Biophysical Chemistry, 32 (1988) 239–245
Elsevier

BPC 01329

The friction constants of bundle-like particles in solution

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Received 24 May 1988
Revised manuscript received 14 August 1988
Accepted 10 October 1988

Friction constant; Bundle-like particle

In our previous experimental work the following was found: an I-Z-I particle, having a bundle-like macromolecule composed of many thin filaments of actin, had a much larger rotational friction constant than that of a single sarcomere having a more compact bundle-like structure composed of not only actin but also myosin filaments. In this paper, the reason for such a large friction constant of I-Z-I is elucidated by a simple theory (based on the Helmholtz principle in hydrodynamics) in terms of the solvent permeability through the particles. It is demonstrated that if the filaments on the bundle surface are composed of flexible segments, each of which has a local Brownian rotation, the solvent-flow along the surface is strongly depressed because of increased energy dissipation and the solvent permeates through the bundle. The large friction constant of an I-Z-I is suggested to be due to this effect. This situation is very different from that of a single sarcomere without flexible filaments on the surface.

1. Introduction

It is known that solvent permeability plays an important role for the transport properties of chain-like polymers in solution, such as viscosity and self-diffusion constant. The theoretical expression for the hydrodynamic friction constant of a polymer has been derived using the concept of solvent permeability through the polymer from the Kirkwood-Riseman theory [1,2] (K-R theory) based on Oseen's equation. In our previous studies [3,4], we obtained the experimental rotatory friction constants, D, of two sorts of bundle-like polymers of the same size, a single sarcomere and an I-Z-I particle, each composed of many thin filaments of 1-μm length, by tracing the rotatory Brownian motions with an optical microscope. The results showed that the rotatory diffusion constant of a single sarcomere was 1.3×10^{-2} (s^{-1}), while that of an I-Z-I particle was 5.5×10^{-4} (s^{-1}), as seen in table 1.

It is an essentially important point that the I-Z-I particle has a remarkably small diffusion constant D_I as compared with the diffusion constant D_s of a single sarcomere (25 times smaller than D_s) and also that D_s itself is three times smaller than the value predicted by Tirado's theory [5] valid for cylindrical polymers. It seems very difficult to explain these results simply in terms of the difference of the solvent penetrability through the bundle between both particles, since according to the K-R theory, the value of D_I is calculated to be almost the same as the value of D_s despite a low number density of filaments in I-Z-I. This results from the fact that the Oseen effect strongly depresses the solvent permeability through the bundle even in the case of a low density of the hydrodynamic resistance units (segments) in the polymer.

In our present study, to solve this disagreement with the experimental results, we have taken into consideration the fluctuation effect of filaments in

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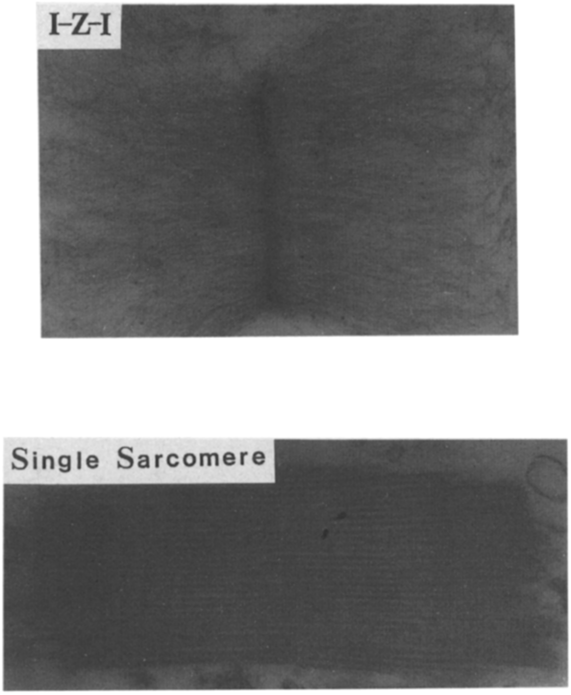

Fig. 1. Electron microscope pictures of the cross sections of I-Z-I and a single sarcomere.

the surface domain on an I-Z-I particle. Introduction of the concept of the local motion of the filaments on the bundle surface seems to be relevant in view of the situation of the filaments on I-Z-I as depicted in our recent electron microscope picture of I-Z-I (fig. 1). This effect is important if Brownian rotation of segments takes place, as the segment rotation forced by the shear gradient of solvent flow is generally accompanied by a large increase of the energy dissipation.

