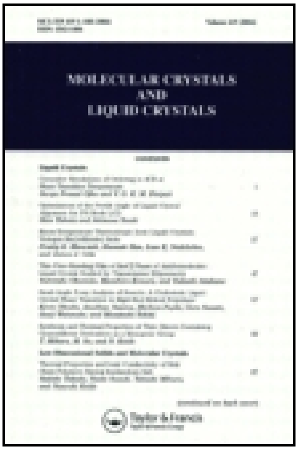

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Periodic Table of the Elements - the Basis for Forecasting Extreme Properties and for the Creation of New High-Performance Oriented Materials (Fibres, Whiskers). Theory, Practice and Prospects

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Periodic Table of the Elements – the Basis for Forecasting Extreme Properties and for the Creation of New High-Performance Oriented Materials (Fibres, Whiskers). Theory, Practice and Prospects

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Results of many years of theoretical and experimental studies of conditions to be met in order to obtain heavy-duty materials, particularly fibres and whiskers, were presented. Independent methods for the determination of theoretical and limited attainable strength, developed by the author, as well as two new indices (intermolecular and interatomic energy density) enabling to forecast mechanical properties, were discussed. Questions concerning maximal values of mechanical and thermal properties as well as the nature of elements of periodic table and their compounds having these properties were answered in the paper.

Keywords: periodic table of elements; extreme mechanical properties; extreme thermal properties; fibres; whiskers

INTRODUCTION

Great achievements have been made in the field of creating materials characterised by extreme mechanical and thermal properties, including fibres and whiskers. The problem of limitations of properties (theoretical and limited attainable strength, modulus and thermal stability) of these materials is of great importance both from scientific point of view and from that of practical discovering of real materials with maximal values of particular properties. Hence the following questions arise: "What are the limits of properties of materials? What kind of a material will have limited, the best properties among the elements and compounds imaginable on the basis of D. I. Mendeleev's periodic table of the elements? What are the optimal ways for further achievements in the area of high-performance materials?" With time passing these interesting, not much investigated, questions converted into an important theoretical problem of solid-state physics and mechanics of anisotropic polymeric materials such as fibres, films, whiskers, and in consequence for discovering real materials with maximal values of their properties.

More than 45-year theoretical and 30-year practical experience in this field helped me to answer these questions. A complex of scientific foundations and practical applications was created in these important areas. In this paper the current state of knowledge of limited mechanical and thermal properties of fibres, films and whiskers for different purposes was presented, including results of author's studies carried out for many years.

The author has developed different independent ways for the determination of theoretical and limited attainable strength and modulus by using structural mechanics methods. New characteristics of importance to practice - limited attainable properties - were suggested for defectless ideal crystalline materials (at definite temperature - time T - τ conditions) in contrary to the theoretical properties (at absolute zero temperature $T \to 0$ K by H. Mark and G. Bartenev). Two new indices - intermolecular and interatomic energy density - which are of importance to predicting mechanical properties were suggested, as well.

Chemical bond energy densities and extreme properties of many elements of Mendeleev's periodic table and their compounds were calculated and examined. The substances of linear and layered structures with maximal chemical bond energy density were determined. The same calculations were performed for all main linear oriented polymeric fibres. Many of the results are the first reported in the literature. The calculations of extreme properties were carried out by using two independent methods and verified by some experimental physical methods.

Comparative analysis of specific (related to mass or volume unit) interatomic interaction energies for different substances show that trivalent and tetravalent „light" elements of the second and third groups of the periodic table and their compounds - carbides, nitrides, oxides - have maximal energy density. These substances can be the materials with the highest values of properties. Oriented polymeric materials (fibres, whiskers) belong to this group, too.

The oriented polymeric materials with maximal theoretical and limited obtainable properties were determined. The linear carbon polymer, β-carbine, has the highest modulus and is the strongest material in nature. Not far from it are hexagonal structures - boron nitride, graphite and silicon carbide. The most thermally stable among polymeric materials is graphite and this fact was confirmed by calculations performed and relevant experimental results.

The theoretical and limited attainable elasticity modulus and strength as well as limited thermal stability properties of many conventional fibres and special high-performance fibres were calculated by different independent methods on the basis of ideal crystal model of linear (aliphatic and aromatic) and layer structures. The results were correlated with interatomic chemical bond energy density in axial direction. Many of the results were the first in scientific literature, for example those for β-carbine and para-aramides. Some of literature data were corrected and completed, too.

