Turbulent Drag Reduction and Degradation of DNA

H. J. Choi,[1] S. T. Lim,[1] Pik-Yin Lai,[2,3] and C. K. Chan[3,*]

[1]*Department of Polymer Science and Engineering, Inha University, Incheon, 402-751, Korea*
[2]*Department of Physics and Center for Complex Systems, National Central University, Chungli , Taiwan 320, Republic of China*
[3]*Institute of Physics, Academia Sinica, Nankang, Taipei, Taiwan 115, Republic of China*

(Received 15 November 2001; published 2 August 2002)

Turbulent drag reduction induced by λ-DNA is studied. The double-stranded DNA is found to be a good drag reducer when compared with the other normal linear polymers. However, this drag reducing power disappears when the DNA denatures to form two single-strand molecules. Mechanical degradation of DNA is also different from that of the normal linear-chain polymers: DNA is always cut in half by the turbulence. Our results suggest that the mechanism for turbulent degradation of DNA is different from that of the normal flexible long-chain polymers.

DOI: 10.1103/PhysRevLett.89.088302 PACS numbers: 83.60.Yz, 47.27.Gs, 87.15.–v

It is well known that the drag in a turbulent flow can be drastically reduced [1] momentarily by the addition of a minute amount of suitable polymers. In general, the most effective drag-reduction (DR) polymers are found to possess a linear flexible structure and a high molecular weight. These long-chain polymers will be broken by the turbulence and DR will be degraded soon after the polymers are added. It is found that this mechanical molecular degradation (MMD) and DR efficiencies are correlated [2]. Usually, poor drag reducers will be degraded more rapidly by the flow. Although both MMD and DR properties of polymers have been known for a long time and are vital for real applications, the basic mechanism for both of them still eludes explanations [3]. Presumably, there are nontrivial interactions of the flow and the polymers at the molecular level which produce both MMD and DR. An understanding of MMD and DR requires the investigation not only of the phenomena associated with turbulence itself but also of the nonlinear interaction between flows and the polymers which has been shown in experiments to be of fundamental importance [4].

Although the interaction between turbulence and polymers cannot be studied directly at the molecular level yet, careful measurements have been carried out to study the polymer dynamics in simple flow configurations. These experiments found that polymers can be stretched by weak flows [5] and broken by strong extensional flows [6]. Since there are strong correlations [7] between drag-reduction efficiency and extensional viscosity in polymer solutions, a study of MMD will provide a good opportunity not only in the understanding of the basic DR mechanism but also in the turbulent structures at such a small scale in the presence of polymers. Ideally, data from these MMD experiments can be used to construct degradation dynamics with consistent turbulence models. In this Letter, we report results of DR efficiency and MMD measurement with λ-DNA molecules in a rotating disk apparatus (RDA). DNA molecules are chosen for two reasons. First, the length of a fully stretched DNA can be comparable to the microscales of the turbulent flow. Since the length of the polymers traditionally used in DR studies are much smaller than the microscales of the turbulence, the interactions between the polymer and the flow might be quite different. Second, a large change in molecular properties of DNA can be induced by a change in *p*H or temperature. At the appropriate *p*H or temperature, a DNA molecule will change from its double-stranded (ds) natural state to its denatured state of two single-strand (ss) molecules. This configurational change might help to probe the details of the DR phenomenon.

Hand and Williams [8] reported in a brief note that the natural state of DNA is preferable to the denatured state for maximum drag reduction in an experiment using calf-thymus DNA with *p*H control. But no explanation was given. In our experiments, we find that the same phenomena can be observed in our system, and this change in DR behavior of the DNA can be attributed to the change in mechanical properties of the two different states of DNA. It is found that the drag reducing power of the molecule is controlled mainly by the stiffness of the molecule. Different from other linear polymers, our MMD results indicate that dsDNA molecules are always cut exactly in half by the turbulence even when the microscale of the turbulence is smaller than the size of the DNA. This simple degradation suggests that the dynamics of degradation of dsDNA is different from those of normal linear chain polymers. It is possible that the difference is due to the long persistence length of the double helix. Furthermore, the simple degradation and monodispersity of DNA allow the construction of a simple degradation model. From this model, various physical parameters not easily accessible or reported before can be estimated.