In this paper, to clarify the above experimental results, we derive a simple theoretical expression for the friction constant of a bundle on the basis of a hydrodynamical 'two-phase model', in which a bundle is assumed to be composed of two phases, the surface and the interior crystal phase, each having a different solvent velocity. It will be demonstrated that the rotational Brownian motion or fluctuation of the segments on the surface filaments of an I-Z-I particle depresses the surface solvent flow on the bundle and indirectly increases the permeability through the interior bundle crystal structure.

2. Materials and methods

A single sarcomere, which has a bundle-like form as mentioned in our previous paper, was extracted from muscle fibers according to Perry's method. A muscle fiber has two optical bands, referred to as the I-band and A-band. A single sarcomere is constructed by an A-band. Since the I-band is mechanically much weaker than the A-band, the muscle fibers can be easily cut into many pieces of single sarcomeres by stirring strongly in a homogenizer.

I-Z-I samples were obtained in the following way. After the treatment of muscle fibers with a high salt solution including ATP, crude I-Z-I was first precipitated by centrifugation and it was again dissolved into a fresh solution. By repeating this procedure, we obtained the final pure sample of I-Z-I.

The samples for electron microscope observation were prepared as follows. After washing the final sediments obtained by centrifugation, the particles contained in the sediments were prefixed by immersion in cold 3% glutaraldehyde buffer solution (pH 7.2), and the materials were finally fixed in solvent composed of cold 1% OsO_4 solution buffered at pH 7.2 by cacodylic acid. After a gradual dehydration with ethyl alcohol, the materials were embedded in Quetol 812 (Nisin EM Co.). Sections were made with an ultramicrotome and mounted on a carbon-coated collodion film, stained in 1% uranyl acetate and/or $Pb(NO_3)_2$. These sections were examined with a Hitachi JEM 100-C Electron Microscope.

3. Results

The data of the diffusion constants found in our previous paper [3] are again listed in table 1. An observed electron microscope picture of an I-Z-I particle is shown in fig. 1, together with a picture of a single sarcomere. As seen in these photos, the surface structure of I-Z-I seems to be

Table 1

The rotational diffusion constants obtained previously [3]

Samples	Experimental D (s^{-1})
I-Z-I [a]	$5.5 \pm 1.5 \times 10^{-4}$
Single sarcomere in relaxation solution	$1.3 \pm 0.5 \times 10^{-2}$

[a] Bundle diameter is 1 μm, and nearest filament-filament distance is 200–300 Å.

very flexible, while a whole single sarcomere looks very rigid.

4. Analyses

4.1. A simple expression for the total friction constant of a bundle-like particle

According to Ishihara's theory [7], there is a hydrodynamic relationship between the rotational diffusion constant D and the translational diffusion constant D_t of a chain-like polymer; that is, D_t is expressed by $D_t = \alpha L^2\ D$, where α is a numerical constant and L is the axial length of the polymer. In this paper we apply this relation to discuss the motion of bundle-like macromolecules by using D_t (in the direction perpendicular to the bundle axis).

In order to elucidate the essential part of the experimental results theoretically, it is not always necessary to construct and solve the complicated Navier-Stokes hydrodynamic equations. That is, for a brief discussion of the solvent permeability, it is sufficient only to introduce a rational hydrodynamical model for the macromolecule in question and to apply the 'minimum energy dissipation principle (the Helmholtz principle in hydrodynamics)' [8] to this model to determine the flow conditions. (This principle is applicable only to the stationary system of slowly flowing and incompressible fluids.)

