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# DNA Dynamics under Turbulent Flow

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**Summary:** Using high molecular weight DNAs (λ-and calf-thymus (CT) DNAs), we investigated their flow characteristics under turbulence in a rotating disk geometry. By putting emphasis on the effect of CT-DNA concentration, its turbulent drag reduction (DR) characteristics were compared with that of λ-DNA based on both DR efficiency and mechanical degradation under turbulence. Addition of spermidine into the turbulent flow as a condensing agent showed abrupt change of the DR efficiency of λ-DNA in a turbulent flow.

**Keywords:** coil-glouble transition; DNA; mechanical degradation; turbulent drag reduction

## Introduction

Dynamical behavior of polymers in both laminar and turbulent flows has been widely investigated not only due to its wide range of engineering applications but also due to its scientific interests,[1,2] One of such interesting behaviors of the polymer chains including DNA observed in nonhomogeneous laminar flow fields is called ''polymer migration'' and it results in a spatially nonuniform concentration profile.[3,4] Concurrently, it is well known that flexible polymers in extremely dilute solution can reduce turbulent losses significantly. This very striking phenomenon is known to be ''turbulent drag reduction (DR)'', in which turbulent drag of flowing medium is drastically reduced by even minute amounts of suitable additives.[5] Despite its extensive studies, a satisfactory explanation of DR still eludes its fundamental and general interpretation[6] due to a coupled mechanism of both turbulence and polymer dynamics. Several intrinsic structural characteristics are playing crucial roles in controlling and evaluating the fundamental features of DR phenomena. Both water-soluble and oil-soluble polymers have been investigated as DR additives. Recently, with respect to the structural effect on the DR, monodispersed high molecular weight λ-DNA[7] has been adopted for the DR study. Double helix structure of the DNA in turbulent flow showed strong resistance to turbulent flow and mechanical molecular degradation.

Among various DANs, we selected both λ-DNA and CT-DNA for our DR studies, in which the CT-DNA possesses polydispersity and higher molecular weight compared with monodisperse λ-DNA. Since DNAs have a unique helical molecular structure compared to conventional flexible polymeric drag reducers, we could expect this characteristic will make it possible to show more detailed understanding in the DR experiment and analysis.[8] In addition, DNA chains exhibit a highly elongated coiled state in a flow field of an aqueous buffer solution in the absence of condensation agents. This structural feature of single and long duplex DNAs exhibits a large discrete transition between elongated coil and compact globule states in some specific condition.[9–11]

Recently, Takahashi *et al.*[9] reported that individual DNA molecules undergo a first-order transition between an elongated coil state and a compacted globular state with the addition of various kinds of condensing agents, such as neutral flexible

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polymer,[12] cationic and neutral surfactant,[13] alcohol,[14] polyamine spermidine[15] and inorganic metal cation.[16] Nonetheless, the conformation change of DNA in bulk solution under a flow field has not been well studied.

In this study, we chose high molecular weight DNAs, and then examined both DR efficiency and coil-glouble transition under a turbulent flow, in which this transition can be observed indirectly via DR efficiency. From DR phenomena of CT-DNA as a function of pH, Hand and Williams[8] observed that the less flexible helical conformation is preferable to the random coil for maximum drag reduction. Recently, we[7,17] have reported that double-stranded DNA is found as a good turbulent drag reducer when compared with linear flexible polymers such as poly(ethylene oxide) and poly(acryl amide). Furthermore, it is natural to conjecture the result of DNA compaction[18] by condensing agent in turbulent flow, resulting in deterioration of DR efficiency. However, it was not easy to find the experimental evidence and quantitative analysis with respect to the correlation of conformational change and drag reduction, and the role of condensing agent in turbulent flow.