A RDA [9] is used to study both the DR efficiency and the transient behavior of MMD of DNA chains in a turbulent flow. The device consists of an aluminum [10] disk with dimensions of 14.5 cm in diameter and 0.32 cm in thickness, enclosed in a cylindrical, temperature-controlled container composed of aluminum with inner diameter

16.3 cm and height 5.5 cm. An electric transducer was used to monitor the torques on the rotating disk. Turbulence is produced for rotational Reynolds number $\mathrm{Re} > 3 \times 10^5$ or, equivalently, rotational speed of the disk $(\omega) >$ 570 rpm [11]. Here $\mathrm{Re} = \rho r^2 \omega/\eta$, where r is the radius of the disk, and ρ and η are the fluid density and viscosity, respectively. The temperature of the system is maintained at $25 \pm 0.5\,^{\circ}\mathrm{C}$. The DR percentage (%DR), defined as $\%\mathrm{DR} = (T_S - T_P)/\mathrm{T}_S \times 100$, is measured, where T_P and T_S are the torques needed to maintain the disk to rotate at a particular Re with and without polymer, respectively.

DNA sample (λ-DNA) with 48 502 bp in size was used. Drag reduction was performed in a buffer solution (10 mM tris-HCL, 10 mM NaCl, and 1 mM EDTA, $p\mathrm{H} = 7.8$) for λ-DNA. The buffer solution is chosen as the flow medium to preserve the ds helical structure of the DNA. It is well known that dsDNA will decompose into ssDNA in deionized water. The RDA reservoir was first filled with 370 ml of buffer solution and %DR was then obtained as a function of time by injecting measured quantities of stock solution (0.515 mg/ml DNA) directly into the turbulent flow field generated by the RDA. The mixing time of the polymer with the flow can be estimated as the turnover time of the largest eddies which is always less than the 0.2 s injection time. Since all the data reported below are sampled in the time scale of a few seconds, the mixing of polymers with the flow in these experiments can be considered as instantaneous. A high molecular weight (M_w) water-soluble poly(ethylene oxide) (PEO) was also selected for the comparison of λ-DNA drag reducing behavior; namely, $M_w = 5 \times 10^6$.

Figure 1 shows the %DR for 2.70 and 1.35 wppm DNA as a function of time at relatively high Reynolds number ($\mathrm{Re} \approx 1.2 \times 10^6$). As shown in this figure, the λ-DNA showed a relatively high drag-reduction effect that increases with concentration. However, the measured %DR decreases with time due to the degradation of DNA. A remarkable feature in Fig. 1 is that although λ-DNA degraded in high turbulent flow, the overall %DR efficiency was maintained for a long time. This long time characteristic behavior is the main difference from other kinds of drag reducing linear long-chain molecules. The inset of Fig. 1 shows the the main difference in %DR of the λ-DNA compared with that of PEO at short time scales. The slopes of the drag reduction of DNA for 1.35 and 2.70 wppm were almost the same, and they were less steep than that of PEO. Note that the reported %DR are relatively low because we added a small amount of DNA and DR from rotating disk flow is always smaller than that from pipe flows.

