Mills, T.Cvitas, K.Homann, N.Kallay, K.Kuchitsu, Eds., (International Union of Pure and Applied Chemistry, Division of Physical Chemistry), "Quantities, Units and Symbols in Physical Chemistry", Blackwell Scientific Publications, Oxford 1988.

## Symbols for Languages

D = German (deutsch), F = French, G = (classic) Greek, L = (classic) Latin.

The Greek letter  $\upsilon$  (upsilon) was transliterated as "y" (instead of the customary "u") in order to make an easier connection to written English (example:  $\pi\omicron\lambda\upsilon\varsigma$  = polys (many)). For the same reason,  $\chi$  was transliterated as "ch" and not as "kh."

## Symbols for Chemical Structures

- A: symbol for a monomer or a leaving group (polycondensations)
- B: symbol for a monomer or a leaving group (polycondensations)
- L: symbol for a leaving molecule, for example,  $\text{H}_2\text{O}$  from the reaction of  $-\text{COOH} + \text{HO}-$
- R: symbol for a monovalent ligand, for example,  $\text{CH}_3-$  or  $\text{C}_6\text{H}_5-$
- Z: symbol for a divalent unit, for example,  $-\text{CH}_2-$  or  $-p\text{-C}_6\text{H}_4-$
- Y: symbol for a trivalent unit, for example,  $-\text{C}(\text{R})<$  or  $-\text{N}<$
- X: symbol for a tetravalent unit, for example,  $>\text{C}<$  or  $>\text{Si}<$
- \*: symbol for an active site: radical ( $\bullet$ ), anion ( $^\ominus$ ), cation ( $^\oplus$ )
- pPh* *para*-phenylene (in text)
- p-C<sub>6</sub>H<sub>4</sub>* *para*-phenylene (in line formulas)

## Mathematical Symbols (IUPAC)

=	equal to	>	greater than
≠	not equal to	≥	greater than or equal to
≡	identically equal to	>>	much greater than
≈	approximately equal to	<	less than
~	proportional to (IUPAC: ~ or ∞)	≤	less than or equal to
≐	corresponds to	<<	much less than
→	approaches, tends to	±	plus or minus
Δ	difference	sin	sine of
δ	differential	cos	cosine of
f	function of (IUPAC: <i>f</i> )	tan	tangent of
Σ	sum	cot	cotangent of
∫	integral	sinh	hyperbolic sine of
Π	product	arcsin	inverse sine of
lg	logarithm to the base 10		(IUPAC: lg or log <sub>10</sub> )
ln	logarithm to the base e (natural logarithm)		(IUPAC: ln or log <sub>e</sub> )



**Averages and Other Markings**

- line above letter indicates common average, for example,  $\overline{M}_n$  = number-average of molar mass (note: subscript is not italicized since it does *not* represent a physical quantity that is kept constant)
- ~ tilde indicates a partial quantity, for example,  $\tilde{v}_A$  = partial specific volume of component A
- [ ] square brackets surrounding the symbol of the substance indicates the amount concentration ("mole concentration"), usually in mol/L
- ⟨ ⟩ angled brackets surrounding a letter indicate spatial averages, for example,  $\langle s^2 \rangle$  = mean-square average of radius of gyration

**Exponents and Superscripts**

- ° degree of plane angle [= ( $\pi/180$ ) rad]
- ' minute of plane angle [= ( $\pi/10\ 800$ ) rad]
- " second of plane angle [= ( $\pi/648\ 000$ ) rad]
- o pure substance
- $\infty$  infinite, for example, dilution or molecular weight
- m amount-of-substance related quantity if a subscript is inexpedient. According to IUPAC, <sup>m</sup> can be used as either superscript or subscript
- (q) qth order of a moment (always in parentheses since it does not represent a power)
- ‡ activated quantity, for example,  $E^\ddagger$  = activation energy
- $\alpha$  exponent in the intrinsic viscosity–molecular weight relationship,  $[\eta] = K_v M^\alpha$

