ENGINEERING TRANSACTIONS • *Engng. Trans.* • **49**, 2–3, 315–358, 2001
Polish Academy of Sciences • Institute of Fundamental Technological Research
10.24423/engtrans.562.2001

LIQUID CRYSTALLINE PROPERTIES OF SYNOVIAL FLUID

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The main aim of this paper is to discuss the liquid crystalline concept of synovial fluid. This new concept was proposed by KUPCHINOV [30] and was next developed in [31, 32, 58]. According to these papers, the crystalline structure of synovia ensures very low friction coefficient in human (and animal) joints. Experimental data in favour of crystalline structure of synovia have been discussed. Mathematical model of such fluid, based on ERINGEN'S theory [14, 15], has been advanced.

1. INTRODUCTION

The mysteries of lubrication mechanism in animal and human synovial joints fascinated scientists and engineers since centuries and stimulated the research in this field. Biotribological phenomena occuring in synovial joints are still not well understood. Synovial joints are very effective natural bearings of the body providing motions with a very small friction coefficient. They can perform their work up to nine decades under a wide range of loading conditions with very little wear and tear of the articulating cartilage layers. The coefficient of friction in healthy synovial joints is very small and can reach even the value equal to 0.001. We recall that compressive stresses in the human hip and knee joints range from several MPa to about 18 MPa [39]. Typical human joint, like hip and knee joints, experience about one million cycles per year. The lubrication minimizes the wear of synovial joints. It is generally believed that these excellent kinematic and load bearing characteristics are due to material structure of the synovial joint, i.e. biophysical, biochemical and biorheological properties of synovial fluid and anatomy of the articulating surfaces of the joint. The properties of articular cartilage are fundamentally different from the properties of materials used in a typical engineering bearing. When synovial joint degenerates or becomes damaged, its characteristics drastically deteriorate. It should be cured and often total joint replacement is needed. Thus a clear understanding of synovial joints would aid to improve the existing mechanical bearings and the replacement of human joints. The main difficulty in the study of synovial joints is that in vivo experimental measurements are limited. Thus the researchers look for a theory, which would provide reasonable quantitative and qualitative predictions. Over the past fifty years, many investigators have intensively studied the friction, lubrication and wear processes in synovial joints [3, 4, 12, 17, 18, 24, 31, 39, 41, 50, 58]. However, our understanding of the mechanisms governing the lubrication of joints and its modelling are far from being complete. Many modes of lubrication have been proposed (hydrodynamic, hydrostatic, elasto-hydrodynamic, boundary, weeping, mixed, boosted), though some of the theories proposed to explain experimental data are sometimes limited to a particular loading configuration. The substantial reviews can be found in the papers [31, 39, 40].

In essence, theoretical studies on joint lubrication can be divided into two groups. More standard is the approach used in tribology. The synovial fluid is then modelled as a Newtonian or non-Newtonian fluid [27, 29, 41, 43, 53]. The second approach is based on modelling the cartilage behaviour as a multiphase porous material, cf. [33, 39, 40]. Recently GU *et al.* [23] developed a mixture theory to model the mechano-electrochemical behaviour of charged-hydrated soft tissues containing electrolytes. Such a theory applies also to cartilage, cf. also [25]. TELEGA and WOJNAR [59] proposed a macroscopic model of cartilage which was derived by using homogenization methods, cf. also [60]. Such an approach enables to take into account the microstructure of cartilage. However, the basic problem is how to model this complicated microstructure. A simple hexagonal model permits to describe the anisotropy of cartilage and change of material coefficients along the thickness.

New experimental data obtained in the past decade threw new light on the microscopic structure of biobearings. It seems that in the light of recent investigations, the concept of synovial fluid exhibiting a liquid crystalline structure and properties is very promising. This idea is due to KUPCHINOV [30], cf. also [31, 58]. The model of liquid crystalline structures has also been advanced with regard to articular cartilage and other connective tissues, cf. [22, 31, 46]. This new concept offers a deeper insight into understanding of biofluids, electrical processes in synovial joints and, first of all, can probably explain exceptionally low friction coefficient in such joints. It seems that liquid crystals play a fundamental role in many anisotropic processes in biology.

Liquid crystals exhibit specific properties, cf. [1, 7, 11, 32, 37, 57]. For instance, their optical properties are typical for the crystalline state and their mechanical properties are typical for liquid state (fluidity and surface tension). Of special importance is that they are "smart" materials, that means – they possess memory. Moreover, liquid crystal materials are very sensitive to weak external fields. They are sensitive to mechanical stresses, electrical and magnetic fields, confining surfaces, etc.

Surprisingly, this new concept of synovia has not yet been formalized, i.e. no mathematical model of synovial fluid with crystalline structure seems to be available. In our opinion, a micropolar fluid theory can be applied to model the behaviour of such a fluid.

The aim of this paper is to describe the crystalline structure of synovial fluid and experimental data in favour of such a concept. Moreover, we propose a mathematical model of liquid crystals and discuss a micropolar fluid model and the results obtained.

The paper is organised as follows. In Sec. 2, a brief review of the structure of liquid crystals and their properties are described with particular reference to data in synovial joints. Synovial joints and their structure are introduced in Sec.3. Experimental data are presented in Sec.4. Section 5 is concerned with a mathematical model of liquid crystals. Next, in Sec. 6 we discuss the micropolar fluid model of synovial fluid in the context of an approximation of liquid crystal model. We also discuss the restrictions enabling this passage. Main conclusions and future perspectives are outlined in Sec. 7.

2. SOME ASPECTS OF LIQUID CRYSTALLINE STRUCTURES

In this section we briefly review liquid crystalline structure and properties of synovial joints. We observe that liquid crystalline structures are found in numerous tissues and organs of living organisms, like grey nerve cells (brain), axons of nerve cells, DNA chains, liver, spleen, bile, hyaline body of eye, spermatozoon, biological membranes, etc.

2.1. Intermediate states

The solid crystalline state is characterised by a fixed 3-dimensional arrangement of molecules. The solid crystalline phase is a result of long-range orientational as well as positional ordering of molecules. Solid crystals are usually anisotropic.

In the conventional liquid phases, the molecules can translate and rotate. There is no long range ordering. Usually, liquids possess isotropic properties.

Some intermediate states can exist between the solid phase and the isotropic phase, often called soft matter states. In those states one kind of movements is permitted while another is forbidden. Two main intermediate states are known: a plastic state and a liquid crystalline state. For instance, in liquid crystalline state (mesomorphic state) the molecules can translate and, additionally, rotate especially around long axes. Consequently, long-range orientational ordering exists [61]. The asymmetry (called anisosymmetry by SONIN [54]) of forms of structural units (elements, molecules) and their rigidity are responsible for the liquid crystallinity of the state.

2.2. Liquid crystalline states

The intermolecular forces are responsible for formation of the mesomorphic phase. From the thermodynamic point of view, liquid crystals can be divided into two main classes. The first class comprises thermotropic liquid crystals resulting from the melting of some solids; the mesomorphic phase arises as an intermediate phase between ordinary crystal and ordinary (isotropic) liquid phases. To the second class belong lyotropic liquid crystals in which the mesomorphic phase arises under the action of a solvent on the substance having amphiphilic character of solubility (polar and nonpolar simultaneously). The liquid crystal phases in such multi-component systems exist in certain regions depending upon the concentration and the temperature. In some cases the structural units may be represented by single molecules, in other by molecular aggregates like micelles, columns, oligomers, systems like viruses, lamellae, tubular or spherical complexes [1, 2, 8]. The lyotropic liquid crystals are typically colloidal. They are found in living organisms, where water is a polar solvent.

According to the geometrical shape of the molecules, liquid crystals may be divided into phases with rod-like molecules, phases with brick-like molecules and phases with disc-like (discotic) molecules.

The liquid crystal structures are divided into two main groups: N – nematic phase with parallel ordering of the long molecule axes, and Sm – smectic phases with additional layered structure. When formed by optically active molecules, the liquid crystals phases develop chiral modifications, namely chiral nematics and chiral smectics. Chiral nematic N* and chiral modifications of smectics Sm* form long-range helicoidal structures. The ester derivatives of cholesterol and other sterols form similar structures, namely helical arrangements of sheets of molecules (Ch – cholesteric liquid crystals).

In the nematic phase the molecules are free to translate and rotate, especially around the long axes. Because of additional layered ordering exibited by smectic phases, besides the parallel arrangement of long molecular axes, translations in these phases are more or less forbidden. These phases are highly differentiated with regard to the type of ordering inside the layers (e.g. parallel or anti-parallel positions of molecules) and the variety of tilted arrangement of the molecules in the layers (different slopes of long axes of molecules in a layer).

In essence, the separate classification of chiral materials or cholesterics is due to unusual optical properties of the structure. These materials are optically extremely active since they rotate the plane of linearly polarised light. They have a naturally twisted structure. A chiral nematic can be depicted as the stack of nematic pseudo-layers. Within each plane layer, the parallel alignment of the molecules is like that of a nematic liquid crystal. Each layer, however, is slightly twisted with respect to the next layer. It means that the preferred direction of long molecular axes, being an average direction defined by a unit vector or the so-called director, must be shifted by a constant angle when passing from one layer to the adjacent layer. On a large scale the director describes a helix in the physical space. The degree of twist is characterised by the pitch of the helix. The pitch itself is defined as the longitudinal distance in which the director performs one complete rotation. The helical pitch can vary from that of pure compound to infinity (it means that the cholesteric becomes a nematic). The resultant helicoidal arrangements are labelled as left or right-handed. They depend on the chemical structure of the molecules, especially on their functional side groups.

In the case of chiral smectics the helicoidal arrangement is also possible; then the long molecular axes can be considered as being placed on some conical surface in which they are shifted by a constant angle when passing from one layer to the next one. Consequently, the twisted, tilted smectics also behave optically as cholesterics [2].

In discotic liquid crystalline phases, the molecules with flexible constituents are attached symmetrically to the central flat aromatic core. Two main types of discotic liquid crystals are the discotic nematic phase and columnar phase. In the discotic nematic phase the molecules lie parallel to each other, without any additional ordering. For optically active discotic molecules the chiral discotic nematic phase arises with the helicoidal arrangement. In columnar phases the discotic molecules are stacked one upon another in the form of columns [2].

Some mesomorphic materials are polymesomorphic. It means that they possess more than one liquid crystalline phase. Moreover, an exactly specified sequence of phase transitions exists. The thermal sequence of phase transitions with increasing/decreasing temperature is as follows:

solid state ⇔ liquid crystal (mesophase) ⇔ isotropic liquid.

The following sequence of phase transitions exists in the range of mesophase:

Sm ⇔ N(Ch).

We should take into account all modifications of smectic thermotropic materials: A, B, L, E, I, J, K, F, G, H as well as chiral modifications of smectics Sm*: C*, I*, J*, K*, F*, G*, H* and chiral modification of nematic – N*.

Generally, the liquid crystals are characterised by the facility to order-order transitions. It means that intermesophase transitions are preferable. Sol-gel, liquid crystal-gel, liquid crystal-liquid crystalline gel, liquid crystal-plastic state, liquid crystal-liquid crystalline plastic state transitions are also possible [1, 7, 8, 11].

2.3. Liquid crystal polymers

Liquid crystal polymers can be divided into two main classes: main-chain and side-chain (comb-like) polymers. It means that polymeric liquid crystals appear when mesogenic groups are included as parts of main chain or are attached to polymer chains as their side-groups [2], see Fig. 1. It is possible that a macromolecule is composed of flexible, semiflexible and sometimes nonmesogenic "spacer" groups. In the liquid crystal states, the rod-like or the discotic mesogenic groups assume parallel configurations to form the nematic or smectic phases.

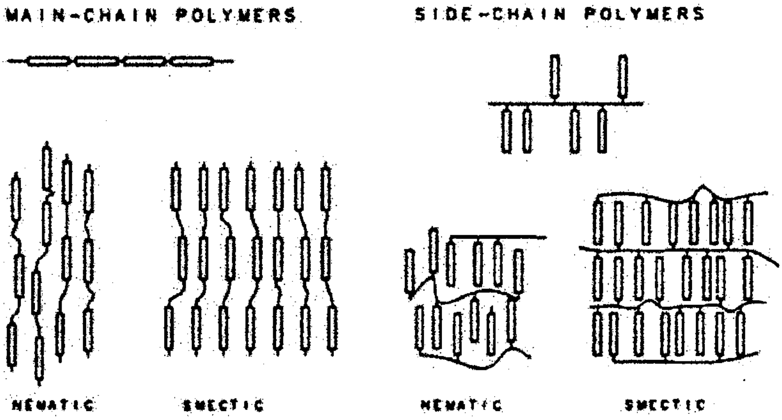


FIG. 1. Schematic diagram of liquid crystal polymers, after ADAMCZYK [2].

However, the main polymer chain remains unoriented. Sometimes, because of unusually high levels of orientation, the macromolecular liquid crystal forms fibers.

2.4. Some physical properties of mesomorphic materials

Various theories have been proposed to model the behaviour of liquid crystals. The first of them is the well-known director theory. This theory was initiated by OSEEN [44] and developed by FRANK [20]. It is based on a fundamental concept that material points of liquid crystals are oriented points, and they are like rigid rods which possess orientational degrees of freedom. Later, with the works of ERICKSEN and LESLIE [13, 34], these early developments were recast into the language of contemporary continuum mechanics. New continuum theories appeared for a different type of liquid crystals incorporating additional properties of nematics, smectics, cholesterics, ferroelectrics, and nonchiral or chiral liquid crystals.