To study the degradation of λ-DNA, the length of the DNA is measured by the electrophoresis method. Samples from high turbulent flow drag-reduction ($\mathrm{Re} \approx 1.2 \times 10^6$) experiments were concentrated to perform the electrophoresis [12]. These measurements show that, instead of a wide range of length distribution, there is only one length (in units of base pair, bp) of the DNA left after the degradation which is 23.1 kbp [13]. Since the original length of

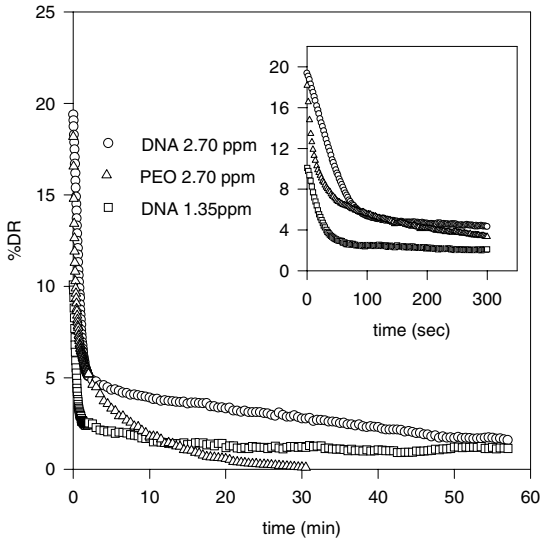

FIG. 1. Time dependence of drag-reduction percentage for 1.35 and 2.70 wppm λ-DNA in buffer solution compared with PEO ($M_w = 5 \times 10^6$) at 1980 rpm ($\mathrm{Re} = 1.2 \times 10^6$). Inset shows the same data at early times.

the DNA is 48 kbp, this result supports the previous theory that the overall degradation of polymer under stretch mainly occurred in the half position of the polymer. Furthermore, it can be seen from Fig. 1 that the drag reducing power of DNA with length L (at $t = 0$ min) is higher than that of $L/2$ (for $t > 50$ min). In order to test if the $L/2$ length DNA will degrade into even smaller fragments, a degradation experiment lasting 8 h has also been performed. The result of this long duration experiment shows that no more degradation after the DNA has been cut into half at this Reynolds number.

If one thinks of the degradation in which L goes to $L/2$ as a first order reaction with α being the rate, the number of DNA with length L at time t, $N_L(t)$, can then be expressed as $dN_L/dt = -\alpha N_L$ and $N_{L/2}(t)$ will increase as $N_{L/2}(t)/2 + N_L(t) = N_L(0)$. Since %DR is proportional to polymer concentration for dilute solution, data in Fig. 1 can be fitted to the form $\%\mathrm{DR}(t) = w(L)N_L(t) + w(L/2)N_{L/2}(t) + c$ 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. In this simple model, all the physics of DR and MMD is contained in the two time independent parameters, α and w. The solid lines in Fig. 2 are the fit of the data to this functional form to get $\alpha = 0.02\ \mathrm{s}^{-1}$, $w(L) = 21$, $w(L/2) = 4.5$, $c = -0.21$ for the 2.7 wppm data and $\alpha = 0.04\ \mathrm{s}^{-1}$, $w(L) = 12$, $w(L/2) = 3.8$, $c = -1.5$ for the 1.35 wppm data, respectively.

Ideally, both the fitted rate constant and the ratio $w(L)/w(L/2)$ for both concentrations should be the same. The fitted values for the two concentrations agree only within a factor of 2. Furthermore, the fitted value of $w(L)$ is always found to be more than 3 times larger than $w(L/2)$, supporting the fact that longer polymers are better drag reducers [1]. Therefore, our model is probably too