**Indices and Subscripts**

- o standard or original state, for example,  $T_o$  = reference temperature
- 0 state at time zero
- 1 solvent
- 2 solute (usually polymer)
- 3 additional component (for example: salt, precipitant, etc.)
- $\infty$  final state
- A substance A, e.g.,  $M_A$  = molar mass of substance A
- a group, monomeric unit from substance A, e.g., mass  $m_a$
- a number of functional groups A in a molecule  $A_aB$
- am amorphous
- B substance B
- b group, e.g., monomeric unit from substance B
- b number of functional groups B in a molecule  $AB_b$
- bd bond, especially chain bond
- be effective bond length (= length of monomer unit projected onto the chain direction)
- bp boiling temperature (boiling point)
- br branch, branched
- c ceiling, for example,  $T_c$  = ceiling temperature
- ch chain (L: *catena*)
- comb combination

cr	crystalline
crit	critical
cryst	crystallization
cycl	cyclic
D	related to diffusion
eff	effective
end	end-group
eq	equilibrium
f	free or functional
G	glass transformation
g	any statistical weight, e.g., n, m, z or x, w, Z
H	hydrodynamically effective property or hydration
h	hydrodynamic average
I	initiator, $I^\bullet$ = initiator radical
<i>i</i>	<i>i</i> th component (variable)
i	initiation
i	isotactic diad (IUPAC recommends m = <i>meso</i> ; see Chapter 4)
ii	isotactic triad (IUPAC: mm)
is	heterotactic triad (IUPAC: mr)
<u>is</u>	sum of heterotactic triads, <u>is</u> = is + si
it	isotactic
<i>j</i>	variable
<i>k</i>	variable
L	liquid, melt (L: <i>liquidus</i> )
l	liquid
M	melting
M	monomer molecule
Mt	metal
m	molar (also as superscript)
md	median
mol	molecule
mon	monomer
mu	monomeric unit
n	number average (note: not in italics since it does not refer to a physical quantity that is held constant)
P	polymer
<i>p</i>	quantity at constant pressure
p	propagation
pm	polymerization, especially propagation
pol	polymer

q	index, defined differently for each section or chapter
R	reactive entity
R	retention
r	relative (only in $M_r$ = relative molecular mass = molecular weight)
r	based on end-to-end distance, e.g., $\alpha_r$ = linear expansion coefficient of a coil (with respect to the end-to-end distance)
red	reduced
rel	relative
rep	repeating unit
S	solvating solvent
s	syndiotactic diad (IUPAC recommends $r$ = <i>racemo</i> )
s	related to radius of gyration
seg	segment
si	heterotactic triad (IUPAC: $rm$ )
soln	solution
ss	syndiotactic triad (IUPAC: $rr$ )
$T$	quantity at constant temperature
t	termination
tr	transfer
u	monomeric unit in polymer
u	conversion
$V$	quantity at constant volume
v	viscosity average (solutions)
w	mass average ("weight average"); note: not in italics since it does not refer to a physical quantity that is held constant
x	crosslink(ed)
z	z average

**Prefixes of Words** (in systematic polymer names in *italics*)

alt	alternating
at	atactic
blend	polymer blend
block	block (large constitutionally uniform segment)
br	branched. IUPAC recommends sh-branch = short chain branch, l-branch = long chain branch, f-branch = branched with a branching point of functionality $f$
co	joint (unspecified)
comb	comb
compl	polymer-polymer complex
cyclo	cyclic
ct	cis-tactic

eit	erythrodiisotactic
g	graft
ht	heterotactic
ipn	interpenetrating network
it	isotactic
net	network; $\mu$ -net = micro network
per	periodic
r	random (Bernoulli distribution)
sipn	semi-interpenetrating network
star	star-like. $f$ -star, if the functionality $f$ is known; $f$ is then a number
st	syndiotactic
stat	statistical (unspecified distribution)
tit	threodiisotactic
tt	trans-tactic