Aliphatic and aromatic compounds, including para-aramide carbocyclic and heterocyclic polymers and fibres were analysed; their theoretical and limited attainable mechanical properties in comparison with maximal real indices were determined. The same work was performed for carbon fibres. New indices were introduced, namely structural perfection coefficients which characterise integral perfection of production method. All these calculations permit to evaluate a possibility of improvement in properties of materials produced today and those of new high-performance materials.

Very interesting investigation, from the point of view of practice, was carried out in order to establish how to obtain fibres of maximal mechanical properties on the basis of rigid-chain and layer polymers. Results of the above investigation were compared with results of calculations performed in order to establish how to get polymeric fibres of maximal properties and discussed from the point of view of thermodynamics and kinetics. A difference exists between thermodynamic and kinetic requirements for achievement of structural maximum regularity and maximal properties approach. Therefore the ideal molecular structure has to be corrected for the control of kinetic flexibility in fibre-forming processes and fibre treatment. The main principles of controlling the structure and properties of high-performance fibres are discussed.

Nowadays, the oriented PBZ - structures (poly-p-phenylene-bis-benzothiazole and oxazole) are the most perspective as real fibres with extreme mechanical and thermal properties among linear polymers, and carbon graphite structure among layered polymers.

All calculations mentioned above and results of investigation of limited properties can (or have to) be used for the prediction of the most perspective oriented polymeric materials (especially fibres and whiskers) and for finding ways of achievement of maximal values of properties in practice.

ENERGY CHARACTERISTICS OF SOME ELEMENTS AND COMPOUNDS

Extreme mechanical and thermal properties correspond to solids with maximal interatomic interaction energy and fully ordered structure.

Energy density of homodynamic compounds can be calculated from equation:

$$U_B = \frac{\Sigma U}{V_M} = \frac{\Sigma U}{M_A} \cdot \rho_{CR} \qquad (1)$$

Energy density of chain or layer polymers can be calculated for polymer chains (layers) without side substituents from equation:

$$U_B = \frac{\Sigma U}{V_M} = \frac{\Sigma U}{M_E} \cdot \rho_{CR} \qquad (2)$$

U - interatomic bond energy per one g-atom or mole;

V_M - atomic or molar volume (element, compound, polymer elementary link);

M_A - atomic or molecular mass (element, compound);

M_E - molecular mass of polymer elementary link;

ρ_{CR} - material or crystal (crystallite) density (element, compound, polymer);

ρ'_{CR} - polymer crystal (crystallite) density for atoms of main chains (layers) without side substituents.

After analysis and calculations of energy characteristics for many elements and compounds we have found that maximal chemical bond energy density corresponds to the 2-nd and the 3-rd periods of periodic table of elements.

The data on the energy density of some elements of the 2-nd and the 3-rd periods are presented in Table 1. The same data concerning some oriented polymers (polymeric crystals and fibres) are presented in Table 2 as an example.

TABLE 1 Structural characteristics of some elements and compounds of the 2-nd and 3-rd periods of periodic table of elements in comparison with maximal axial elasticity modulus

Elements, compounds	Atomic, molecular mass	Density, g/cm^3	Atomisation energy, KJ/g-atom, kJ/mole	Atomisation energy density, kJ/cm^3	Elasticity modulus, GPa
B	10.60	2.310	562	120.0	400–420
C (diamond)	12.01	3.520	712	209.0	1080–1100
C (graphite)	12.01	2.270	717	135.0	960–1040
BN (cubic)	28.20	3.450	1320	161.5	810–970
SiC (cubic)	30.10	3.215	1251	133.5	480–560

TABLE 2 Energy characteristics of fibres and correlation of their mechanical properties

Materials	Energy density, kJ / cm^3		Elasticity modulus, GPa	
	Interatomic	Intermolecular	Axial	Transverse
Polyethylene				
- crystallite	17–22	0.3–0.37	230–250	3.2–4.3
- fibre	–	–	2.4–15	2.0–3.0
Poly-*para*-phenylene-terephthalamide				
- crystallite	52–54	0.9–1.0	220–230	–
- fibre	–	–	130–170	7–8
Graphite				
- single crystal *	130– 49	7–8	1000	48
- graphitised fibres **	–	–	500–700	6

Notes: * Needle crystals grown from gas phase
** Polyacrylonitrile-based high-modulus fibre

The energy data given above (Table 2) for linear macromolecule chain enable to explain the reason for the chemical constitution of the fibres - C, N, O atoms. Moreover, layer and three-dimensional structures of other materials with extreme properties consist of B, C, N, O, Al and Si atoms and compounds (borides, carbides, nitrides, oxides). Results of calculations and experiments, performed in order to determine extreme mechanical and thermal properties of fibres and whiskers, were presented in previous papers [1-10], including results of studies carried out by the author.