A unified continuum theory for all class of liquid crystals was proposed by ERINGEN [15]. This micropolar continuum theory concerns liquid crystals whose molecular elements possess the following degrees of freedom: translations, rotations, expansions and contractions. Nonlinear constitutive equations unify different classes of liquid crystals. For instance, chiral and nonchiral liquid crystals are singled out by means of symmetry groups, smectics are singled out by constraints. Also thermodynamic restrictions are imposed.

The domain of applicability, fundamental nature of basic laws and potential applications of the unified continuum theory of liquid crystals are discussed in Sec. 5. However, it is not clear how to determine the material coefficients involved in this theory.

Thin layer problems of liquid crystalline materials are considered in many technical applications and in modelling of organic materials. Consequently, both the bulk and surface (interface) properties of liquid crystals will be discussed below.

2.4.1. Bulk properties

Viscosity

According to LESLIE [34], an anisotropic liquid crystalline material with a nematic structure is described by the viscous stress tensor $\boldsymbol{\Psi}$ of the following form:

$$(2.1)\qquad \Psi_{ij} = \alpha_1\chi_k\chi_p A_{kp}\chi_i\chi_j + \alpha_2\chi_i N_j + \alpha_4 A_{ij} + \alpha_5\chi_i\chi_k A_{kj} + \alpha_6\chi_j\chi_k A_{ki} + \alpha_3\chi_j N_i,$$

where $\chi(\mathbf{r},t) = (\chi_i(\mathbf{r},t))$ denotes the director, and N_i – are components of the director velocity with respect to motionless fluid.

We have

$$\mathbf{N} = \frac{d\chi}{dt} - \omega \times \chi, \tag{2.2}$$

where the angular velocity ω is given by

$$\omega = \frac{1}{2}\operatorname{rot}\mathbf{v}. \tag{2.3}$$

After LESLIE [7, 34] we write

$$A_{ij} = \frac{1}{2}\left(\frac{\partial v_j}{\partial x_i} + \frac{\partial v_i}{\partial x_j}\right), \tag{2.4}$$

where $\mathbf{v} = (v_i)$ denotes the velocity vector.

The coefficients α_n, $n = 1, 2, ..., 6$, are rheological coefficients called Leslie's coefficients. Only five of Leslie's coefficients are independent. The coefficient α_4 corresponds to the viscosity in an isotropic fluid; α_2 and α_3 describe orientational effects whilst α_5 and α_6 are due to anisotropy of the liquid crystal.

Already in the thirties, MIĘSOWICZ [1, 37] performed experiments with liquid crystals well-oriented by a magnetic field. In essence, the experiments can be described as follows. A layer of liquid crystal was placed between two plates, one of which was fixed. The second plate moved slowly in order not to change the orientation of molecules. Three typical "geometries" were realised depending on the orientation of molecules, thus the director orientation, the direction of flow velocity and the velocity gradient, see Fig. 2. MIĘSOWICZ [37] determined three viscosity coefficients η_1, η_2 and η_3 characterising the flow of nematics. The

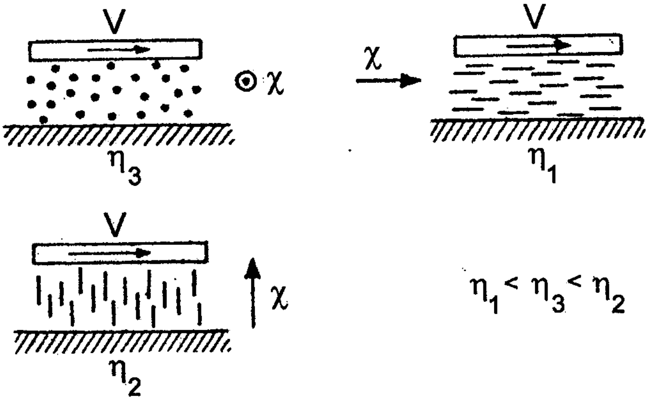


FIG. 2. Scheme of experiment for viscosity coefficients measurements in a nematic liquid crystal, after MIĘSOWICZ [37] and ADAMCZYK [1].

viscosity coefficients are measured when molecules are oriented along the moving plate, transversely and perpendicularly, respectively. We have the following inequalities: $\eta_1 > \eta_3 > \eta_2$. Anisotropic viscosity is a property specific only for mesomorphic materials. Usually the viscosity anisotropy is given by the ratio η_1/η_2, which exceeds 4.

The Mięsowicz coefficients η_1, η_2 and η_3 can also be determined by studying a flow of liquid crystal in a rectangular tube. In Fig. 3 three MIĘSOWICZ coefficients are represented for a nematic liquid crystal being from a homologue of cyanobiphenyl. We note three coefficients for the mesophase; obviously, the isotropic phase is characterised by one coefficient.

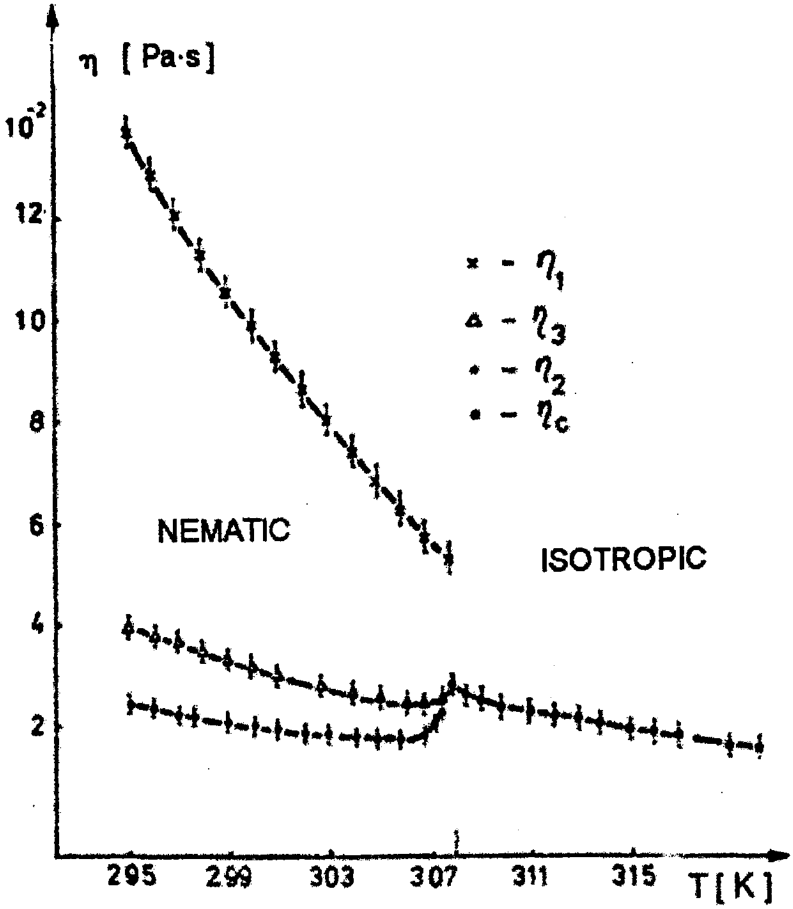


FIG. 3. Temperature dependence of the Mięsowicz viscosity coefficients for a nematic liquid crystal pentyl-cyano-biphenyl in nematic phase (η_1, η_2, η_3) and isotropic phase (η_c), after SZWAJCZAK [56].

We observe that nematics have relatively low viscosity. The viscosities of cholesterics and smectics are usually higher. In general, the viscosity, including shear viscosity, of liquid crystal materials is much lower than that of conventional materials at a comparable molecular weight.

The Leslie and Mięsowicz coefficients are interrelated by:

$$\begin{aligned} \eta_1 &= \frac{1}{2}\alpha_4, \\ \eta_2 &= \frac{1}{2}\left(\alpha_3+\alpha_4+\alpha_6\right), \\ \eta_3 &= \frac{1}{2}\left(-\alpha_2+\alpha_4+\alpha_5\right). \end{aligned} \tag{2.5}$$

Deformability

The energy related to deformation of liquid crystals is small. Consequently, their molecular structure can easily be changed by external fields, e.g., by mechanical stresses, temperature, electric field and so on. Hence one can easily control their properties, particularly in the case of cholesteric liquid crystals. The energy related to parallel ordering of molecules in nematic liquid crystals is of the order of a fraction of J/mol. The energy stored up in cholesteric helix amounts to about 10^{-5} of the energy of nematic ordering.

According to the continuum theory, liquid crystals are treated as liquids. It means that elasticity is almost negligible. However, additional kind of deformations is introduced, namely such deformations which do not change the distance between the points of the body considered. Those deformations change orientation of the field of director χ. The director is parallel to the privileged direction of molecular ordering. Additional assumptions concerning the nematic phase are formulated as follows:

(i) the ordering of the molecules in the bulk of sample assumes the same direction;

(ii) discontinuities of ordering of molecules occur only on lines of disinclination; the direction of ordering changes in a continuous manner;

(iii) the distribution and orientation of molecules of liquid crystal on confining surfaces remains unchanged.

Let the director χ be directed along the z-axis. Then the ordering of molecules can be characterised by the six components of $\boldsymbol{\nabla}\cdot\chi$ [1, 7, 20]:

$$
\begin{aligned}
&s_1 = \frac{\partial \chi_x}{\partial x}, &\qquad& s_2 = \frac{\partial \chi_y}{\partial y},\\
(2.6)\qquad &t_1 = \frac{\partial \chi_y}{\partial x}, && t_2 = -\frac{\partial \chi_x}{\partial y},\\
&b_1 = \frac{\partial \chi_x}{\partial z}, && b_2 = \frac{\partial \chi_y}{\partial z}.
\end{aligned}
$$

The density of the free energy for all types of mesomorphic phases is given by [1, 20]:

$$
\begin{aligned}
(2.7)\qquad g = \frac{1}{2}K_{11}(s_1 + s_2 - s_o)^2 + \frac{1}{2}K_{22}(t_1 + t_2 - t_o)^2 + \frac{1}{2}K_{33}(b_1 + b_2)^2\\
+ K_{12}(s_1 - s_2)(t_1 + t_2)^2 - (K_{22} + K_{21})(s_1 s_2 + t_1 t_2).
\end{aligned}
$$

where: s_1, s_2 – the splay or transverse deformations, t_1, t_2 – the twist deformations, b_1, b_2 – the bend deformations, s_o, t_o – the spontaneous deformations.

The last deformations take into account the fact that in the state corresponding to the minimum value of the free energy, the homogeneous deformation does not disappear; for instance, it may be due to the spontaneous twist of cholesteric liquid crystals.

In nematics three basic types of deformations are possible: splay, twist and bend [1, 7, 20], see Fig. 4. They are characterised by three elastic constants: K_{11}, K_{22}, K_{33}, respectively, cf. [1] and the references therein.

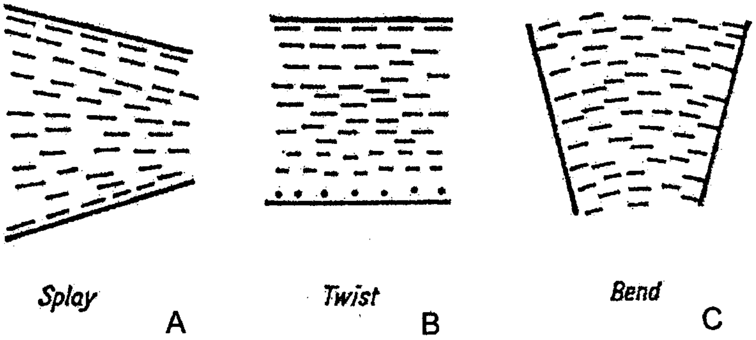


FIG. 4. Basic distortions of liquid crystalline nematic structures: (A) – splay, (B) – twist, (C) – bend, after ADAMCZYK [1].

Let g_s, g_t, and g_b denote the free energy density for the splay, twist and bend deformation, respectively. We have [20]

$$
\begin{aligned}
g_s &= \frac{1}{2}K_{11}\,(\operatorname{div}\chi)^2 ,\\
g_t &= \frac{1}{2}K_{22}\,(\chi \cdot \operatorname{rot}\chi) ,\\
g_b &= \frac{1}{2}K_{33}\,(\chi \times \operatorname{rot}\chi) ,
\end{aligned}
\tag{2.8}
$$

For nematics we have: $s_o = t_o = K_{12} = 0$. Moreover, K_{21} is also negligible. This is possible provided that spontaneous polarisation is absent and physical states corresponding to χ and $-\chi$ are identical.

The alignment of the bulk liquid crystal is determined by minimizing the sum of the elastic energies. This leads to the differential equation

$$\frac{dF}{dz} = 0,$$

where

$$F = \int\limits_V g dV,$$

whilst V denotes the volume of the sample.