## Experimental Part

Concentrated stock solution of the DNA, both monodispersed λ-DNA[7] (Promega Corporation, USA with 48.5kbp (32,300 kD) and polydispersed calf-thymus DNA (CT-DNA, ~75.7kbp (~50,000kD), Sigma, USA), was separately injected into a buffer solution in rotating disk apparatus (RDA, dimension of disk: 140 mm diameter × 3 mm thickness) as shown in Figure 1.[19] The fluid container consists of an aluminum cylinder, whose dimensions are 170 mm diameter × 25 mm wall thickness, and a removable 30 mm thick aluminum lid to seal the solution. The torque required to rotate the disk for pure solvent ($T_S$) at a given speed was measured first. The percent DR (%DR) was then calculated

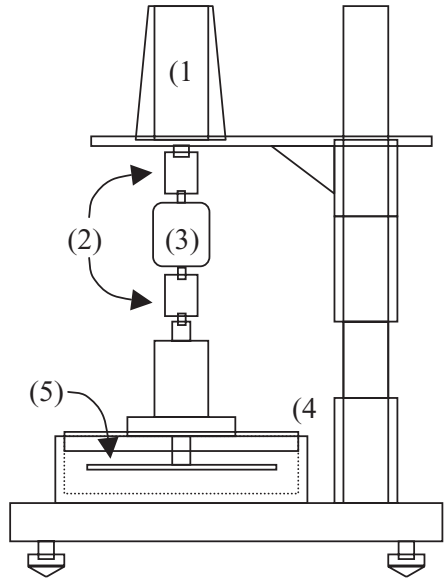


***Figure 1.***
(1) Motor (2) Flexible couple ring (3) Torque sensor (4) Fluid container (5) Rotating disk.

by measuring the corresponding torque required for a dilute polymer solution ($T_P$) at the same $\omega$ as:

$$\%DR = \left(\frac{T_s - T_p}{T_s}\right) \times 100\,(\text{at a given } N_{Re} \text{ number}). \quad (1)$$

Note that $N_{Re} \equiv \rho r^2 \omega/\mu$ is based on the rotational speed of RDA,[20,21] in which $\rho$ is the fluid density, $\mu$ is the fluid viscosity, and r is the radius of the disk. The %DR was then obtained as a function of time. The temperature of the system was maintained at 25 °C.

At first, the RDA reservoir was filled with the buffer solution (10 mM tris-HCl, 10 mM NaCl and 1mM EDTA) for DNA. The DNA was injected into the turbulent flow field for the DR measurement. The %DR was then obtained as a function of time by injecting measured quantities of stock solution directly into the turbulent flow field generated by the RDA.

For coil-globule transition, polyamine (Spermidine (SPD), $C_7H_{19}N_3 \cdot 3HCl$,

 

Sigma, USA) was introduced into solution into flowing medium containing DNA.

## Results and Discussion

Figure 2 shows %DR of the CT-DNA for two different concentrations as a function of time at relatively high Reynolds number ($N_{Re}$~1,000,000; 1,980 rpm). Although CT-DNA degraded initially, the overall DR efficiency was maintained for an hour. However, when compared to λ-DNA, CT-DNA did not seem to show any measurable DR effect at a very low DNA concentration with similar conditions in our previous experiment of λ-DNA (1.35 wppm and 1980 rpm) as shown in the inset, despite the higher molecular weight of the CT-DNA. The difference can be explained based on polydisperse molecular weight characteristics of the CT-DNA.[22]

Based on the higher molecular weight of CT-DNA and the analysis of λ-DNA under turbulent flow condition,[7] it can be assumed that major mechanical degradations of the CT-DNA intensively occurred right after the injection of CT-DNA into turbulent flow just in a few second. As far as this half-cut phenomenon is concerned, Sasaki *et al.*[23] explained that molecular degradation in an elongational flow field proceeds as a two-stage process; molecular stretching and fracture. In this process, scission of the extended conformation always occurs near the midpoint, where the stress in the molecule reaches a maximum value.[24] Regarding the critical condition of molecular degradation, they[23] also explained that the critical strain rate ($\dot{\varepsilon}_f$) for fracture is related to the contour length (L) and molecular weight ($M_w$) of a polymer chain in an inverse square mode as:

$$\dot{\varepsilon}_f \propto \frac{1}{L^2} = \frac{1}{M_w^2} \tag{2}$$

This indicates that a polymer chain including DNA chain, after its first mechanical degradation, will need a much higher strain rate for its second generation scission. In a sufficiently strong turbulent flow, multi-generation degradation process can occur.[25] Nonetheless, this model strongly supports the single step half-cut degradation process of both CT-DNA and λ-DNA in this study, if the critical strain rate for