EXTREME VALUES OF MECHANICAL AND THERMAL CHARACTERISTICS

The extreme mechanical and thermal characteristics values concern fully ordered „ideal" structures without any defects. Theoretical and limited attainable mechanical and thermal properties are the limits for oriented structures, which can be achieved in principle. They are calculated in the main axial directions for defectless single crystals with extended chains or layers of very high molecular mass (for three-dimensional solids in the main axis of crystal structure) [1-10].

Theoretical elastic modulus and strength E_t ; σ^*_t are determined at 0 K, when all thermal motions are freezed, whereas limited attainable elastic modulus and strength E_{lim} ; σ_{lim}^* are determined at a definite temperature - time conditions. Frequently these conditions are T = 293 K and breaking time τ^* = 10 - 20 s, but other conditions may be used as well.

Theoretical elastic modulus of polymeric crystal may be calculated by different methods:

1. On the basis of molecular chain axial (or layer) collective deformation

$$E_t = N\sum\lambda / \sum(\Delta\lambda / f_b) \qquad (3)$$

E_t - theoretical elasticity modulus;
N - number of molecular chains;
λ - interatomic bond length projection to chain axis;
f_b - bond deformation force in chain axis direction.

2. On the basis of theoretical elastic impulse velocity along molecular chains. This velocity is determined on the basis of bond vibration frequency:

$$E_t = v^2\rho' ; \qquad v = 4cv\lambda \qquad (4)$$

v - bond valence wavenumber;
ρ' - calculated density of chain „skeleton" (without side substituents);
c - electromagnetic wave velocity;
ν - bond vibration wavelength.

3. On the basis of the dependence of elastic modulus on thermal expansion coefficient in chain or layer direction :

$$E_t = 0.5(Nk\,\lambda\alpha_c) \qquad (5)$$

k - Boltzmann constant;
α_c - chain axial thermal expansion coefficient.

Limited attainable modulus values E_{lim} can be determined experimentally by X-ray method on the ground of variation in interatomic distances in crystallites at axial loading of highly-oriented fibres or films. Theoretical and limited attainable values of the modulus are in a good agreement: $E_t \approx E_{lim}$, because the modulus changes only slightly with temperature due to the thermal expansion of chemical bond length. Theoretical and limited attainable modulus values are presented in Table 3.

TABLE 3 Theoretical and limited attainable mechanical properties for polymeric crystals*

Polymers (fibres)	$E_{TH} \approx E_{LIM}$; GPa	σ^*_{TH} ;GPa	σ^*_{LIM}; GPa	σ^*_{EXT}; GPa**
Polyethylene	230–250 (240)	26–27(26)	21–23.5 (22)	7–10
Polyvinyl alcohol	200–255 (230)	22–23 (22)	17–19 (18)	4.5–8
Polyethylene terephthalate	140–150 (145)	20.5–22 (21)	14–17 (16)	4–6
Cellulose	120–180 (160)	14–18 (16)	10–12 (11)	3.3–6.5
Poly-p-phenylene terephthalamide	230–240 (235)	29–31 (30)	23–25 (24)	9–12
Polyamidebenzimidazole	220–240 (230)	21–25 (24)	16–20 (18)	9–11
Graphite	960–1040 (1000)	120–140 (130)	100–107 (105)	-

Notes: * Values for K_E and K_σ calculations are given in parentheses.
** σ^*_{EXT} - Extrapolation of technical data to 100% crystallinity and disorientation angle of 0^0.

Theoretical strength of polymeric crystal can be calculated by different methods.

1. By using stress values at collective deformation of polymeric chains (layers) to their rupture:

$$\sigma_t^* = N\left(f_b^* \cdot \cos\psi\right) \tag{6}$$

f^*_b - bond rupture force;

ψ - angle between bond and chain axis direction (temperature - time dependence).