The viscosity coefficients η_s, η_t, η_b corresponding to splay, twist, and bend deformations respectively, can be expressed in terms of Leslie and Mięsowicz coefficients as follows [1]:

$$
\begin{aligned}
\eta_s &= \alpha_3 - \alpha_2 - \frac{\alpha_3^2}{\eta_1},\\
\eta_t &= \alpha_3 - \alpha_2,\\
\eta_b &= \alpha_3 - \alpha_2 - \frac{\alpha_2^2}{\eta_2}.
\end{aligned}
\tag{2.9}
$$

Equivalently, if only the Leslie coefficients are used, then:

$$
\begin{aligned}
\eta_s &= \alpha_3 - \alpha_2 - 2\frac{\alpha_3^2}{\alpha_3 + \alpha_4 + \alpha_6},\\
\eta_t &= \alpha_3 - \alpha_2,\\
\eta_b &= \alpha_3 - \alpha_2 - 2\frac{\alpha_2^2}{\alpha_4 + \alpha_5 - \alpha_2}.
\end{aligned}
\tag{2.10}
$$

Orientation by flow

Orientation of molecules of liquid crystalline substances is also influenced by the flow processes. The appearance of optical anisotropy under flow can be caused by the orientation of anisometrical particles with their long axes in the direction of flow. It can take place in some organic substances and is also possible in non-organic substances, e.g. in sols of vanadium pentoxide, V_2O_5, cf. [54]. It was shown that in liquid crystals and in liquid crystalline polymers, the orientational effects are caused mainly by orientational flows. The effects of various flow fields, including shear and radial flow, on the orientation of liquid crystalline polymer molecules were discussed in [54].

2.4.2. Influence of confining surfaces

Liquid crystals are substances with properties which can be modified by very weak surface influences. Main reasons of confined surface/layer of material interactions are: the arrangement (orientation) due to curvature of the solid surface, electrostatic influence of double electric layer, immediate adhesion and intermediate adhesion. The direct adhesion consists in creation of van der Vaals bonds or hydrogen bonds between molecules of the liquid crystals and molecules belonging to the solid confining surface. The indirect adhesion consists in creation of bonds between the molecules of liquid crystal and molecules of substance constituting mono- or multi-molecular layer separating the layer of liquid crystal from the surface of solid body. The ordering influence of confining surfaces is a result of physico-chemical interactions of surface molecules with molecules of liquid crystal.

The surface roughness, for instance created by grooves, also plays a significant role. According to ADAMCZYK [1], the arrangement of nematic molecules parallel to the bearing surfaces and to the direction of grooves on these surfaces is advantegeous from the energetic point of view. Indeed, if molecules are oriented perpendicularly to a sinusoidal surface (see Fig. 5), the free energy density resulting from splay and bend deformation is given by

$$\begin{aligned} g &= \frac{1}{2}\left\{K_{11}\left[\left(\frac{\partial\varphi}{\partial y}\right)^2 + \left(\frac{\partial\varphi}{\partial z}\right)^2\right]\right\}, \\ \varphi &= Aq\cos(qy)e^{-qz}. \end{aligned} \tag{2.11}$$

Here φ denotes the angle between the axis z and the long axis of molecule (in the vicinity of the surface). Moreover, A denotes the amplitude of the perturbed surface and $2\pi/q$ is the wavelength of perturbation. Assuming that $K_{11} = K_{33}$, we get

(2.12) $$q = \frac{1}{2} K_{11} A^2 q^4 e^{-2qz}.$$

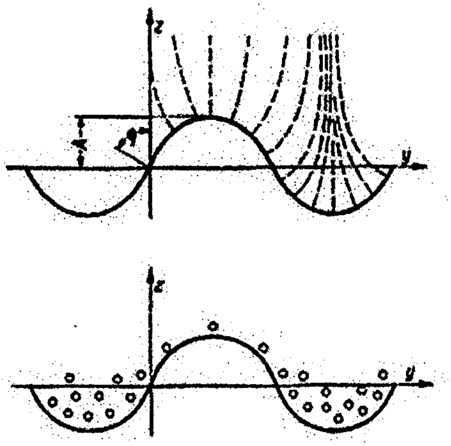


FIG. 5. Orientational effect of curved surfaces, after ADAMCZYK [1].

If molecules are parallel to the surface, the energy is equal to zero. Hence, from the energetic point of view, such planar texture is durable where the long axis of molecule coincides with the groove direction.

It was found that the elastic energy in the bulk is minimized by having the director oriented more or less parallel to the grooves. A nematic spontaneously orients its molecular axis parallel to the grooves. The parallel alignment of molecules can be achieved most easily by rubbing the adjacent surface or other techniques of generation of some grooves. The orientation determined by physico-chemical interactions depends on the difference in surface tension of the substratum and the liquid crystalline substance. When the critical surface tension of the surface exceeds the surface tension of the liquid crystal, the alignment of liquid crystalline molecules is parallel to the surface, and when the reverse case takes place, the alignment is perpendicular.

The alignment is extremely important because it determines the physical properties and the manner in which the molecules are reoriented by an applied external field [36]. Examples of the alignments are shown in Fig. 6. The thickness of the liquid crystal layer between the confining surfaces has significant effects on the physical properties. The alignment of molecules of thin liquid crystalline film may be determined practically only by the interaction with the confining surfaces. The thickness of oriented layer can attain 1 mm [1].

The external field applied deforms the texture generated by the described surface interactions. Once the external field ceases to operate, the same surface interactions restore the primal texture in a layer of liquid crystal.

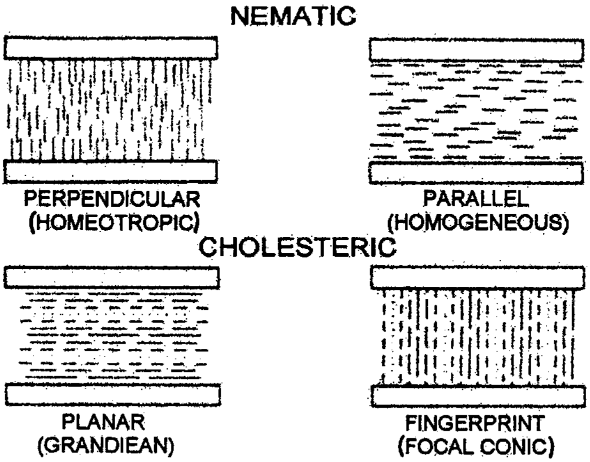


FIG. 6. Basic alignment modes of nematic liquid crystals (A), and cholesteric liquid crystals (B), after MARGERUM *et al.* [36].

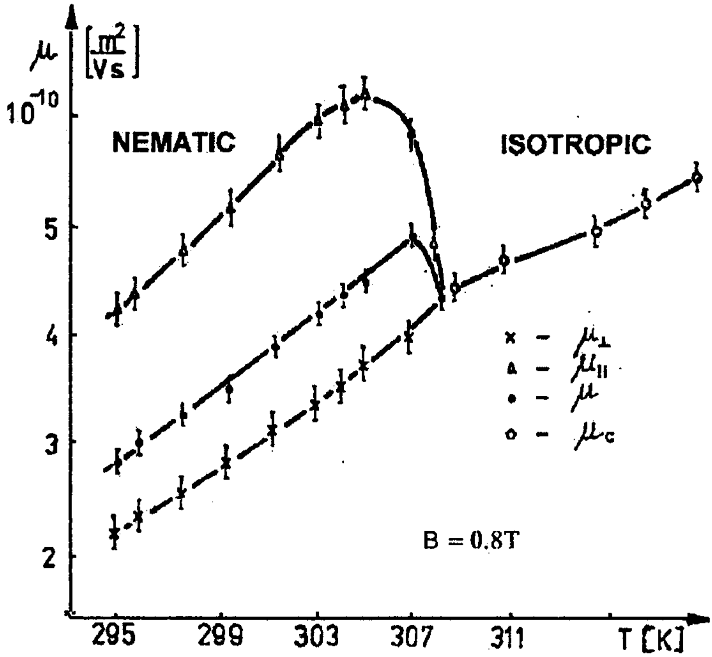


FIG. 7. Temperature dependence of ions mobility for a nematic liquid crystal pentyl- cyano-biphenyl in nematic phase and isotropic phase. In the nematic phase: $\mu_\perp$ – for orientation by magnetic field B perpendicular to direction of electric field E, $\mu_\parallel$ – for orientation parallel to direction of electric field E, μ – without orientation, and isotropic phase – (μ_c), after SZWAJCZAK [56]; B=0.8 T.

2.5. Some experimental data

Liquid crystals subject to large external fields exhibit isotropic behaviour [56]. We present some results of measurements for a nematic liquid crystal which belong to cyano-biphenyls. This type of liquid crystalline material is mainly used in displays as well as in thermography as admixture. Molecules of material are arranged by magnetic field B. Fig. 7 depicts the temperature dependence of the mobility of charge carriers. As we can see, material is anisotropic in the range of mesophase. Above the point of phase transition, the material is isotropic. Also, the anisotropic viscosity is observed for temperature below the temperature of phase transition, and isotropic viscosity above the phase transition, cf. also Fig. 3. In Fig. 8 the anisotropic electric conductivity of the same material is presented. We see that the anisotropy decays for higher electric fields.

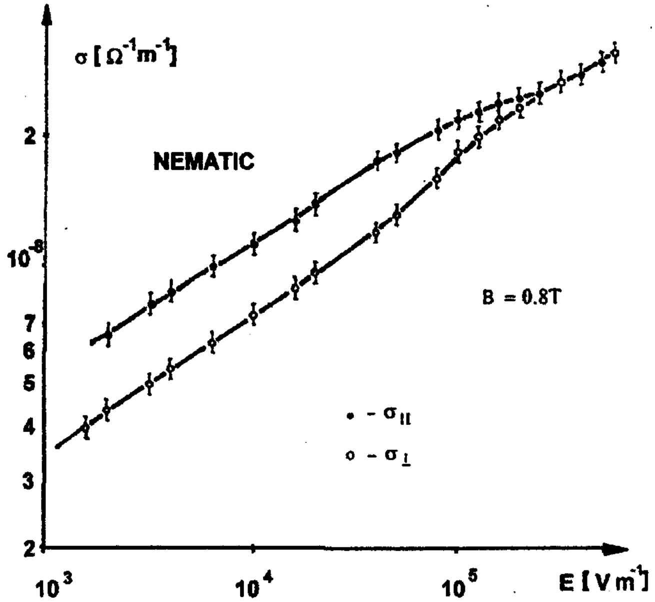


FIG. 8. Dependence of the electric conductivity on electric field for an oriented nematic liquid crystal pentyl-cyano-biphenyl: $\sigma_\perp$, $\sigma_\|$ – for orientation by magnetic field B perpendicular and parallel to direction of electric field E, respectively, after SZWAJCZAK [56]; B=0.8 T.

3. STRUCTURE OF NATURAL JOINTS

Considerable attention has been paid to studies of tissue composition, structural factors, and biochemical and biophysical properties of normal and diseased synovial joints. In this section we briefly describe the chemical composition, molecular structure and ultrastructural organisation of synovial joints and relate them to their biomechanical properties and functions.

The synovial joint may be described as a system of two articulating bones covered with layers of hydrated soft tissue called cartilage. Articular cartilage layers and meniscus are important elements of joints. The joint cavity is filled with the synovial fluid. The synovial joint is enclosed by a strong fibrous capsule.

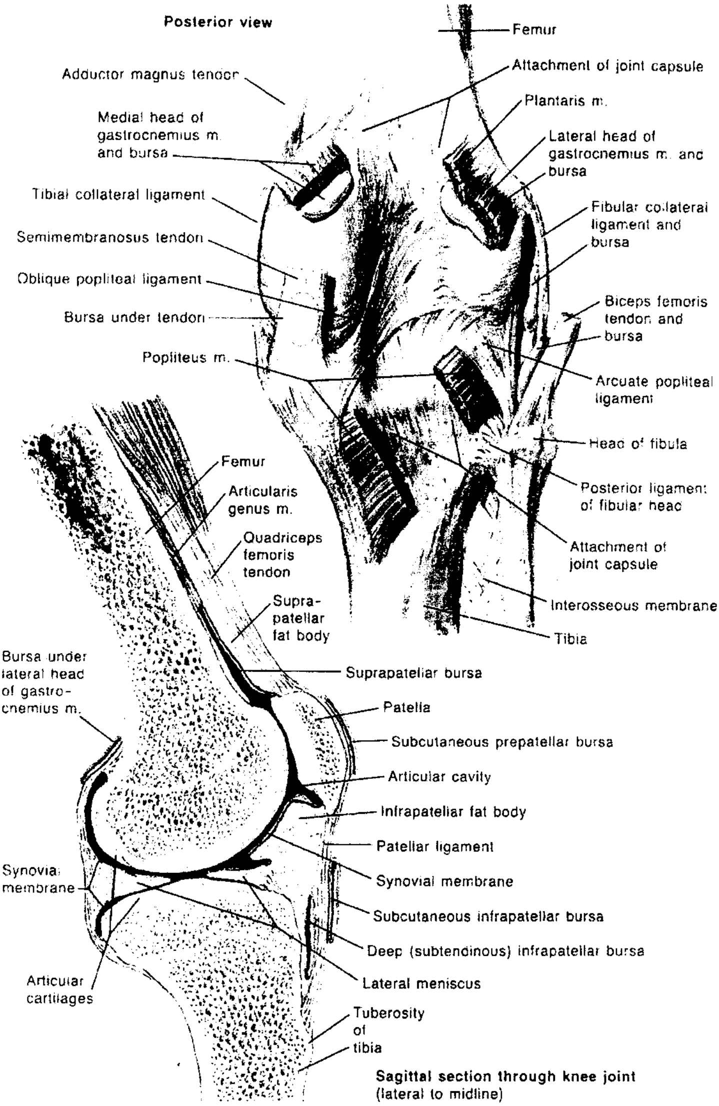


FIG. 9. Knee joint, after CIBA-GEIGY catalogue [10].