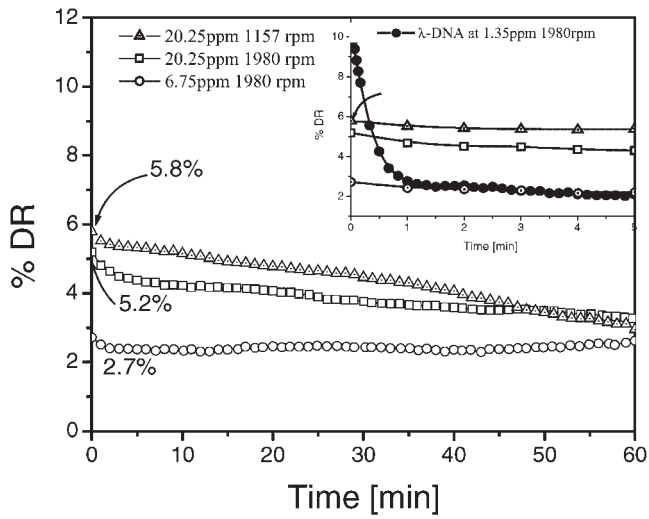


***Figure 2.***
DR efficiency of CT-DNA for different concentration and RPM. The inset shows the comparison of initial drag-reducing efficiency of CT-DNA with λ-DNA.

 

half-cut of both CT-DNA and λ-DNA is much higher than the flow field.

In addition, we performed another experiment at lower rpm (1,157 rpm, $N_{Re} = 7 \times 10^5$) with same DNA concentration (20.25 ppm) to identify the nature of flow condition at 1,980 rpm. As a result, we obtained slightly higher %DR value (5.8% in Figure 2) contrary to the initial speculation (lower DR efficiency at low rpm). This indicates that the experimental condition at 1,980 rpm was already above the critical point. On the other hand, the %DR saturation values (asymptote values) of different flow conditions are almost the same with each other. Accordingly, this trend of the same saturation value indicates that the minor degradation below the critical rotation speed, from which apparent degradation starts, was not a function of rotation speed but rather a function of time, which is distinct from the drag reducing behavior of linear polymers.

Figure 3 represents the DR efficiency as a function of time for five different rotation speeds, representing overall trends of similar and distinct decrease of DR efficiency.[17] A numerical fitting was attempted using the simple first order degradation model. If we consider the characteristic degradation of λ-DNA in turbulent flow as mid-point scission, the degradation can be expressed as a function of molecular length scale (L) and half-length (L/2), which is the size of degraded λ-DNA. Namely, we can describe the degradation of DNA, in which the length of λ-DNA (L) goes to L/2 as a first order reaction with $\alpha$ being the rate. Using the number of λ-DNA with length L at time t, $N_L(t)$ and the mass balance of λ-DNA molecules, Lim *et al.*[7] derived the DR efficiency as a function of time as given in Eq. (3). So the degradation features in different rotation speeds can be fitted and numerically expressed by the coefficients in the Eq. (3) in first order approximation where w(L) is the drag reducing power dependence on L and c is some background contribution due to systematic uncertainties in the measurement.

$$\begin{aligned} \%DR(t) &= w(L)N_L(t) + w(L/2)N_{L/2}(t) + c \\ &= w_1 e^{-\alpha t} + 2w_2(1 - e^{-\alpha t}) + c \end{aligned} \quad (3)$$

Using this equation, the physical interpretation of DR and mechanical molecular degradation can be described with two time

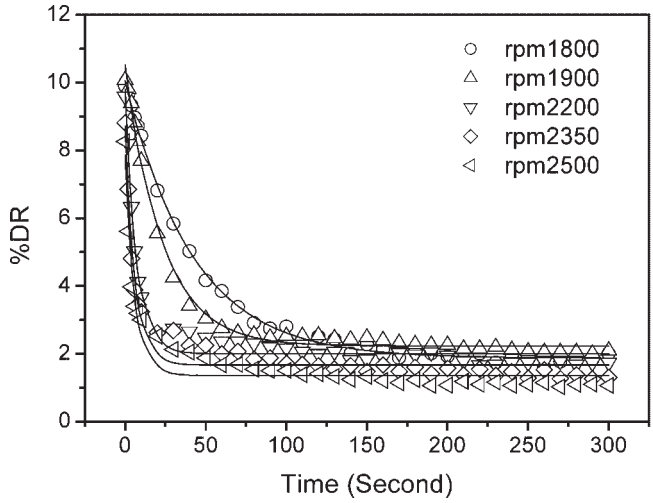


***Figure 3.***
DR efficiency as a function of time for 1.35 wppm of λ-DNA at different RPM. Solid line represents values obtained from Eq. (3).