In this study, we assume at first that there are two hydrodynamical phases in a bundle-like molecule, i.e. the surface phase and the interior phases, each having a different average velocity of solvent flow. By applying the above energy dissipation minimum principle to this 'two-phase' model, we can evaluate the solvent permeability in the interior phase and calculate the entire friction.

Now, let U and U' be the solvent velocities in x-direction (relative to the bundle) at places far from the bundle and just on the bundle surface, respectively, and let u be the averaged solvent velocity in the interior domain of the bundle (see fig. 2). Since the velocity gradient of the solvent is usually not so sharp except just near the boundary between the both domains, the velocity of inner flow u may be regarded to be a constant over the interior domain.

The total friction constant due to the inner filaments, denoted by ξ_k, can be calculated from Kuwabara's theory [9] in which the total friction exerted on the solvent flowing through a crystal structure composed of N parallel rods has been derived. This theory gives an expression for the friction constant per filament in the interior domain, ξ_k/N in the following form

$$\xi_k/N = 4\pi\eta_0 L_0 [\ln(\ell/a) - 0.75)]^{-1} \qquad (1)$$

where η_0 is the solvent viscosity, L_0 the length of a filament, ℓ the diameter of an area shared by each rod (or in other words, the distance between the rods), and a is the diameter of the filament, respectively. If the number of the interior filaments is sufficiently large, ξ_k may be thought to be proportional to N. The above formula predicts a value of ξ_k/N smaller than the Stokes' friction constant for an isolated rigid filament. The energy dissipation in the inner domain, W_{in}, is expressed by $W_{in} = \xi_k u^2$.

On the other hand, in order to evaluate the friction force acting on the surface domain, it is

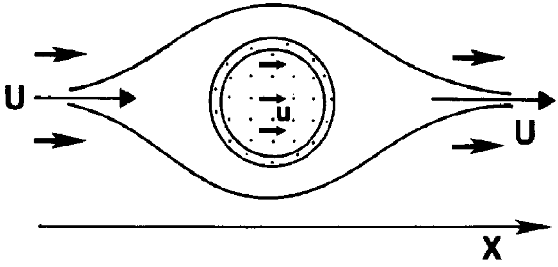

Fig. 2. A hydrodynamical two-phase model for a bundle-like particle.

convenient to classify the total frictions into the following three contributions.

The first one is the Stokes' force due to the disturbance of the flow on the bundle surface. It should be expressed in the form of $\xi_s(U-u)$ with the friction constant ξ_s given by the Stokes' law for a compact cylinder, since the relative velocity of the solvent flow $(U-u)$ causes the effective Stokes' force, or from another point of view, the flow has a slipping effect on the bundle surface with a slipping factor u/U. The energy dissipation due to this effect, W_1, is written by $W_1 = \xi_s(U-u)^2$.

The second one is the direct friction force in the flow direction acting on each filament in the surface domain against the surface flow, which may be expressed as $\xi_s'(U'+u)/2$ as an average in this region (ξ_s' represents a proportionality constant).

The third one, that is emphasized here, is an increase of the friction energy due to the local rotation of segments of the surface filaments in the domain of thickness δ forced by the shear gradient $(U'-u)/\delta$. The projection of this rotation exhibits a two-dimentional filament rotation on the plane orthogonal to the bundle-axis. The important fact is that this rotation is taking place against the two dimensional Brownian rotation of the filaments (see fig. 3) and it contributes strongly to the increase of energy dissipation, W_3, although it does not cause any additional friction force against the solvent on the average because it is laminar flow. This W_3 is written in the form, $W_3 = \xi_s^0(U'-u)^2$, in the case of a thin core, with a new constant ξ_s^0, which includes δ.