### Other Abbreviations

AIBN	$N,N'$ -azobisisobutyronitrile
BPO	dibenzoylperoxide
Bu	butyl group ( $i$ Bu = isobutyl group; $n$ Bu = normal butyl group (according to IUPAC, the normal butyl group is not characterized by $n$ , which rules out Bu as an unspecified butyl group); $s$ Bu = secondary butyl group; $t$ Bu = tertiary butyl group)
Bz	benzene
C	catalyst; $C^*$ = active catalyst or active catalytic center
cell	cellulose residue
Cp	cyclopentadienyl group
DMF	$N,N$ -dimethylformamide
DMSO	dimethylsulfoxide
Et	ethyl group
G	gauche conformation
glc	glucose
GPC	gel permeation chromatography
I	initiator
IR	infrared
L	solvent (liquid)
LC	liquid-crystalline

Me	methyl group
Mt	metal atom
Np	naphthalene
NMR	nuclear magnetic resonance
P	polymer
Ph	phenyl group
Pr	propyl group
SEC	size exclusion chromatography
THF	tetrahydrofuran
UV	ultraviolet

### Quantity Symbols (unit symbols, see Chapter 17, Appendix)

Quantity symbols follow in general the recommendations of IUPAC.

$A$	absorption ( $A = \lg(I_0/I) = \lg(1/\tau_t)$ ); formerly: extinction
$A$	area; $A_c$ = cross-sectional area of a chain
$A$	Helmholtz energy ( $A = U - TS$ ); formerly: free energy
$A^\ddagger$	pre-exponential constant (in $k = A^\ddagger \exp(-E^\ddagger/RT)$ )
$a$	thermodynamic activity
$a$	linear absorption coefficient ( $a = (1/L) \lg(I_0/I)$ )
$b$	bond length; $b_{\text{eff}}$ = effective bond length
$C$	number concentration (number of entities per total volume); see also $c$
$[C]$	amount-of-substance concentration of substance $C$ = amount of substance $C$ per total volume = "molar concentration of $C$ "
$C$	transfer constant (always with index, e.g., $C_r$ of a regulator, $C_s$ of a solvent)
$C$	heat capacity (usually in J/K); $C_p$ = isobaric heat capacity (heat capacity at constant pressure $p$ ); $C_V$ = isochoric heat capacity (heat capacity at constant volume $V$ ); $C_m$ = molar heat capacity (heat capacity per amount-of-substance $n$ )
$C_{tr}$	chain transfer constant of polymerizations ( $C_{tr} = k_{tr}/k_p$ )
$c$	specific heat capacity (usually in J/(g K)); $c_p$ = isobaric specific heat capacity; $c_V$ = isochoric specific heat capacity. Formerly: specific heat
$c$	concentration = mass concentration (= mass-of-substance per total volume) = "weight concentration." IUPAC calls this quantity "mass density" (quantity symbol $\rho$ ). The quantity symbol $c$ has, however, traditionally been used for a special case of mass concentration, i.e., mass-of-substance per volume of solution and the quantity symbol $\rho$ for another special case, the mass density ("density") = mass-of-substance per volume of substance.
$d$	diameter; diameter of blob ( $d_{\text{blob}}$ ), of sphere ( $d_{\text{sph}}$ ), etc.
$E$	energy
$e$	parameter in the $Q,e$ copolymerization equation
$e$	cohesion energy density