The capsule is linked with the synovium. The last secretes the synovial fluid and provides the nutrients for tissues [40]. Stability of the system is provided by ligaments, tendons and muscles.

Only synovial joints or diarthrodial joints allow for a large degree of relative motion between the contacting bones. The shoulder, elbow, hip, knee and ankle are synovial joints [10]. An example of this type of junctures is shown in Fig. 9.

The geometrical structure of the synovial joint and physical, chemical, mechanical, electrical and biological processes which take place in it insure the unfailing functioning in the case of healthy system and low friction lubrication mechanisms. Our study is focused on the system the synovial fluid – cartillage.

3.1. *Synovial fluid*

The synovial fluid is a yellow and clear, highly viscous liquid. Small amounts of this fluid, to 5 ml, are contained in animal and human synovial joints [40]. It exists as a thin film layer. The thickness of the layer varies and depends on the specific joint and location within it. It ranges from 6 μm to 1 mm.

The synovial fluid is a dialysate of blood plasma; it contains about one-third of the protein concentration of the plasma. The synovial fluid of the healthy joint consists to 94% of water, contains a very specific polymer known as the hyaluronic acid, hyaluronan or hyaluronate (HA) – $(2-3)\%$ by weight. Moreover, the synovial fluid contains some macromolecular components like glycoproteins, phospholipids and low molecular compounds, e.g., liquid crystalline cholesterol esters and small ions [31].

Hyaluronan is an unbranched macromolecule which is composed of repeating disaccharide units. A length of a typical macromolecule chain is of the order of 0.5 to 5μm. The molecular weight of the hyaluronan is up to 10^6 [39]. At a certain critical concentration a three-dimensional molecular network of HA can form a gel (at least 1% – solutions), cf. [50] and the references therein.

The viscosity of the synovial fluid depends on the concentration of the hyaluronic acid macromolecules [21, 41], cf. Fig. 10, and on the length and conformation of hyaluronan chain. For healthy human synovial fluid it ranges from 100 Pa·s at low shear rate ($0.1\ \mathrm{s}^{-1}$) to 0.02 Pa·s at much higher shear rates ($1000\ \mathrm{s}^{-1}$). Rheological properties are affected by pathological processes [12], see Fig. 11. The viscosity coefficients of synovial fluid from degenerated joints are smaller than those exhibited by the normal synovial fluid [21].

Since the synovial fluid contains different macromolecular components, in water it exhibits non-Newtonian behaviour. The synovial fluid serves as the lubricant for the synovial joints. Furthemore, the synovial fluid provides the necessary nutrients for cartilage, protects the cartilage against enzyme activity, and acts as a medium in morphological fields. FUNG [21] discussed the viscoelastic property of the synovial fluid. The elasticity was confirmed in 1953 by Ogston and Stanier, see FUNG [21].

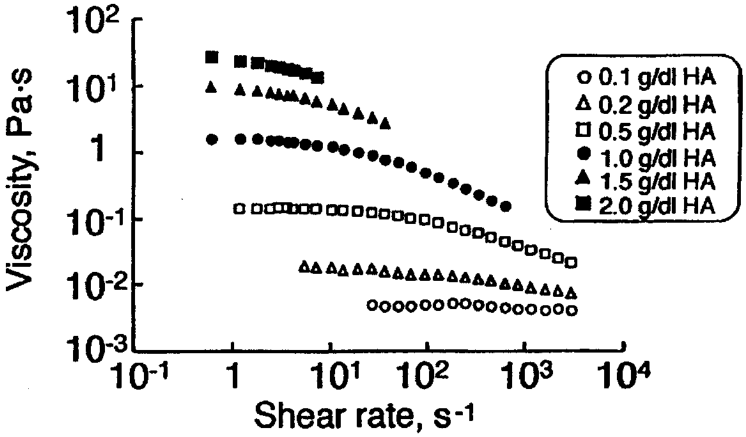


FIG. 10. Saline solution of sodium hyaluronate for various HA concentrations, after MURAKAMI *et al.* [41].

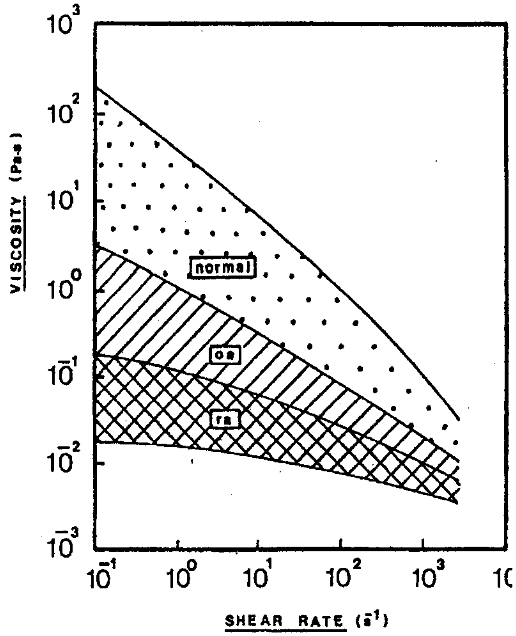


FIG. 11. Viscosity of normal and pathological human synovial fluid, after DOWSON [12], oa – osteoarthritis, ra – rheumatoid arthritis.

3.2. Articular cartilage

The articular cartilage is a soft, charged, porous and hydrated connective tissue. The cartilage material is a viscoelastic material. It combines the properties of elastic solid and viscous fluid. It can be regarded as some kind of composite or as multiphase material. It is composed of a fluid phase (water and dissolved electrolytes) and a solid phase (collagen – mainly type II, proteoglycans, proteins, glycoproteins, chondrocytes).

According to MOW *et al.* [39], water and dissolved ions like Na^+, Ca^{++}, Cl^- comprise approximately 68% to 85% of the wet weight of the healthy articular cartilage. The solid phase comprises 15% to 32% of normal articular cartilage by wet weight. Quantitatively major non-aqueous components of cartilage are collagen II (10–20)% and proteoglycan called aggrecan (5–10)%. Hyaluronan, link proteins, collagens type I, collagens type V, collagens type VI, collagens IX, collagens type XI, small proteoglycans and other quantitatively minor ingredients like metabolically active chondrocytes, comprise about 5% of cartilage.

Collagen exhibits a high degree of structural organisation. There are at least 25 different types of collagen [31]. All collagens have a basic structure of three chains forming a triple helix. Types I, II, X, XI are fibrillar collagens, whereas types VI and IX are nonfibrillar collagens. Type II collagen is the primary collagen of articular cartilage. The basic structural unit of this collagen is formed of three α polypeptide chains, namely α chains. Each of the α chains is the left-handed helix which together form a right-handed helix called tropocollagen. We observe similarity to the liquid crystalline helix. Tropocollagen molecules in cartilage are polymerised extracellularly to form microfibrils. The diameter of microfibrils is of the order of 3 – 5 nm whilst that of fibrils is up to 100 nm or more. In the next stage, the fibres are constituted. Their diameter is up to 10 μm [31]. Schematic representation of collagen fibers is presented in Fig. 12.

The primary function of the collagen appears to ensure good tensile properties to the tissues. The collagen of type IX stabilizes the three-dimensional organisation of the collagen network. The collagen of type IX is covalently linked to at least one collagen molecule of type II.

The molecules of proteoglycans are very large since their molecular weight is of the order of 10^6. Their macromolecular organisation is quite complex. Numerous glycosaminoglycan chains, being chondroitin sulfate and keratan sulfate, are attached to a single protein core molecule. Furthermore, proteoglycans are attached by a link protein to a single hyaluronate chain, thus forming a proteoglycan aggregate. The molecular weight of this supra-molecule can be as high as $2{\cdot}10^8$. Schematic representation of proteoglycan aggregate is shown in Fig. 13.

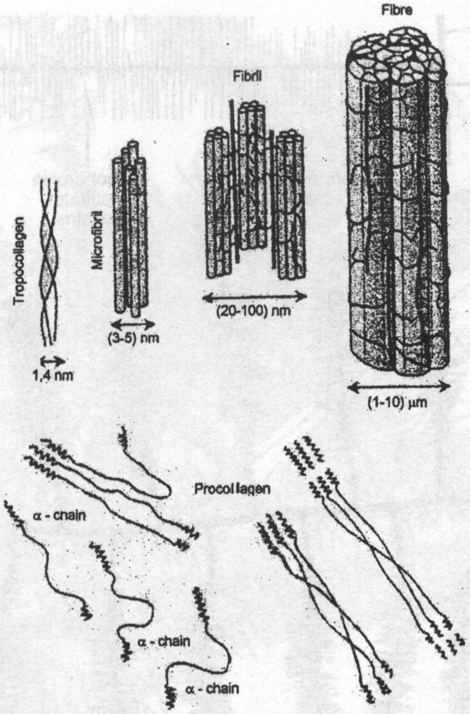


FIG. 12. Scheme of collagen structure in articular cartilage, after KUPCHINOV *et al.* [31].

Each proteoglycan is a macromolecular polymer, similar to a bottle-brush. The conformation of proteoglycans and proteoglycan aggregates resembles the conformation of typical side-chain liquid crystalline polymers. Glycosaminoglycans chains are bound to protein chains as their side-groups and proteoglycans are attached to hyaluronan chains as their side-groups, [2, p. 6], cf. Fig. 13.

The collagen and proteoglycan network constitute the solid entanglement which contains the interstitial water [31]. A schematic diagram of the proteoglycan-collagen matrix is shown in Fig. 14.

The cartilage can be regarded as porous and permeable solid matrix. According to HUYGHE [25], it contains two fluid compartments: the intrafibrillar water inside the collagen fibrils and the extrafibrillar water outside the collagen network. In undamaged (healthy) cartilage, the portion of extrafibrillar water constitutes approximately 70%. The extrafibrillar water may jointly move throughout the tissue with ions. The water content is regulated by pressure gradients between the extra- and intrafibrillar compartments. The water plays a significant role in governing the equilibrium swelling, compressive stiffness and elasticity of the tissue. Other mechanical, elastic, rheological and electrokinetic properties also depend on the content of water.

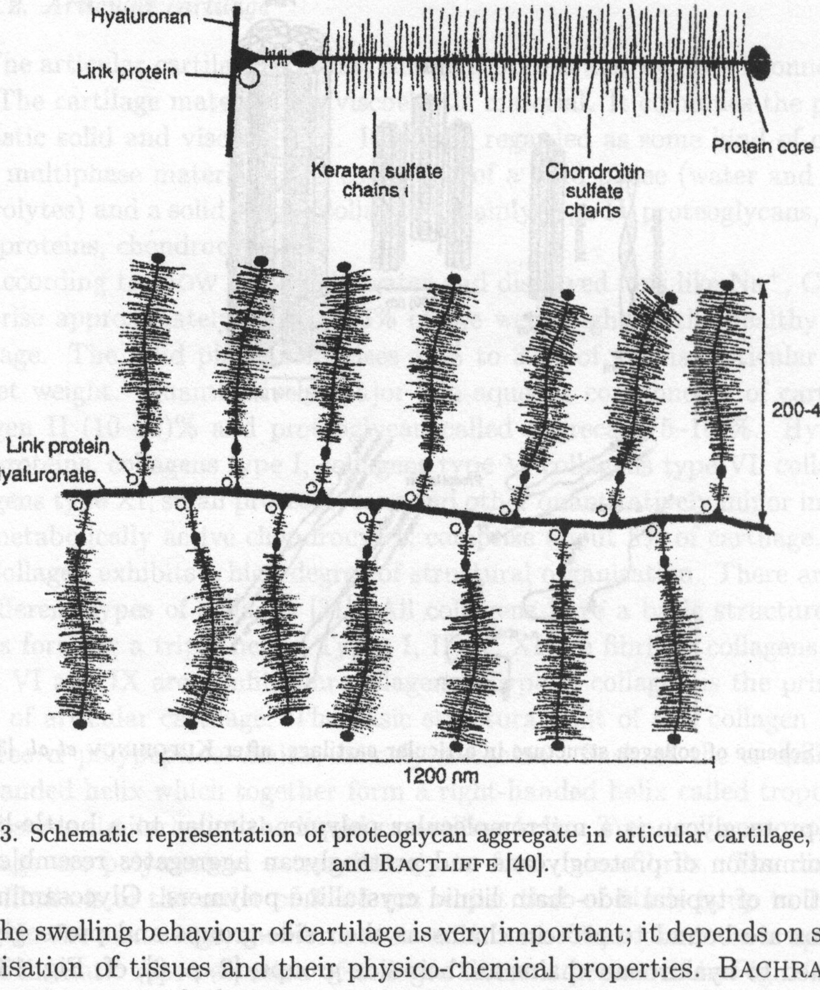


FIG. 13. Schematic representation of proteoglycan aggregate in articular cartilage, after MOW and RACTLIFFE [40].