The above second effect on the surface friction, $\xi_s'(U+u)/2$, may be neglected, since the number of the filaments in the surface core is negligibly small as compared with the number of the inner filaments. We assume here for convenience that U' is close to U considering that Stokes' friction arises mainly in the vicinity of the inner surface of the bundle, although the Stokes' region itself spreads beyond the domain δ to the space whose width is the order of a. Under this assumption, U' may be put roughly equal to U and the total surface friction force, f, may be simply written as $f = \xi_s(U-u)$. The total friction force of the bundle, defined by $\Xi\, U$, can be expressed in the following form

$$\Xi U = \xi_s(U-u) + \xi_k u. \tag{2}$$

On the other hand, the total dissipative energy, W, given by $W = W_{in} + W_1 + W_3$, is expressed as

$$W = \xi_s^*(U-u)^2 + \xi_k u^2 \tag{3}$$

where

$$\xi_s^* = \xi_s + \xi_s^0. \tag{3'}$$

In eq. 3, the first term represents the energy dissipation in the surface domain including the third effect introduced above and arising under the shear rate, $(U-u)/\delta$.

Let us apply the 'energy dissipation minimum principle', e.g., the condition

$$\partial W/\partial u = 0$$

to eq. 3, we then obtain the following equation with respect to u/U

$$u/U = \xi_s^*/(\xi_s^* + \xi_k). \tag{4}$$

Substituting eq. 4 into eq. 2, we finally obtain the following simple expression for the total lateral friction constant

$$\Xi = \xi_k(\xi_s + \xi_s^*)/(\xi_s^* + \xi_k). \tag{5}$$

This Ξ may be simplified in three special cases as shown in the following.

4.2. *The special cases of Ξ*

(1) *When the surface friction is very large.* Since we may posit in eq. 5

$$\xi_s^* \gg \xi_k \tag{6}$$

the total friction of this bundle becomes

$$\Xi = \xi_k. \tag{7}$$

Fig. 3. A model of an actin filament. The filament has n pieces of segments, each of which may be thought to be an independent fluctuating unit.

As shown in the next section, the above situation, $\xi_s^* \gg \xi_k$ is realized and eq. 7 is established, if each segment is allowed to make a local Brownian rotation as mentioned above. In such a case, the total friction constant is given by the sum of the friction constants of all the inner filaments, ξ_k, shown in eq. 3. Eq. 7 is thought to apply in the case of I-Z-I.

(2) *Absence of flexible filament on the surface.* If there is no filament in the surface phase out of the bundle, ξ_s^* should be equal to ξ_s (or $\xi_s^0 = 0$), and ξ_k should be much larger than ξ_s (because of N filaments). Hence we have in this case the following relation for a compact bundle

$$\Xi = 2\xi_s. \tag{8}$$

This situation, $\xi_s^0 = 0$, obviously occurs in the case of a single sarcomere having a completely compact cylindrical form without flexible filaments on the surface.

The friction constant ξ_s for a compact cylinder having the length L and the diameter d can be calculated by Tirado's equation [5]

$$\xi_s = 4\pi\eta_0 L\left[\ln(L/d) + 0.839 + 0.185(d/L) + 0.233(d/L)^2\right]^{-1}.$$

From this equation we obtain the value $\xi_s/\Xi = 0.50$, which corresponds well to the experimental value for a single sarcomere indicated in table 1.

(3) *When all filaments are flexible in the whole domain.* In this case, since there is no surface domain, $\xi_s^0 = 0$, and ξ_k is expressed by the total segmental friction constant

$$\xi_k = N_b \xi_b = 6\pi\eta b N_b$$

where ξ_b is the friction constant of each (spherical) segment, b is the segment radius and N_b is the number of segments. If N_b is large, ξ_k becomes large and Ξ is given from eq. 5 by

$$\Xi = 2\xi_s. \tag{8'}$$

This result shows the tendency of the non-free-draining character of the bundle arising in the case of a large value of ξ_k.

4.3. Evaluation of ξ_s^0 in the case of the fluctuating surface filaments

It has been demonstrated by Happel and Brenner that if the thermal motion of a macromolecule is taken into consideration [11], the intrinsic viscosity $[\eta]$ of the macromolecule becomes apparently much larger than that predicted by simple hydrodynamic theories. For example, in the case of an isolated ellipsoidal macromolecule with axial ratio 50, the usual hydrodynamic calculation in the absence of Brownian motion gives for $[\eta]$ a value of 3.79. However, it increases to 178 in the presence of a full angle rotatory Brownian motion. The evaluation made use of the following equations of Kuhn's theory [12]

$$[\eta] = 2.5 + 0.4075(p-1)^{1.508}, \; (1 < p < 15)$$

$$[\eta] = 1.6 + p^2\left[(3(\ln 2p - 1.5))^{-1} + (\ln 2p - 0.5)^{-1}\right]/5, \; (p > 15) \tag{9}$$

where p is the axial ratio.