$F$	force
$f$	fraction (unspecified), see also $x$ , $w$ , $\phi$ , etc.
$f$	functionality
$G$	Gibbs energy ( $G = H - TS$ ); formerly: free enthalpy
$G$	statistical weight fraction ( $G_i = g_i / \sum_i g_i$ )
$g$	statistical weight (for example: $n$ , $x$ , $w$ ). IUPAC recommends $k$ for this quantity which is problematic because of the many other uses of $k$ . Similarly, $K$ cannot be used for the statistical weight fraction because of the many other meanings of $K$ .
$g$	parameter for the ratio of dimensions of branched macromolecules to those of unbranched macromolecules of equal molecular weight (branching index); $g_h$ = branching index from hydrodynamic measurements
$H$	height
$H$	enthalpy; $\Delta H_{\text{mix}}$ = enthalpy of mixing, $\Delta H_{\text{mix,m}}$ = molar enthalpy of mixing
$h$	Planck constant ( $h = 6.626\,075\,5 \cdot 10^{-34}$ J s)
$I$	light intensity
$i$	radiation intensity of a molecule
$i$	variable ( $i$ th component, etc.)
$J$	flux (of mass, volume, energy, etc.)
$K$	general constant; $K_n$ = equilibrium constant
$k$	rate constant (always with index); $k_i$ = rate constant of initiation; $k_p$ = rate constant of propagation, $k_t$ = rate constant of termination, $k_{tr}$ = rate constant of transfer
$k_B$	Boltzmann constant ( $k_B = R/N_A = 1.380\,658 \cdot 10^{-23}$ J K <sup>-1</sup> )
$L$	length (always geometric); $L_{\text{chain}}$ = true (historic) contour length of a chain (= number of chain bonds times length of valence bonds); $L_{\text{cont}}$ = conventional contour length of a chain (= length of chain in the all-trans macroconformation); $L_K$ = length of a Kuhn segment; $L_{ps}$ = persistence length; $L_{\text{seg}}$ = segment length
$l$	length
$M$	molar mass of a molecule (= physical unit mass/amount of molecule, e.g., g/mol). $\overline{M}_n$ = number-average molar mass; $\overline{M}_w$ = mass-average molar mass; $M_{\text{crit}}$ = critical molar mass; $\overline{M}_{R,n}$ = number-average molar mass of reactants (= polymer plus monomer)
$M_r$	relative molar mass = relative molecule mass = molecular weight (physical unit: 1 = "dimensionless"); $\overline{M}_{r,n}$ = number-average molecular weight
$m$	mass; $m_{\text{mol}}$ = mass of molecule
$N$	number of entities
$N_A$	Avogadro constant ( $N_A = 6.022\,136\,7 \cdot 10^{23}$ mol <sup>-1</sup> )
$n$	amount of substance (in mol); formerly: mole number
$n$	refractive index
$p$	conditional probability
$p$	pressure
$p$	extent of reaction (fractional conversion); $p_A$ = extent of reaction of A groups
$p$	number of conformational repeating units per completed helical turn

$Q$	electric charge = quantity of electricity
$Q$	heat
$Q$	parameter in the $Q,e$ copolymerization equation
$Q$	polymolecularity index (= "polydispersity index"), e.g., $Q = \overline{M}_w/\overline{M}_n$
$Q$	intermediate variable or constant, usually a ratio; varies with section
$q$	charge of an ion
$R$	molar gas constant ( $R \approx 8.314\ 510\ \text{J K}^{-1}\ \text{mol}^{-1}$ )
$R$	radius; $R_d$ = Stokes radius, $R_v$ = Einstein radius
$R$	rate of reaction; $R_p$ = rate of propagation, etc.
$R$	dichroic ratio
$r$	spatial end-to-end distance of a chain, usually as $\langle r^2 \rangle^{1/2}$ with various indices
$r$	copolymerization parameter
$r_0$	initial ratio of amounts of substances in copolymerizations
$S$	entropy; $\Delta S_{\text{mix}}$ = entropy of mixing, $\Delta S_{\text{mix,m}}$ = molar entropy of mixing
$S$	solubility coefficient
$T$	temperature (always with units). In physical equations always as thermodynamic temperature with unit kelvin; in descriptions, either as thermodynamic temperature (unit: kelvin) or as Celsius temperature (unit: degree Celsius). Mix-ups can be ruled out because the physical unit is always given. IUPAC recommends for the Celsius temperature either $t$ as a quantity symbol (which can be confused with $t$ for time) or $\theta$ (which can be confused with $\Theta$ for the theta temperature). $T_c$ = ceiling temperature, $T_G$ = glass temperature, $T_M$ = melting temperature
$T$	transparency
$t$	time
$t$	rotational angle around helix axis
$U$	internal energy
$u$	fractional conversion of monomer molecules ( $p$ = fractional conversion of groups; $y$ = yield of substance)
$u$	excluded volume
$V$	volume; $V_h$ = hydrodynamic volume, $V_m$ = molar volume; $\tilde{V}_m$ = partial molar volume
$v$	specific volume; $\tilde{v}$ = partial specific volume
$v$	linear velocity ( $v = dL/dt$ )
$W$	work
$w$	mass fraction = weight fraction
$X$	degree of polymerization of a molecule with respect to monomeric units (not to repeating units); $\bar{X}_n$ = number-average degree of polymerization of a substance; $\bar{X}_w$ = mass-average degree of polymerization of a substance
$x$	mole fraction (amount-of-substance fraction); $x_u$ = mole fractions of units, $x_i$ = mole fraction of isotactic diads, $x_{ii}$ = mole fraction of isotactic triads, etc.
$x_{br}$	degree of branching
$Y$	refractive index increment ( $= dn/dc$ )