The swelling behaviour of cartilage is very important; it depends on structural organisation of tissues and their physico-chemical properties. BACHRACH *et al.* [4] and MOW *et al.* [40] observed anisotropic swelling behaviour of the cartilage. The anisotropy of cartilage is primarily due to the collagen fibres orientation. The collagen fibres form in the cartilage surface a system of anisotropic microreliefs, see Fig. 15A. The collagen-fiber network exhibits a nonlinear and anisotropic behaviour, cf. [40] and the references therein. However, in confined compression the behaviour of articular cartilage seems to be isotropic, cf. [39, p. 278].

The human articular cartilage thickness varies among joints and ranges from 1 to 6 mm. The normal pore size in cartilage ranges from 2.5 to 6.5 nm. The macromolecular solutes of the synovial fluid are too large to penetrate into the cartilage tissue [23]. Only the water and small particles like ions and low molecular liquid crystals can penetrate into the cartilage. The roughness of the articular surface is of the order several of nm. The thickness of the fluid film during the boundary lubrication is of the same order.

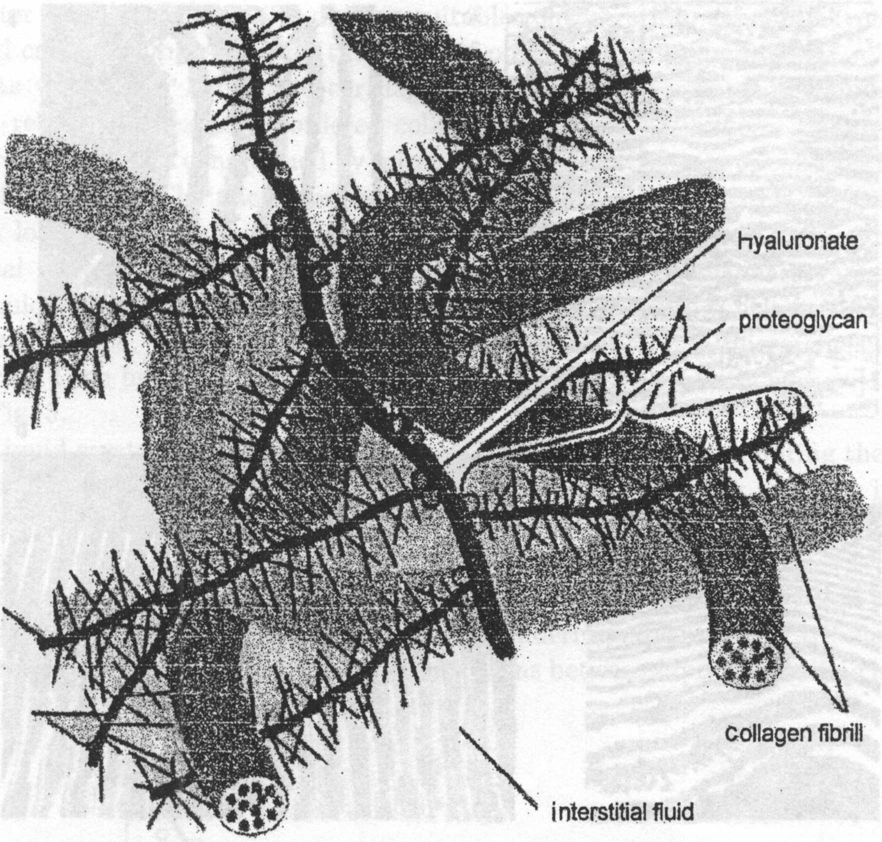


FIG. 14. Scheme of structural organisation of articular cartilage, after KUPCHINOV *et al.* [31].

The surface porosity plays the role in the regulation of the frictional response of cartilage. According to ATESHIAN *et al.* [3], the friction coefficient is load and time-dependent.

The flow of fluid within the joint cavity and the exchange of fluid to and from the cartilage plays an important role in joint functioning and nutrition of cartilage. It also includes the regulation (change) of synovial fluid volume as well as of concentration of HA and low molecular contents in water.

The joint system as a whole is electroneutral under normal, physiological conditions. The interstitial fluid with dissolved electrolytes and the solid skeleton of proteoglycans are charged, cf. [23]. Tissues, similarly to liquid crystals, are typical dielectrics [45, 51]. The structure parameters and phase transitions strongly depend on the water content, head-group structure (hydrophobic, hydrophilic), length and branching of carbon chains of macromolecules and charge of proteoglycans.

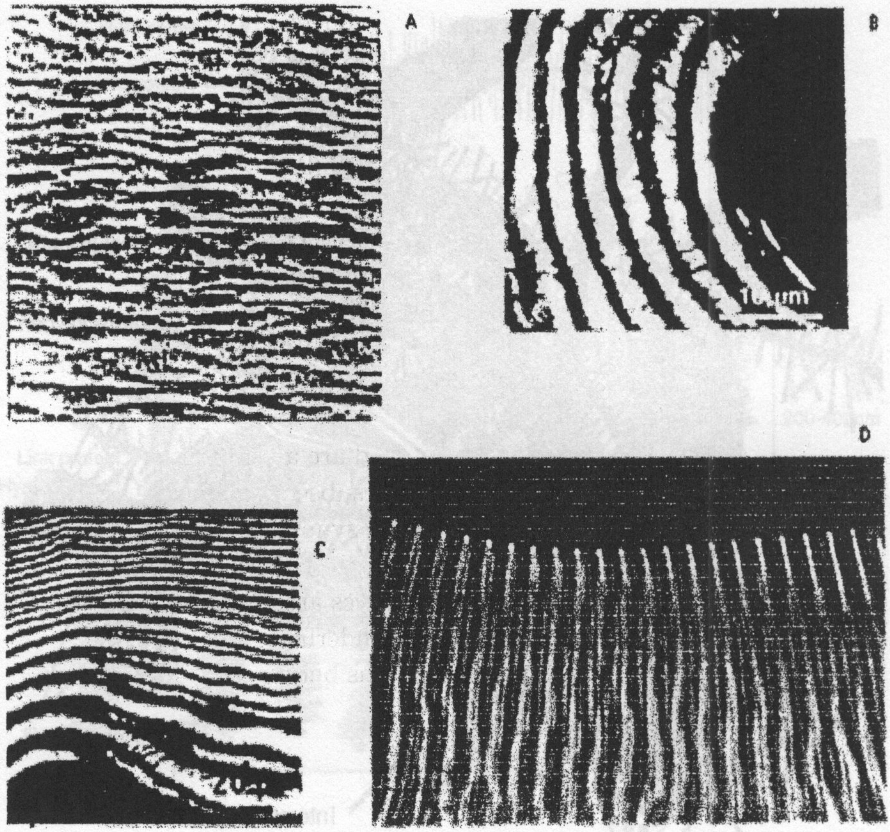


FIG. 15. Examples of liquid crystalline structures of selected biological materials: (A) – surface of an articular cartilage (magnification × 300), after KUPCHINOV *et al.* [31], (B) – single osteon from a human compact bone (magnification × 2000), after GIRAUD-GUILLE [22], (C) – decalcified crab cutile (magnification × 250) after GIRAUD-GUILLE [22], and for comparison (D) – thin layer of liquid crystal with typical cholesteric texture, after ADAMCZYK [1].

4. LIQUID CRYSTALLINE STRUCTURES IN SYNOVIAL JOINTS: EXPERIMENTAL DATA

KUPCHINOV *et al.* [31, 32] and BELOYENKO *et al.* [6] investigated the synovial fluid and compared its response with the response of various artificial fluids [19, 32, 48]. Optical methods like polarisation microscopy, raster microscopy, scanning electron microscopy, birefringence, and DTA (differential temperature analysis), chromatography method, tribological and rheological methods were used.

It has been found that synovial fluid exhibits liquid crystalline behaviour. The synovial fluid contains low-molecular weight cholesterol esters and high-molecular weight hyaluronic acid. The hyaluronan, like cellulose derivatives, is a water soluble polymer and, under a suitable concentration, generates lyotropic liquid crystalline phase within the range of physiological temperatures.

As we already know, the cartilage surface exhibits anisotropic roughness. Such reliefs are created by ordered collagen structures. Submicro-, micro- and macro-roughness is conditioned by micro- and fibrillar structure of collagen fibers [17, 18, 19, 31]. The microgrooves and grooves are oriented in the direction of main locomotion of synovial joint, see Fig. 15A. The privileged axes of liquid crystal molecules coincide with the axes of grooves. This planar organisation of liquid crystalline molecules corresponds to the minimum of elastic energy of molecules [1, 5, 7, 19]. If liquid crystal molecules adjust to microreliefs, they deform. Then bend and splay deformations demand higher elastic energy [1, 19], see Fig. 5.

Liquid crystalline molecules of synovial fluid are also oriented during the flow according to the flow direction. The flow is a subsequent factor assisting in arrangement of liquid crystalline molecules of the synovial fluid along microgrooves on the surface of cartilage.

According to the papers [19, 31], these grooves are covered by very thin layers of liquid crystals of the synovial fluid, thus rendering advantageous tribological characteristics. Weak intermolecular interactions between liquid crystalline layers

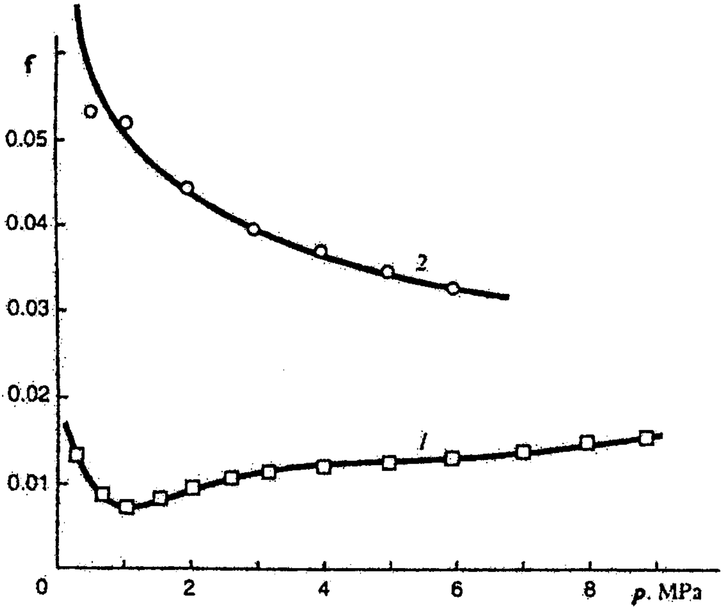


FIG. 16. Anisotropy of friction coefficient f versus pressure p for cartilage-glass system lubricated by synovial fluid: 1 – the friction direction along microgrooves of cartilage; 2 – the direction perpendicular to microgrooves of cartilage, after KUPCHINOV *et al.* [31].

ensure low friction, being anisotropic. This fact was confirmed experimentally for the cartilage-glass system lubricated by the synovial fluid. The friction coefficient along the direction of preferred motion in the joint is twice smaller than the friction coefficient in the direction perpendicular to microgrooves of cartilage, see. Fig. 16.

Similar behaviour was revealed for other systems, like animal cartilage-cartilage (in vitro), cartilage-glass, cartilage-steel, glass-glass, etc., lubricated by the lubricant with liquid crystals as dopant (admixture). Various multicomponent artificial lubricants were investigated. Synthetic lubricants, which contain cholesteric liquid crystals, have properties similar to healthy synovial fluid [6, 32].

Rheological and tribological investigations of synovial fluid with various concentration of cholesteric compounds and pseudosynovial fluids with various concentration of cholesterol ester admixtures manifest the important role of liquid crystalline components of synovial fluid which is treated as a lubricant, cf. Fig. 17.

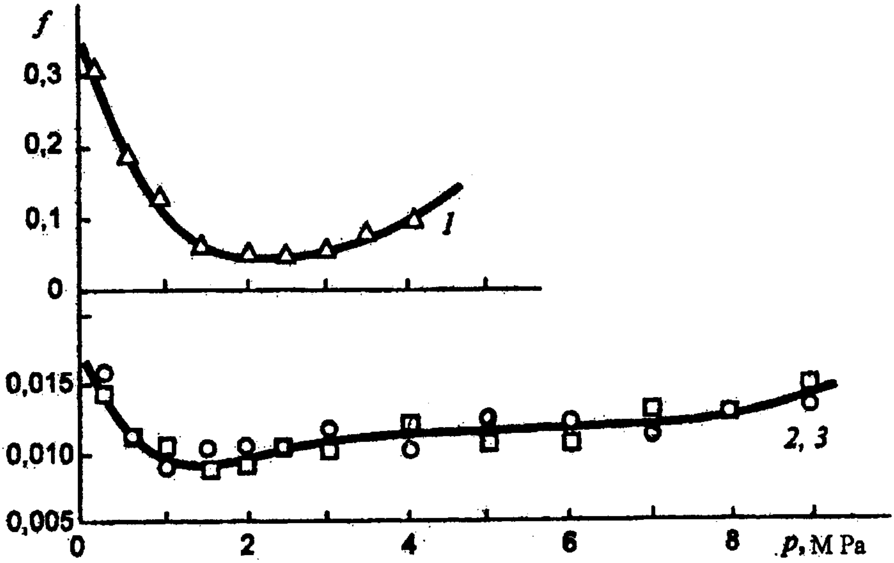


FIG. 17. Friction- pressure $f(p)$ characteristics for cartilage-glass system lubricated by: 1 – pseudosynovial fluid, 2 – synovial fluid, 3 – pseudosynovial fluid with liquid crystalline dopant, after KUPCHINOV *et al.* [31].