In the case of the I-Z-I particle, this friction increase, ξ_s^0, is brought about by the Brownian rotation of the segments of the surface filaments, which gives a disorientation effect on the segment rotation made by a velocity gradient of solvent flow existing in the surface domain. Since the surface filaments are assumed to be flexible, as observed in the electron microscope pictures, it is possible that this effect justifies eq. 6.

Let us evaluate the concrete value of ξ_s^0 to examine whether eq. 6 is established or not. For this purpose the Debye theory [10] may be applied. The energy dissipation, W_3, due to the segmental rotation in the surface domain is expressed in two ways, a microscopical and a macroscopical way, which must give the same values. We thus have the following equation

$$W_3 = \Delta\eta G^2 S\delta = \xi_s^0 (U-u)^2 \tag{10}$$

where S is the area of the bundle envelope, δ the thickness of the surface domain, $\Delta\eta$ the viscosity increment due to the existence of the partial rotation of the segments in each filament in the surface domain and G the velocity gradient existing between the outer and inner surfaces of that domain, respectively. Since $G = (U - u)/\delta$, we have the following expression for $\xi_{\rm s}^0$

$$\xi_{\rm s}^0 = (S/\delta)\Delta\eta. \tag{11}$$

Note that $\Delta\eta$ may be written as $\Delta\eta = [\eta]\eta_0\phi$, with $[\eta]$ the viscosity increment per filament and ϕ the volume fraction of the filaments.

If one assumes that a single filament is composed of n segments, each of which has a freedom of rotation within a certain solid angle (Ω), $\xi_{\rm s}^0$ in eq. 11 may be put into the following form for I-Z-I particle

$$\xi_{\rm s}^0 = 6\pi \times 10^{-6}\Omega n[\eta]. \tag{12}$$

Here δ and ϕ have been put equal to 20 nm and 0.030, respectively, and S and η_0 to 4π μm^2, and 0.010 poise, respectively. Using for $[\eta]$ the values given by eq. 9 and for a filament in the surface domain (assumed to be an ellipsoid) $p \gg 1$, we obtain numerical values of $\Delta\eta$ larger than unity, as seen in table 2.

Table 2 indicates the calculated numerical values of $\xi_{\rm s}^0$ for limited segmental Brownian motions of the segments at various n and Ω. The conditions where $\xi_{\rm s}^0 > \xi_{\rm k}$ are indicated with bold lines in table 2. From this result, we may conclude that the relation $\xi_{\rm s}^0 > 7 \times 10^{-3}$ (g/s) is realistic. Therefore, the condition $\xi_{\rm s}^0 \geq \xi_{\rm k}$ in eq. 8 is definitely satisfied, since $\xi_{\rm k}$ is $\overline{\xi_{\rm k}} = 6.85 \times 10^{-3}$ (g/s) for I-Z-I [4].

Table 2

The values of friction constants, $\xi_{\rm s}^0$, in the surface layer due to the segment fluctuation, calculated on the basis of Debye's theory [10] and Kuhn's theory [12]

p, the axial ratio of a segment, which is a fluctuating unit of a filament; n, the number of segments in a filament, whose segments are freely movable within a certain solid angle (Ω). $\xi_{\rm k} = 6.85 \times 10^{-3}$ (g/s).

	p	800	400	200	100	50	20	10
	n	1	2	4	8	16	40	80
	[η]	25869	7233	2052	594.0	177.0	38.87	13.70
	$\Delta\eta$ (p.)	7.76	2.17	0.616	0.178	0.053	0.012	0.004
ξ_s^0 $\times 10^{-3}$ (g /sec)	Ω ×1	488	273	155	89.6	53.4	29.3	20.7
	×1/4	122	68.2	38.7	22.4	13.4	7.33	5.17
	×1/10	48.8	27.3	15.5	8.96	5.34	2.93	2.07
	×1/30	16.3	9.09	5.16	2.99	1.78	0.98	0.69