2. On the ground of temperature for strength dependence, extrapolated to absolute zero temperature:

$$\tau^* = \tau_0 \exp\left[\left(U_0 - \gamma\sigma\right)/kT\right] \tag{7}$$

$$\sigma_t = U_0\left(T = 0\right)/\gamma_t \tag{8}$$

τ^* - life time;

τ_0 - constant, $\tau_0 \approx (10^{-13} - 10^{-13})$ c.;

U_0 - activation energy of mechanical dissociation;

γ - structural coefficient;

γ_t - theoretical structural coefficient;

T - absolute temperature.

Theoretical strength can be determined also by the extrapolation of limited attainable strength to absolute zero temperature (see below). Another approximate evaluation can be performed by using the Orovan equation:

$$\sigma_t \approx \xi E_t \tag{9}$$

ξ - coefficient (Orovan equation), $\xi \approx 0.1$

The correlation between results obtained by these methods is good. The theoretical strength values are presented in Table 3.

Limited attainable strength can be determined by two methods:

1. By temperature - time dependence starting from theoretical strength values:

$$\sigma_{lim} = \sigma_t - \left[\left(kT/\gamma_t\right)\ln\left(\tau^*/\tau_0\right)\right] \tag{10}$$

σ^*_{lim} - limited attainable strength.

2. By maximal breaking stresses for chemical bonds which can be determined by using spectroscopic methods (valency vibrations changed by loading).

All results of limited strength calculations are presented in Table 3.

The values of technical extrapolated strength are of importance to practical examination of fibre strength reserve. These values are determined by the extrapolation of experimental results to 100% crystallinity and full orientation (see Table 3). They are essentially lower (by about 1/3 - 1/2) than limited attainable strength values because of the presence of different structural defects. If the dimensional effect of breaking characteristics is taken into consideration, the indices of technical extrapolated strength will have higher values and differences with limited attainable properties will be smaller.

Maximal thermal resistance of polymeric and other materials corresponds to thermal radical dissociation of chemical bonds. The values of limited thermal resistance indices T* can be calculated for polymeric crystals from the strength - temperature - time dependence and are considered as extrapolated temperature of zero-point strength $\sigma = 0$ at definite time to break τ^*:

$$T^* = U_0 / k\ln\left(\tau^* / \tau_0\right) \qquad (11)$$

T^* - limited attainable thermal stability.

The calculated indices are presented in Table 4 as a dependence of interatomic bond energy on time.

TABLE 4 Limited thermal stability for polymeric crystals (calculated values).

Time	U_0, kJ/mol					
	100	200	400	600	800	1000
	Limited thermal stability, K					
10 s	370	740	1480	2230	2970	3710
1 h (3600 s)	310	620	1340	1860	2480	3100
100 h (3.6×10^5 s)	280	560	1120	1680	2240	2800
10000 h (3.6×10^7 s)	250	500	1010	1510	2020	2620

Now the answer can be given to principally important questions: "What are the maximal values of mechanical and thermal properties? What kind of elements or compounds can have these properties?" The necessary conditions for the achievement of extreme properties are the highest dissociation energy and homogeneity of all chemical bonds which correspond to maximal energy density of chemical bonds in the solid state. These requirements are met for the elements (and their compounds) of the 2-nd and 3-rd periods mentioned above, especially for carbon polymers (graphite, carbine), hexagonal silicon carbide and boron nitride. The data concerning these substances are summarised in Table 5 [1, 11, 12].

TABLE 5 Theoretical and limited attainable mechanical properties of hexagonal layered structures of BN, SiC, graphite and β-carbine

Polymeric structures	$E_{TH} \approx E_{lIM}$; GPa	σ^*_{TH}; GPa	σ^*_{LIM}; GPa
Boron nitride (hexagonal)	105	120–135	95–105
Silicon carbide (hexagonal)	110	100–120	65–90
Graphite	960–1040 (1000)	120–140 (130)	100–107 (105)
β-carbine	180	22–23	19–20

Note: *Average values for K_E and K_σ calculations of graphite are shown in parentheses.