KUPCHINOV *et al.* [31, 32] studied the decrease of amplitude in the classical test when the pendulum tribometer is used, cf. Fig. 18. The largest amplitude was observed in the case of lubrication of cartilage with the synovial fluid containing a significant amount of cholesteric materials, i.e., liquid crystalline cholesteric esters. Also, pseudosynovial fluids with liquid crystalline admixtures revealed to be good lubricants. The larger the liquid crystals concentration, the smaller the friction coefficient, cf. [32]. The frictional parameters of pairs like cartilage-cartilage, cartilage-glass, steel-steel, etc., lubricated with pseudosynovial fluids containing liquid crystals, are similar to parameters of those systems lubricated with the synovial fluid.

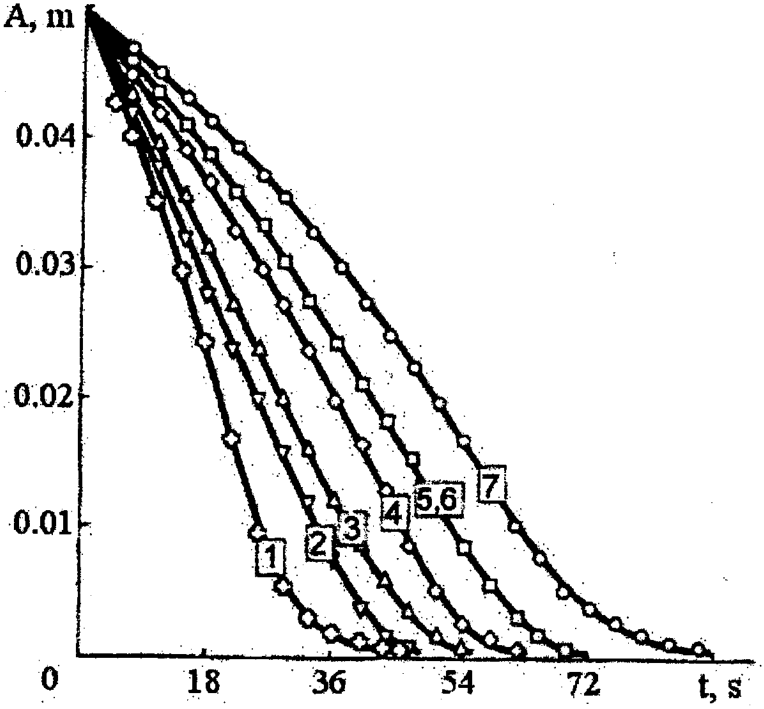


FIG. 18. Dependence of damping amplitude in the case of cartilage friction and lubrication with: 1 – distilled water, 2 – silicone-organic fluid used in joint treatment, 3 – pseudosynovial fluid (water solvent of carboxymetylcelulose), 4 – silicone-organic fluid containing 2% admixture of cholesteric liquid crystals, 5 – pseudosynovial fluid containing 2% admixture of cholesteric liquid crystals, 6 – synovia containing 0.9 mmol/l of cholesteric liquid crystals, 7 – synovia containing 4.09 mmol/l of cholesteric liquid crystals, after KUPCHINOV *et al.* [32].

Microscopic investigations of GIRAUD-GUILLE [22] show that three-dimensional arrangements of biological polymers in living cells and tissues, such like compact bones and crab cuticles, are similar to arrangements of molecules in liquid crystals. Collagen fibrillar networks follow the same spatial distribution as those described in cholesteric liquid crystalline materials. The particular geometry of collagen fibrils in compact bone, cartilage and cuticle is analogous to the distribution of molecules in cholesteric liquid crystals, see Fig. 15. Moreover, a specific state of matter combining both fluidity and order was obtained in highly concentrated solutions of collagen molecules. Those macromolecules were arranged in a manner reminiscent of cholesteric liquid crystals.

PURSLOW *et al.* [46] found helical structures in soft connective tissues like cuticles, tendons, fascia and arterial walls. Time-resolved X-ray diffraction was used to monitor structure changes during squeeze, strain and creep. Ordering of bundles of collagen fibrils was typical for liquid crystals.

5. MATHEMATICAL THEORY OF LIQUID CRYSTALS

The Newtonian model (by definition) cannot describe fluids which possess substructures, and in particular liquid crystals. The particles of such complex fluids may be of different shape, may shrink and expand, and, moreover, they may rotate independently of the rotation and movement of the fluid. To account for these local aspects, classical concepts of continuum mechanics had to be augmented with additional laws and new constitutive relations should be given.

OSEEN [44] initiated the development of continuum theory of liquid crystals where the material points of liquid crystals are treated as oriented points. A review of various developments was given by STEPHEN and STRAHLEY [55], see also the book by de GENNES [11] and [26]. In 1997 ERINGEN [15] proposed a unified continuum theory of liquid crystals. Below we follow just this unified theory, cf. also [16, 47].

5.1. Kinematics

A micropolar continuum is a collection of orientable material points. A material point P in the reference state $\mathbf{B}_R$ is identified by the position vector (X_K), $K = 1, 2, 3$ and three rigid directors χ_K attached to the point. These directors are used to represent the orientation degrees of freedom of liquid crystals and their existence is an important difference between the theory and the classical continuum theory. The motion, at time t, carries P from (X_K) to a spatial point (x_k) and rotates χ_K to χ_k in the spatial configuration $\mathbf{B}_S$. This rotation is called microrotation. Both X_K and x_k are referred to the same rectangular coordinate system. The motion is expressed by

$$x_k = x_k(X,t); \qquad \chi_{kK} = \chi_{kK}(X,t); \qquad k, K = 1,2,3. \tag{5.1}$$

It is assumed that $(x_k(X,t))$ and $(\chi_{kK}(X,t))$ possess continuous partial derivatives with respect to X_k and t and they are uniquely invertible

$$X_k = X_k(x,t); \qquad \chi_{Kk} = \chi_{kK}^{-1}(x,t), \tag{5.2}$$

so that

$$\chi_{kK}\chi_{lK} = \delta_{kl}; \qquad \chi_{kK}\chi_{kL} = \delta_{KL}, \tag{5.3}$$

$$x_{k,K}X_{K,l} = \delta_{kl}; \qquad X_{K,k}x_{k,L} = \delta_{KL}. \tag{5.4}$$

An index following a comma represents a partial derivative, e.g.,

$$x_{k,K} = \frac{\partial x_k}{\partial X_K}, \qquad X_{K,k} = \frac{\partial X_K}{\partial x_k}, \tag{5.5}$$

and the repeated indices are summed over the range 1, 2, 3.

The presence of directors gives rise to intrinsic angular velocity (microrotation) ν_k

$$\nu_k(x,t) = -\frac{1}{2}\epsilon_{klm}\nu_{lm}, \tag{5.6}$$

$$\nu_{kl} = -\epsilon_{klm}\nu_m, \tag{5.7}$$

where (ν_{kl}) is the gyration tensor defined by

$$\nu_{kl}(x,t) \equiv \dot{\chi}_{kK}\chi_{lK} = -\nu_{lk}, \qquad \dot{\chi}_{kK} = \nu_{kl}\chi_{lK}. \tag{5.8}$$

A superposed dot denotes the material derivative, sometimes denoted by $\frac{D}{Dt}$, and ϵ_{klm} are components of the permutation symbol.

The three directors may be used to form the strain tensor $\boldsymbol{\gamma} = (\gamma_{ij})$:

$$\gamma_{kl} \equiv \frac{1}{2}\epsilon_{kmn}\chi_{mK}\chi_{nK,l} \tag{5.9}$$

and the two deformation-rate tensors, (a_{kl}) and (b_{kl}), defined by:

$$a_{kl} \equiv v_{l,k} + \epsilon_{klm}\nu_m, \qquad b_{kl} \equiv \nu_{k,l}. \tag{5.10}$$

The spin inertia $\dot{\sigma}_l$, is defined by

$$\dot{\sigma}_l = \frac{D}{Dt}(j_{lm}\nu_m). \tag{5.11}$$

The presence of rigid directors implies the rigid body nature of material particles. A material particle is a collection of aggregate molecules with an orientational order. The microinertia tensor (j_{kl}) expresses the angular momentum and the molecular ordering, and is defined by:

$$j_{kl} = J_{KL}\chi_{kK}\chi_{lL}. \tag{5.12}$$

Obviously, (J_{KL}) stands for the microinertia tensor of the material particle in the reference configuration.

5.2. Balance laws

Let us pass to the formulation of balance laws for liquid crystals and let us introduce additional notations:

ρ – mass density,
$\mathbf{t} = (t_{kl})$ – stress tensor,
$\mathbf{f} = (f_l)$ – body force density,
$\mathbf{m} = (m_{kl})$ – couple stress tensor,
$\mathbf{l} = (l_l)$ – body couple density,
ε – internal energy density,
$\mathbf{q} = (q_k)$ – heat vector,
η – entropy density,
T – absolute temperature,
$\mathbf{n}$ – normal vector to the surface S,
h – heat input,
S – surface of discontinuity.

Conservation of mass

$$\begin{aligned} \dot{\rho} + \rho \nabla \cdot \mathbf{v} &= 0 \quad \text{in} \quad V \setminus S, \\ [\rho(\mathbf{v} - \mathbf{u})] \cdot \tilde{\mathbf{n}} &= 0 \quad \text{on} \quad S. \end{aligned} \tag{5.13}$$

Conservation of microinertia

$$\begin{aligned} \dot{j}_{kl} - \nu_{km} j_{lm} - \nu_{lm} j_{km} &= 0 \quad \text{in} \quad V \setminus S, \\ [\rho j_{kl}(v_r - u_r)]\, \tilde{n}_r &= 0 \quad \text{on} \quad S. \end{aligned} \tag{5.14}$$

Balance of momentum

$$\begin{aligned} t_{kl,k} + \rho(f_l - \dot{v}_l) &= 0 \quad \text{in} \quad V \setminus S, \\ [t_{kl} - \rho v_l (v_k - u_k)]\, \tilde{n}_k &= 0 \quad \text{on} \quad S. \end{aligned} \tag{5.15}$$

Balance of moment of momentum

$$\begin{aligned} m_{kl,k} + \varepsilon_{lmn} t_{mn} + \rho(l_l - \dot{\sigma}_l) &= 0 \quad \text{in} \quad V \setminus S, \\ [m_{kl} - \rho \sigma_l (v_k - u_k)]\, \tilde{n}_k &= 0 \quad \text{on} \quad S. \end{aligned} \tag{5.16}$$

Balance of energy

$$\begin{aligned}
&p\dot{\varepsilon} - t_{kl}a_{kl} - m_{kl}b_{lk} - q_{k,k} - \rho h = 0 \quad \text{in} \quad V \setminus S, \\
(5.17)\qquad &\Big[\Big(p\varepsilon + \frac{1}{2}\rho \mathbf{v}\cdot\mathbf{v} + \frac{1}{2}\rho\sigma\cdot\mathbf{v}(v_k - u_k)\Big)\Big] \\
&\qquad\qquad -t_{kl}v_l - m_{kl}v_l - q_k]\,\tilde{n}_k = 0 \quad \text{on} \quad S.
\end{aligned}$$

Second law of thermodynamics

$$\begin{aligned}
&\rho\dot{\eta} - (q_k/T)_{,k} - \rho h/T \geq 0 \quad \text{in} \quad V \setminus S, \\
(5.18)\qquad &\left[\rho\eta(v_k - u_k) - \frac{1}{T}q_k\right]\tilde{n}_k \geq 0 \quad \text{on} \quad S.
\end{aligned}$$

A useful form of Eq. (5.18) is the generalized Clausius-Duhem inequality

$$(5.19)\qquad -\frac{\rho}{T}\left(\dot{\psi} + \eta\dot{\theta}\right) + \frac{1}{T}t_{kl}a_{kl} + \frac{1}{T}m_{kl}b_{lk} + \frac{q_k}{T^2}T_{,k} \geq 0.$$

We note that when microinertia tensor vanishes identically, the classical theory is recovered.

5.3. *Constitutive equations for liquid crystals*

The state of liquid crystals (nematics, smectics, and others) is characterized by a set of constitutive equations that express the dependent constitutive variables (ε,η,q_k,t_{kl},m_{kl}) as functions of independent variables $(\rho^{-1}, T, j_{kl}, a_{kl}, b_{kl}, T_{,k}, \dot{T})$.

The recoverable constitutive variables ${}_RY$ are defined by

$$(5.20)\qquad {}_RY = (\rho^{-1}, T, j_{kl}, \gamma_{kl})$$

and the dynamic part of the independent variables, called also thermodynamic forces, is:

$$(5.21)\qquad {}_DY = (T, T_{,k}, \dot{T}, a_{kl}, b_{kl}).$$

In the paper by ERINGEN [15] general form of constitutive equations for liquid crystals is postulated. Next, they are formulated for each group separately. The constitutive equations are derivable from two functions: the free energy $\psi({}_RY)$ and the dissipation potential $\Phi({}_RY,{}_DY)$. These functions are restricted by the axiom of objectivity. For instance, we will present below constitutive equations for nematics liquid crystals.