5. Discussion

As shown in the above section, the remarkably large friction constant of an I-Z-I particle found in our previous experiments may well be elucidated by the above simple theory. The essential point found in this study is that if the segments (hydrodynamic resistance units) on a filament are allowed to have a local Brownian rotation in the surface domain, the apparent friction constant of the filaments, $\xi_{\rm s}^*$, therein is largely amplified. This effect depresses the solvent flow on the surface and results in a full permeability for the solvent flow inside the bundle to minimize the energy dissipation. In this theory, an independent rotation of each segment has been assumed, which is not unreasonable, since adjacent segments are known to be connected across a bond with a short molecule.

The above theory is not a strict one and is derived particularly to elucidate the dominant natures of the friction constant of bundle-like molecules based on a two phase model. However, we believe that this study has pointed out some important aspects of the hydrodynamic behavior of a bundle-like polymer, such as a free-draining character in the presence of flexible filaments on the surface domain and a non-draining character appearing in the case of a set of all rigid rods or a set of all flexible filaments. Although the above two-phase model seems to be too much simplified, it may explain the hydrodynamic feature of macromolecular rotation if used in combination with the 'minimum energy dissipation principle'.

To examine the validity of eq. 5 in the above simple theory, we have performed a sedimentation experiment [6] with a macroscopic bundle made of many parallel thin glass rods, in which the sedimentation velocity was measured in a highly viscous solvent of sufficiently small Reynolds' number. A part of the results is shown in fig. 4. One of the most important findings in this experiment is that the overall hydrodynamic resistance,

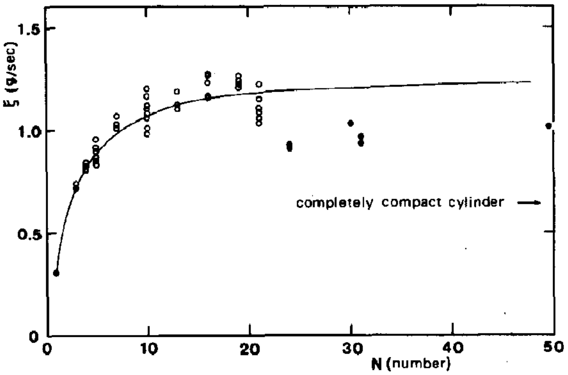

Fig. 4. Friction coefficient, ξ, of a macroscopical bundle-like body (composed of parallel glass rods) measured by a sedimentation experiment as a function of the number of the rods, N (bundle diameter, 3 cm; and rod diameter, 1.2 mm). The open circles (○) and solid circles (●) which represent stable values indicate the experimental ξ values (reduced to the values in water at 20 ° C). The solid curve represents the change of the function coefficient according to our theory [6].

ξ, increases sharply at the initial stage of increasing number density, σ, of rods in a bundle at a fixed outer radius. This tendency has also been demonstrated by Bloomfield et al. for a shell model [13]. But ξ obviously shows after a maximum a decreasing tendency at large N. This suggests that the above two phase model breaks down after the maximum ξ point at large N as a result of a change in the hydrodynamical intra structure situation of the bundle where u becomes much smaller than the value expressed by $u = 0(1/\xi_k)$ in eq. 8 with respect to the friction constant, ξ_k, in the inside phase. However, this situation has not been clarified theoretically as yet. A more strict theoretical treatment is required for a more quantitative analysis of the friction constant of bundle-like macromolecules.

Acknowledgments

The author would like to express his thanks to Prof. N. Imai who has given much valuable advice on this work. He also wishes to express his gratitude to Dr. S. Higashi-Fujime for her guidance about the techniques to obtain ultrathin sections for electron microscope observation. The author is thankful likewise to Dr. S. Matsuura who kindly presented to him the valuable data of the sedimentation experiments on model samples.

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