 

independent parameters, $\alpha$ and w. The solid lines in Figure 3 for five different rotation speeds are obtained from non-linear curve fitting for $\alpha$ and w using the simple first order degradation model. Ideally, the fitting ratio w(L)/w(L/2) should be the same at a fixed concentration (here, 1.35 ppm).

It can be also noted that λ-DNA always displayed nearly the same limiting value, indicating possible evidence of a mid-point degradation of the λ-DNA molecule. The residual maintenance of the drag reducing efficiency was thought to originate from the strong endurance of the λ-DNA molecule in turbulence, indicating that even though the λ-DNA molecules underwent a degradation process in high turbulence, residual short chain molecules that were not degraded under such conditions remained as a drag reducer. The molecular weight change of λ-DNA induced from the DR experiment was confirmed using a very simple but definitive way, electrophoresis method, supporting the previously suggested mid-point molecular scission.

Figure 4 shows the %DR for 1.35 wppm of λ-DNA as a function of time at 1157 rpm (Reynolds number, $N_{Re} \sim 590{,}000$) with and without SPD. To identify the role of SPD in turbulent drag reduction induced by DNA, we differentiated SPD mixing procedure and compared the results with that of simple λ-DNA drag reducing phenomena. As far as the drag reduction of λ-DNA in buffer solution is concerned[5], relatively high drag reducing efficiency even at low DNA concentration (1.35 wppm) was observed and then maintained for an hour with minor degradation of λ-DNA (about 1%) in turbulent flow of this specific Reynolds number. This kind of strong resistance of double strand λ-DNA to turbulent flow was already investigated and the details were explained in our previous work.[7] Briefly speaking, this resistant and residual drag reducing efficiency is originated from the residual half cut DNA in turbulent flow. The resultant asymptote DR efficiency explains the origin of those changes, which can be conclusively verified via the electrophoresis experiment.

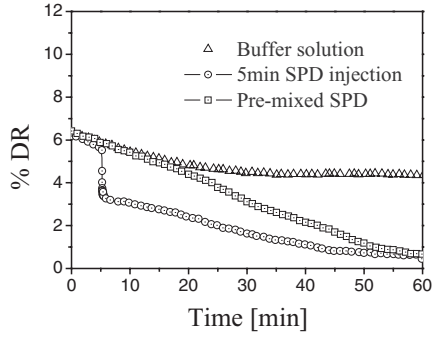


***Figure 4.***
DR versus time for 1.35 wppm λ-DNA at 1,157 rpm with and without SPD.

As a condensing agent, SPD in distilled water (10.615 μM) was prepared to be mixed before and after the injection of λ-DNA into the flow. The dot centered square and circle in Figure 4 show the effects of SPD injection on the DR efficiency change of λ-DNA. The open circle represented the abrupt change of DR efficiency by an addition of SPD after 5 min of λ-DNA injection.[26] SPD altered the conformation of λ-DNA through the complexation of ionic structure. In the case of pre-mixed SPD system, we could find nearly the same initial %DR with buffer solution system (~6.2%). Soon after the complexation started, the progressive decrease of DR effect was found, and finally reached to nearly same saturation %DR value of 5 min-injection case. More detailed understanding of the mechanism for three different methods is under investigation.

On the other hand, there might be one another possible reason, which can explain the gradual slow decrease of DR for DNA-SPD complexes. Although, the extremely strong turbulent flow gave small room for considering the precipitation or aggregation of DNA-SPD complexes in turbulent flow, our extremely severe experimental condition of SPD concentration could give its way to inclusion for the possibility of precipitation or aggregation[27] different from the results of Yoshikawa *et al.*[28].

 

## Conclusions

From both DR efficiency and a mechanical degradation under turbulence, DNA chains having much higher molecular size than that of λ-DNA were observed to be more susceptible to mechanical degradation in a turbulent flow. Furthermore, by adopting the coil-globule transition concept of DNA to turbulent drag reduction phenomena, we can evidently and dramatically examine the structural effect on the drag reduction. The addition of SPD into turbulent flow as a condensing agent of the λ-DNA showed the sudden decrease of drag reducing efficiency.

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