Linear carbon polymer β-carbine has the maximal interatomic energy density and it is a „record-holder" among the elements of periodic table as concerns the extreme mechanical properties. Calculated value for theoretical (and limited attainable) modulus is about *1800* GPa; *theoretical strength*: about 230 GPa, limited attainable strength (at 293 K and 10 s): 200 GPa [11, 12]. Graphite is thermodynamically stable allotrope form of carbon with the highest thermal stability among the elements of periodic system. Calculated and experimentally determined temperatures of thermal dissociation are in a good agreement (for graphite: 3900 - 4100 K). It is worth to add that this value corresponds to radical dissociation in anti-oxidative media. Thus our calculations and experiments have led to the answer: „There are no elements or compounds with mechanical and thermal indices higher than those of carbon".

Maximal experimental values for mechanical properties of fibres and whiskers are extensively investigated. Some results of these studies are summarised in Table 6 [1-9, 13-16].

TABLE 6 Maximal mechanical properties (determined experimentally) for fibres and whiskers*

Polymers (fibres)	E; GPa	σ^*; GPa	K_E	K_σ
Polyethylene	70–160(160)	3–7 (3.5)	0.67	0.160
Polyvinyl alcohol	36–70 (70)	1.2–2.8 (2.5)	0.30	0.140
Polyethylene terephthalate	20–25 (25)	0.8–1.4 (1.4)	0.17	0.090
Cellulose	15–80 (70)	0.85–1.5 (1.2)	0.44	0.110
Poly-p-phenylene terephthalamide	120–170 (170)	3.3–3.8 (3.6)	0.72	0.150
Polyamidebenzimidazole	120–160 (150)	4.2–6 (5)	0.25	0.270
Graphite	500–830 (830)	5–7.2 (7.2)	0.83	0.069
	980–1000** (1000)	21–25** (24.5)	1.00	0.230

Notes: * The 1-st number corresponds to commercial samples; the 2-nd number corresponds to experimental samples. Average values for K_E and K_σ calculations are shown in parentheses.

** Values for whiskers.

Properties of fibres and whiskers achieved in reality were discussed in comparison with limited values [1-9, 13, 14, 16]. This comparison permits to calculate the relative achievement of elastic modulus and strength in real fibres and whiskers:

$$K_e = E / E_{lim} \quad \text{at} \quad E_{lim} \approx E_t \tag{12}$$

$$K_\sigma = \sigma^* / \sigma_{lim} \tag{13}$$

K_E - modulus achievement coefficient;

K_σ - strength achievement coefficient;

σ^* - fibre strength.

The achievement coefficients K_E and K_σ are given in Table 4. Main reasons for the difference and achievement rate are structural irregularity and defectiveness of fibres on all structural levels: molecular, supramolecular and macrolevel (fibre level). Whiskers with single crystal structure are more regular than fibres. Therefore their mechanical properties are higher and achievement coefficients are increased in comparison with fibres and films. It is worth to add that mechanical destruction is a result of mechanochemical reactions. Strength of real polymeric materials – fibres and whiskers - is influenced by impurities, especially by trace amounts of oxidising and hydrolysing compounds.

Real thermal properties of all oriented polymer materials are lower than limited values. This difference is a result of a variety of reactions proceeding in organic polymers during thermal treatment: polymer-analogous transformations, oxidation, hydrolysis, side groups detachment, etc. These reactions can be catalysed by small amounts of impurities. Thermal stability of polymers without side substituents (especially inorganic polymers) can approach the calculated level in anti-oxidative media only.

Achievement of maximal experimental values for mechanical properties of fibres occurs in two main ways: for flexible-chain polymers by maximal orientation at stretching; for rigid-chain polymers by self-ordering effect. Very interesting practical investigations to achieve of maximal mechanical properties of fibres on the basis of rigid-chain polymers have been done. There is a difference between thermodynamic and kinetic requirements for the achievement of maximal structural regularity and maximal properties approach. Therefore the ideal molecular structure is necessary to correct for regulation of kinetic flexibility in fibre-forming processes and fibre processing [17-19]. The problem of forming fibres with maximal mechanical and thermal properties is a subject of another study.

In memory of Professors Mark and Meos

Professor Herman Mark (Brooklyn Polytechnical Institute) has given me the first impulse for this „hobby work" in the period of my university studies and

later. His attention and discussions with him and with Professor Alexander A. Meos, my teacher in the period of studies at St-Petersburg State University of Technology and Design (formerly Leningrad Textile Institute) were of great help to me. Therefore this paper is dedicated to the memory of Professors H. Mark and A. I. Meos.

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