Particles of nematic liquid crystals possess a center of symmetry. Hence the free energy function ψ depends on the invariants of (j_{kl}, γ_{kl}) under the full group of orthogonal transformations. The set of basic invariants consists of 22 elements. The symmetric and skew-symmetric parts of the tensor $\boldsymbol{\gamma}$ are

$$\boldsymbol{\gamma}^s = \frac{1}{2}\left(\boldsymbol{\gamma} + \boldsymbol{\gamma}^T\right), \qquad \boldsymbol{\gamma}^a = \frac{1}{2}\left(\boldsymbol{\gamma} - \boldsymbol{\gamma}^T\right). \tag{5.22}$$

For constitutive equations of the first degree in $\mathbf{j}$ and the first degree in γ, we only need eight of these invariants specified by, cf. [26],

$$\mathrm{tr}\mathbf{j}, \quad J_1 = \mathrm{tr}\boldsymbol{\gamma}^s, \quad J_2 = \mathrm{tr}(\mathbf{j}\boldsymbol{\gamma}^s), \quad J_3 = \mathrm{tr}\boldsymbol{\gamma}^{s2}, \tag{5.23}$$

$$J_4 = \mathrm{tr}(\mathbf{j}\boldsymbol{\gamma}^{s2}), \quad J_5 = \mathrm{tr}\boldsymbol{\gamma}^{a2}, \quad J_6 = \mathrm{tr}(\mathbf{j}\boldsymbol{\gamma}^{a2}), \tag{5.24}$$

$$J_7 = \mathrm{tr}(\mathbf{j}\boldsymbol{\gamma}^s\boldsymbol{\gamma}^a). \tag{5.25}$$

The free energy is then expressed as a polynomial of second degree in $\boldsymbol{\gamma}$ as

$$\begin{aligned} 2\rho\psi = (A_1 + A_2\mathrm{tr}\,\mathbf{j})J_1^2 + A_3J_1J_2 + (A_4 + A_5\mathrm{tr}\,\mathbf{j})J_3 + A_6J_4 \\ +(A_7 + A_8\mathrm{tr}\,\mathbf{j})J_5 + A_9J_6 + A_{10}J_7. \end{aligned} \tag{5.26}$$

A concise form of $\rho\psi$ is given by

$$\rho\psi = \frac{1}{2}A_{ijkl}\gamma_{ji}\gamma_{lk}, \tag{5.27}$$

where

$$\begin{aligned} A_{ijkl} \equiv (A_1 + A_2\mathrm{tr}\mathbf{j})\delta_{ij}\delta_{kl} + \frac{1}{2}A_5(j_{kl}\delta_{ij} + j_{ij}\delta_{kl}) \\ + \frac{1}{2}[A_4 + A_2 + (A_5 + A_8)\mathrm{tr}\,\mathbf{j}]\delta_{jk}\delta_{il} + \frac{1}{2}[A_4 - A_7 + (A_5 - A_8)\mathrm{tr}\,\mathbf{j}]\delta_{ik}\delta_{jl} \\ + \frac{1}{4}(A_6+A_9)(j_{il}\delta_{jk}+j_{jk}\delta_{il})+\frac{1}{4}(A_6-A_9)(j_{jl}\delta_{ik}+j_{ik}\delta_{il})+\frac{1}{4}A_{10}(j_{ik}\delta_{jl}-j_{jl}\delta_{ik}). \end{aligned} \tag{5.28}$$

Here A_i are the material moduli that depend on ρ^{-1}.

The dissipation function is given by

$$\Phi = \frac{1}{2}\alpha_{ijkl}a_{ij}a_{kl} + \frac{1}{2}\beta_{ijkl}b_{ji}b_{lk} + \frac{1}{T}d_{ijk}b_{ji}T_{,k} + \frac{1}{2T^2}k_{ij}T_{,i}T_{,j} \tag{5.29}$$

where α_{ijkl} and β_{ijkl} are of the form of Eq. (5.28) with A_i replaced by α_i and β_i, respectively, whilst d_{ijk} and k_{ij} are given by

$$\begin{aligned} d_{ijk} &= (d_1 + d_2\mathrm{tr}\mathbf{j})\epsilon_{ijk} + d_3\epsilon_{lij}j_{lk}, \\ k_{ij} &= (k_1 + k_2\mathrm{tr}\mathbf{j})\delta_{ij} + k_3j_{ij}. \end{aligned} \tag{5.30}$$

Here α_i, β_i, d_i and k_i are dynamic moduli, being the viscosities and heat conduction coefficients, that depend on ρ^{-1} and T.

The constitutive equations for the nematic liquid crystals are specified by

$$\mathbf{t} = {}_R\mathbf{t} + {}_D\mathbf{t}, \qquad \mathbf{m} = {}_R\mathbf{m} + {}_D\mathbf{m}, \tag{5.31}$$

where

$${}_Rt_{kl} = -\pi\delta_{kl} - {}_R\,m_{kr}\gamma_{rl}, \;\; \pi = -\frac{\partial\psi}{\partial\rho^{-1}}, \tag{5.32}$$

$${}_Rm_{kl} = A_{ijkl}\gamma_{ji}, \qquad {}_Dt_{kl} = \alpha_{ijkl}a_{ij}, \tag{5.33}$$

$${}_Dm_{kl} = \beta_{ijkl}b_{ji} + \frac{1}{T}d_{kli}T_{,i}, \qquad q_k = \frac{1}{T}k_{kl}T_{,l} + d_{ijk}b_{ji}. \tag{5.34}$$

The quantities: ${}_R\mathbf{t}$ and ${}_R\mathbf{m}$ denote recoverable (static) stress and couple-stress tensors whilst ${}_D\mathbf{t}$ and ${}_D\mathbf{m}$ are dynamic stress and couple-stress tensors; π is the thermodynamic pressure, and η is the entropy density.

From the equations written above it is clear that the constitutive behaviour of nematic liquid crystals is governed by the strain tensor (γ_{kl}), the deformation rate tensors $(a_{ij}), (b_{ij})$, temperature gradient and order parameter (j_{kl}).

5.4. Remark

To calculate hydrodynamics and tribological quantities of biobearing we should solve the flow field equations completed by proper boundary and initial conditions. The flow field equations follow from Eqs. (5.13) – (5.18) by taking into account the constitutive equations. The field equations consist of 20 partial differential equations for the determination of 20 unknowns: $\rho, v_{kl}, \nu_k, i_{kl}$ and T. Solution of lubrication problems within the framework of such a theory, presents major mathematical difficulties. The differential equations are nonlinear. Therefore some simplifications should be imposed. The classical approach to the lubrication problem permits to perform some simplification due to the geometry and physics of flow. As the simplest flow model for hip joints lubrications we can consider a squeezed liquid crystal flow between two parallel, flat walls. Mathematical formulation of lubrication problems within the framework of liquid crystal theory was not yet presented, but seems to be promising. Nevertheless it is not known to which group of liquid crystals belongs the synovial fluid. Moreover, in constitutive equations there are too many material constants and presently, experimental values of material moduli are not available.

6. MICROPOLAR FLUID THEORY

It seems that the micropolar fluid model proposed by ERINGEN [14] can describe fluids with a substructure. We recall that the synovia is a fluid with a specific microstructure. Micropolar fluids are usually isotropic, i.e. the microinertial tensor has the following form: $j_{kl} = J\delta_{kl}$. Here J denotes the microinertia coefficient. The deformation of particles is negligible whilst the flow is described by two independent vector fields: $\mathbf{v}$ (the velocity) and $\boldsymbol{\nu}$ (the microrotation). The constitutive equations contain 6 rheological coefficients: $\lambda_v, \mu_v, \kappa_v, \alpha_v, \beta_v$ and γ_v.

The field equations for the micropolar fluid in vectorial form can be written as follows:

$$
\begin{gathered}
(\partial\rho/\partial t) + \nabla\cdot(\rho\mathbf{v}) = \mathbf{0}, \\
-\nabla\pi + (\lambda_v + 2\mu_v + \kappa_v)\nabla\nabla\cdot\mathbf{v} - \frac{1}{2}(2\mu_v + \kappa_v)\nabla\times\nabla\times\mathbf{v} + \kappa_v\nabla\times\boldsymbol{\nu} \\
+\rho(\mathbf{f} - \dot{\mathbf{v}}) = 0, \\
(\alpha_v + \beta_v + \gamma_v)\nabla\nabla\cdot\boldsymbol{\nu} - \gamma_v\nabla\times\nabla\times\boldsymbol{\nu} + \kappa_v\nabla\times\mathbf{v} - 2\kappa_v\boldsymbol{\nu} + \rho(\mathbf{l} - \dot{\boldsymbol{\sigma}}) = 0.
\end{gathered}
\tag{6.1}
$$

The constitutive equations for the micropolar fluid are of the following form

$$
\begin{aligned}
t_{ij} &= (-p + \lambda_v v_{k,k})\delta_{ij} + \mu_v(v_{i,j} + v_{j,i}) + \kappa_v(v_{j,i} - \varepsilon_{klm}\nu_m), \\
m_{ij} &= \alpha_v\nu_{k,k}\delta_{ij} + \beta_v\nu_{i,j} + \gamma_v\nu_{j,i}.
\end{aligned}
\tag{6.2}
$$

The books [35, 38] summarize the results achieved in this field. The micropolar model was applied to modelling the synovial fluid in a biobearing during the squeezing motion [42, 53]. The results obtained suggest that decrease of structure concentration results in lowering the biobearing load capacity and in lowering the response time. The same phenomenon was observed when molecular length of HA was diminished. This result is confirmed by clinical observation and experimental data [12, 21]. In the papers [42, 53] the rheological coefficients were a priori assumed since the experimental data are still lacking. Only the geometrical dimensions were realistic.

6.1. *Micropolar model of liquid crystaline synovial fluid*

A question which naturally arises is how the isotropic micropolar model is related to liquid crystalline model of synovia. As we already know, liquid crystals are anisotropic, but under special conditions become isotropic, as was discussed in Sec. 2. The liquid cystals state of matter exists only in certain interval of concentration and temperature, for instance see Figs. 3, 7. Moreover, liquid crystals in organisms are strongly influenced by various external fields like mechanical, electric, magnetic, and thermal fields. Confining surfaces and flows also can lead to ordering. The concentration of admixtures and a change in chemical composition of solution also strongly influence the behaviour of liquid crystals. Due to porosity of the cartilage, the changes of concentration of HA and other admixtures of synovial fluid are possible. In biological systems with liquid crystaline structure, due to a variety of interactions, the phase changes of type order-order can easily take place (the phase changes in the region of mesophase). The phase changes of the type of liquid crystal-gel, liquid crystal-liquid crystalline gel, or liquid crystal-plastic state are also possible [8]. All external factors acting on a liquid crystal, once a corresponding limit value is exceeded, lead to decay of anisotropy. For instance, a passage to an isotropic liquid is thus possible. According to CHIKAMA [9], the molecules of HA in thin layers are surrounded by protein molecules thus forming ball-shaped complexes. Also, according to KOBAYASHI *et al.* [28], long chains of HA molecules form compact helical structures. The results of CHIKAMA [9] indicate that synovia behaves like an isotropic fluid whilst those due to KOBAYASHI [28] point out its anisotropy.

6.2. Modelling of biolubrication processes based on micropolar fluid theory

In the papers [42, 53] a very simple model of human joint was studied, namely a squeezing flow of micropolar fluid between two parallel plates. The lower plate is a fixed a porous solid. The upper plate, being parallel to the lower plate, is rigid and moves to the lower plate with the velocity U. In the Cartesian coordinate system (x, y, z) the lower plate is described by $y = 0$ whilst the upper plate by $y = h$. The translational velocity of the plate is $(0, -U, 0)$. The motion is very slow. The fluid leaves the domain between the plates in two directions, through the open boundary at $x = a$ and $x = -a$.

For the squeezing motion we assume that: 1) the flow is steady and incompressible, 2) the body forces and body couples are absent, 3) at any time t the thickness h of the gap between the two plates is much smaller than the characteristic in-plane dimension of the plates.

Therefore, analyzing the field equations (6.1) for micropolar fluid one makes the following assumptions:

1) the y-component of the velocity is negligible in comparison with the x-component, 2) the terms of order $(h/a)^2$ in balance Eqs.(6.1) are neglected, 3) the inertia terms in Eqs. (6.1) are neglected.

So, denoting by $\mathbf{V} = (u, v, 0)$ the velocity, by $\boldsymbol{\nu} = (0, 0, \omega)$ the microrotation vector, and by p the pressure, the flow is described by the following system of equations, cf. [35],

$$\frac{\partial p}{\partial x} = \frac{1}{2}\,(2\,\mu_v + \kappa_v)\,\frac{\partial^2 u}{\partial y^2} + \kappa_v \frac{\partial \omega}{\partial y}, \tag{6.3}$$

$$\frac{\gamma_v \partial^2 \omega}{\partial y^2} - 2\kappa_v \omega - \kappa_v \frac{\partial u}{\partial y} = 0, \tag{6.4}$$

$$\frac{\partial p}{\partial y} = 0. \tag{6.5}$$

The boundary conditions are given by

$$u = 0, \quad v = -U, \quad \boldsymbol{\nu} = \mathbf{0}, \tag{6.6}$$

for $y = h$ and

$$u = 0, \quad v = u_1, \quad ,\boldsymbol{\nu} = \mathbf{0}, \tag{6.7}$$

for $y = 0$. Here u_1 denotes the velocity resulting from the porosity of the lower wall [42, 53].