$Y$	degree of polymerization with respect to repeating unit
$y$	yield of substance
$Z$	$z$ fraction ( $Z_i = z_i / \sum_i z_i$ )
$z$	$z$ -statistical weight
$z$	coordination number, number of neighbors
$\alpha$	angle, especially rotational angle of optical activity
$\alpha$	linear thermal expansion coefficient of materials ( $\alpha = (1/L)(dL/dT)$ ). Note: in literature often as $\beta$
$\alpha$	linear expansion of coils ( $\alpha_s$ if radius of gyration; $\alpha_r$ if end-to-end distance; $\alpha_h$ if hydrodynamic dimensions; $\alpha_D$ if diffusion; $\alpha_v$ if viscosity (dilute solutions))
$\alpha$	degree of crystallinity (with index for method: $X = X\text{-ray}$ , $d = \text{density}$ , etc.)
$[\alpha]$	"specific" optical rotation
$\beta$	angle
$\beta$	cubic thermal expansion coefficient [ $\beta = (1/V)(dV/dT)$ ]; in literature usually as $\alpha$
$\gamma$	angle
$\gamma$	crosslinking index
$\delta$	solubility parameter
$\delta$	chemical shift
$\epsilon$	cohesive energy
$\epsilon_r$	relative permittivity (formerly: dielectric constant)
$\eta$	dynamic viscosity, e.g., $\eta_0$ = viscosity at rest (Newtonian viscosity),
$\eta_1$	= viscosity of solvent, $\eta_e$ = extensional viscosity,
$\eta_r$	= $\eta/\eta_1$ = relative viscosity,
$\eta_i$	= $(\eta - \eta_1)/\eta_1$ = relative visc. increment (= specific viscosity $\eta_{sp}$ ),
$\eta_{inh}$	= $(\ln \eta_r)/c$ = inherent viscosity (= logarithmic visc. number),
$\eta_{red}$	= $(\eta - \eta_1)/(\eta_1 c)$ = reduced viscosity (= viscosity number $\eta_{sp}/c$ ),
$[\eta]$	= $\lim_{c \rightarrow 0} \eta_{red,c \rightarrow 0}$ = limiting visc. number (= intrinsic viscosity)
$\Theta$	theta temperature
$\theta$	torsional angle (conformational angle in macromolecular science)
$\vartheta$	angle, especially scattering angle or torsional angle (organic chemistry)
$\kappa$	isothermal (cubic) compressibility
$\Lambda$	aspect ratio
$\lambda$	wavelength ( $\lambda_0$ = wavelength of incident light)
$\mu$	moment of a distribution
$\mu$	chemical potential
$\nu$	moment of a distribution, related to a reference value
$\nu$	kinetic chain length
$\nu$	frequency
$\nu$	effective amount concentration of network chains
$\nu$	velocity



$\Xi$	zip length
$\xi$	frictional coefficient
$\Pi$	osmotic pressure
$\pi$	mathematical constant pi
$\rho$	density (= mass/volume of the same matter)
$\sigma_n$	(number) standard deviation
$\sigma$	cooperativity
$\sigma$	wave number
$\varsigma$	degree of coupling of chains
$\tau$	bond angle, valence angle
$\tau$	relaxation time
$\tau$	light transmission; $\tau_{it}$ = internal transmission; $\tau_{et}$ = external transmission
$[\Phi]$	molar optical rotation
$\phi$	volume fraction; $\phi_f$ = free volume fraction
$\phi$	angle
$\chi$	Flory–Huggins interaction parameter
$\Omega$	angle
$\Omega$	thermodynamic probability
$\Omega$	skewness of a distribution
$\omega$	angular velocity