SINHA *et al.* [53] applied the micropolar fluid theory to the study of human joints lubrication. These authors obtained a solution of Eqs. (6.3) – (6.7) for the hip joint represented by a sphere on a porous surface, which modelled the porous cartillage. The gap was assumed to be very thin so that the system of two squeezing parallel plates with the lower porous plate can be considered as an aproximation for the lubrication in the weight-bearing synovial joint under conditions when the area of contact is maximum such as in standing and jumping.

NIGAM *et al.* [43] considered a model of two parallel plates with reference to load-bearing human joints. The lower plate is porous and consists of three layers of different porosities. The results obtained in [43] and [53] concern the pressure, load capacity and time of approach and are functions of parameters L, N, defined by

$$N = \sqrt{\frac{\kappa_v}{2\mu_v + \kappa_v}}, \qquad L = \frac{c}{l}, \qquad l = \sqrt{\frac{\gamma_v}{4\mu_v}}. \tag{6.8}$$

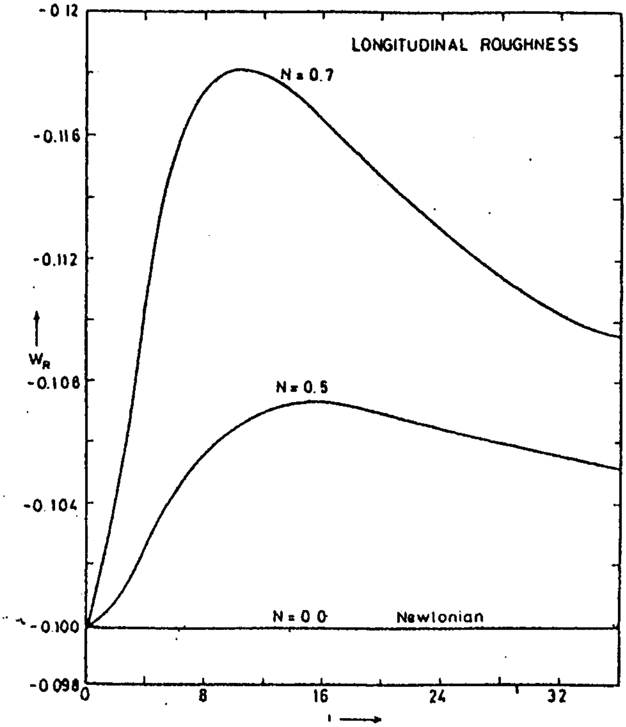
Here c denotes the characteristic height of the channel. The parameter L is the length parameter and N denotes the coupling number. In the classical case, for the Newtonian fluid where microrotations are absent, we have $N = 0$, $L = \infty$.

In addition to the usual dimensionless numbers which appear in the Newtonian theory, to identify the micropolarity of the fluid we need additional parameters to describe the characteristic material length of the microstructure and to characterise the coupling between viscosities κ_v and μ_v (both these viscosities characterise the couple-stresses.

In the present context the hyalouronic acid molecules are treated as microelements. The micromotions of those elements increase the viscosity coefficient κ_v and may be thought of as a measure of the resistance to the rotational motion, in the same way as μ_v characterizes the resistance to the linear motion. Consequently, the parameter N can be thought of as a property that depends on the concentration of the hyaluronic acid molecules. An increase in the concentration of HA molecules results in an increases of N. The parameter l has the dimension of length and may be thought of as a material parameter that depends on the size of the HA molecules. It was shown that for a diseased and elder joints, the HA molecules have lower molecular weight [12, 21]. The lower molecular weight corresponds to a smaller length of the molecule which, for constant c, yields larger values of *L*. In this sense, the influence of size and concentration of the hylauronic acid molecules on the functioning of synovial joints was investigated in [43] and [53] in the standing position. The results obtained indicate an increase in L which implies a decrease in the chain length of the hyaluronic acid molecule. This effect causes a decrease in the load capacity and response time. This may happen in rheumathoid joints. Similarly, a decrease in *N*, which signifies a decrease of the concentration of HA in the synovial fluid, also leads to a decrease in the load capacity and response time. An increase in the porosity results in a decrease in the load and response time. The response time is sufficiently small for high values of porosity, low values of *N* and high values of *L*. It was also shown that the viscosity near the solid boundary increases.

SINHA and SINGH [52] studied the influence of surface roughness on a technical bearing system. Theoretical results obtained in terms of *L* and *N* show that the values of the coefficient of friction are different for the longitudinal and transverse roughness, see Fig. 19.

(A)



(B)

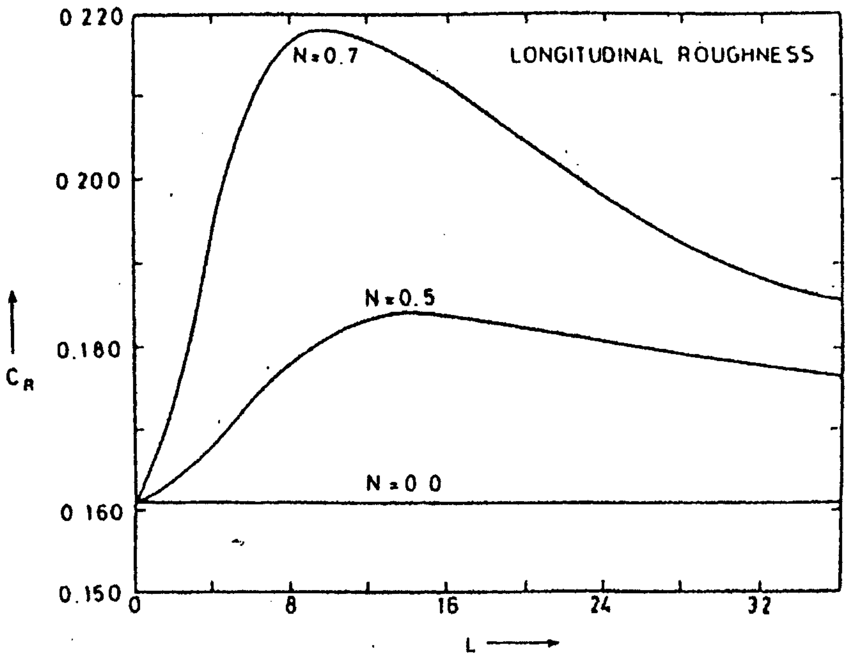


FIG. 19. Examples of tribological quantities: (A) – mean load ratio parameter W_r, and (B) – mean coefficient of friction parameter C_R, in bearing system as functions of parameters L and N, after SINHA *et al.* [52].

Recent experimental data due to MURAKAMI *et al.* [41] confirmed that the HA molecules primarily control the viscous property of the lubricant, cf. also [21]. Earlier, NEGAMI [42] showed that the viscosity of the synovial fluid varies almost linearly with the concentration. DOWSON [12] presented a formula describing the effect of the concentration of HA on the viscosity in the case of low shear rate. The experimental results due to MURAKAMI *et al.* [41] show excellent agreement with the viscosity measured by DOWSON [12], see Figs. 10 and 11. The effect of concentration of the hyaluronic acid on tribological quantities for both the natural and artificial knee joints was also experimentally investigated in [41], see Fig. 20. The results obtained show that the concentration of HA mainly controls the viscous property of the lubricant, and influence on the values of the coefficient of friction as well. It is also worth mentioning that exact values of the rheological coefficients for the synovial fluid, treated as a micropolar fluid, can be determined by performing experiments described in the book by MIGOUN and PROHORENKO [38]. Moreover, in the paper by KUCABA-PIĘTAL, [29], concerning a simplified analysis of the hip joint lubrication, the rheological coefficients of the micropolar fluid were estimated by exploiting the available experimental data concerning the microstructure of the synovial fluid.

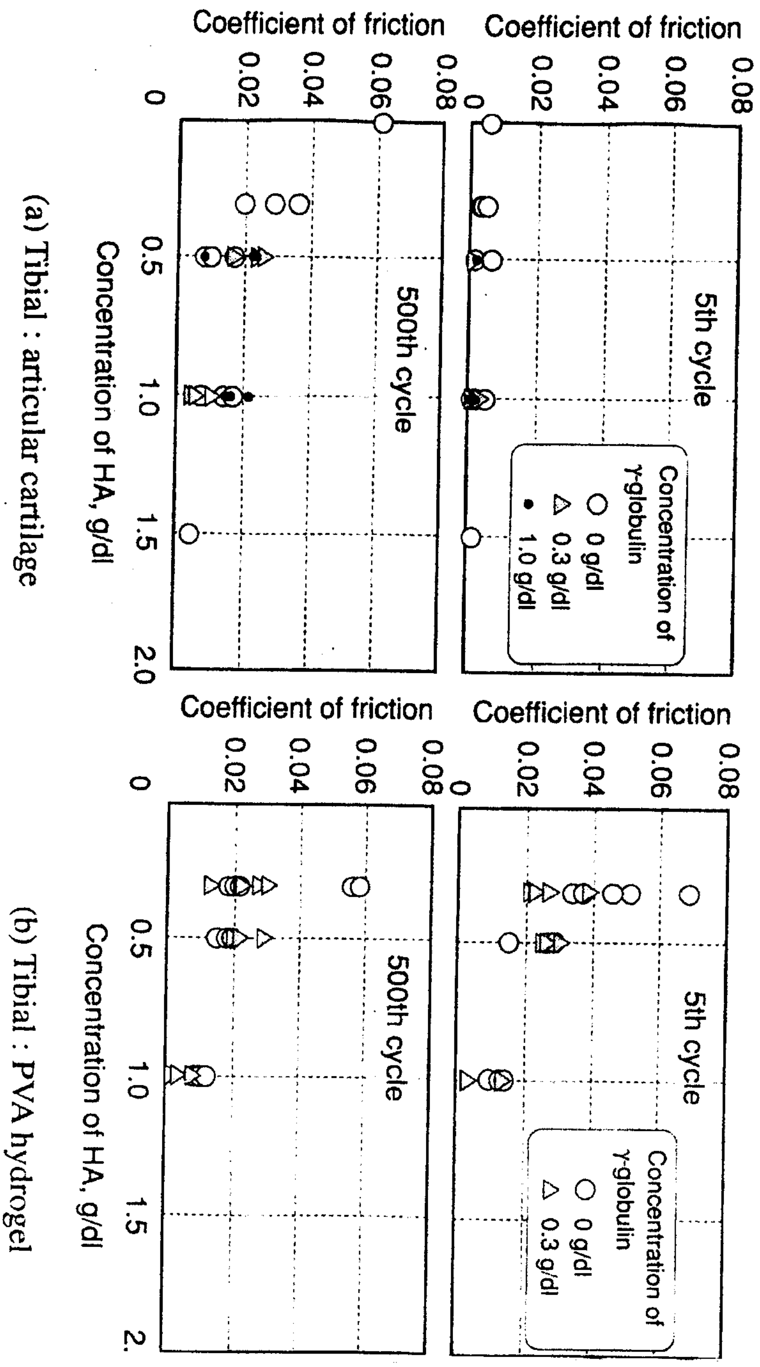


FIG. 20. Effect of HA concentration and γ-globulin addition on friction in knee joint in walking cycles, after MURAKAMI *et al.* [41].

7. CONCLUSIONS AND FINAL REMARKS

As we have seen, experimental data are available which suggest that joint lubrication is linked with liquid crystals theory. The following facts support this hypothesis:

1. The synovial fluid possesses liquid crystalline phases because it contains low-molecular cholesterics like cholesterol esters and high-molecular hyaluronic acid, which is a water-soluble polymer. Under suitable concentration, they can generate lyotropic liquid crystalline phase within the range of physiological temperatures. Collagen fibres in water, at appropriate concentration, form helical structures, see Fig. 4. Additionally, it is evident that there exists a similarity between the form of proteoglycan agregate in cartillage that is composed of aggrecans attached to the molecules of hyaluronian acid and typical side-chain polymers, see Figs. 1, 13.

2. Phase transitions take place in the synovial fluid. They are caused by various internal and external conditions. These transitions, for instance the inter-mesophase transition, anisotropic-isotropic phase transition, affect the rheological properties of the synovial fluid, and consequently the tribological quantities of synovial joints.

3. The cartilage surface exhibits an anisotropic roughness which is generated by ordered collagen fibers. Microgrooves are oriented in the direction of main locomotion of the synovial joint. In a thin layer in the vicinity of cartilage, the molecules of synovium are planar oriented by cartilage microgroves. Additionally, they are oriented during the flow. The anisotropy of the synovial fluid viscosity leads to a decrease of the coefficient of friction.

4. The theory of micropolar fluids seems to be applicable to the description of the synovial fluid. By using this theory one can, for instance, generalize the Reynolds equations derived for Newtonian fluids. It seems that the description of the synovial fluid within the framework of isotropic micropolar fluid theory can be considered as the first approach to modelling of such a fluid. The phase transition occuring in synovia under the influence of such factors as pressure, temperature, etc., support such an approach.

Among open problems one should mention the determination of rheological coefficients of the synovial fluid treated as a crystaline liquid. The second open problem pertains to a developement of the theory of synovial fluid and description of the joint lubrication combined with functioning of cartilage.

ACKNOWLEDGMENT

The second author was supported by the State Committee for Scientific Research (KBN, Poland) through the grant No. 7 T07A 012 18. and the third author through the grant No. 8 T11F 017 18.

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Received September 12, 2000.