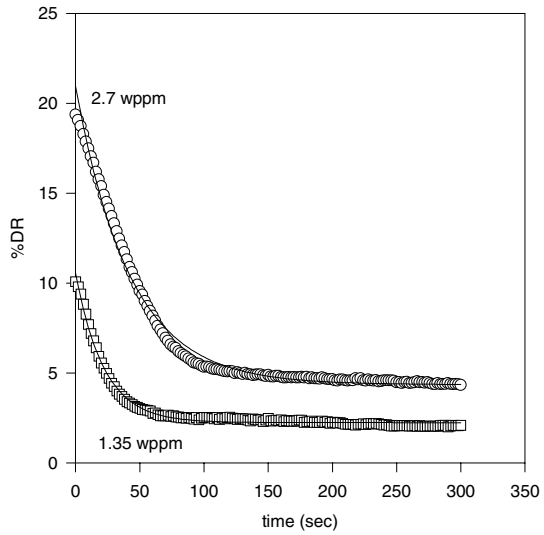

FIG. 2. Fit of early time data from Fig. 1 to the simple first order degradation model in the form $\%\mathrm{DR}(t) = w_1 e^{-\alpha t} + w_2(1 - e^{-\alpha t}) + c$ where $\alpha = 0.02\ \mathrm{s}^{-1}$, $w_1 = 21$, $w_2 = 4.5$, and $c = -0.21$ for 2.7 wppm and $\alpha = 0.04\ \mathrm{s}^{-1}$, $w_1 = 12$, $w_2 = 3.8$, and $c = -1.5$ for 1.35 wppm.

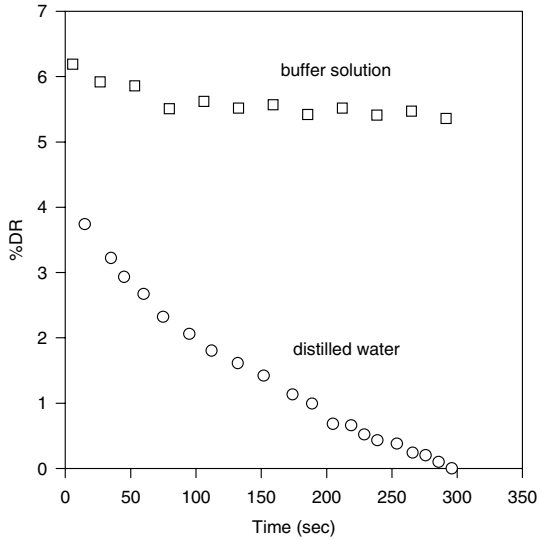

FIG. 3. Difference between measured %DR of 1.35 wppm λ-DNA at $R_e = 7 \times 10^5$ in buffer solution and distilled water.

rough for detailed analysis but is still good for the orders of magnitude estimate. The rate constant α contains information on the dynamics of degradation. If turbulence is space filling, α can be estimated as the inverse of the turnover time for the largest eddy which is about 0.03 s. This suggests that the high shear regions in the turbulent flow are concentrated in a small fraction of the total volume; about $0.02 \times 0.03 \sim 6 \times 10^{-4}$ which is about 2 orders of magnitude smaller than the predictions of current intermittency models [14]. The discrepancy is probably due to the fact that MMD will not only need velocity fluctuations with high shear rate but also would require long enough duration such that the molecules are fully stretched. Presumably, the probability of having both a large and long-life fluctuation is small.

In order to change the structure of DNA in the solution, experiments with distilled water are also performed. Figure 3 shows the result of an experiment in which λ-DNA is injected into the high turbulent flow of both distilled water and buffer solution. In distilled water, DNA molecules will denature into two single strands. The experiment is performed at (1157 rpm, $\mathrm{Re} = 7 \times 10^5$). This is chosen because the degradation behavior of λ-DNA in buffer solution at this speed is not apparent. As shown in the figure, %DR for the buffer solution degraded by less than 15% during the experiment. However, %DR for the distilled water experiment drops to close to zero at the end of the experiment, a degradation of more than 90%. Since both the ssDNA and the dsDNA have more or less the same length, the important difference between them can be only the mechanical response of the molecule to external stress such as stiffness and bending rigidity. Therefore, it seems that the double-stranded structure of the DNA is responsible for the drag reducing power of the DNA.

Since DNA is a double-stranded molecule, the force needed to break it by tensile stretching will be at least twice that of a single-strand molecule. One can consider the dsDNA as having a spring constant about twice as stiff as a ssDNA. Furthermore, due to the double helix structure of the dsDNA, its bending rigidity is larger (about 40 times) than that of its ss counterpart. When the DNA is denatured, its behavior should be similar to normal long flexible chain linear polymers as we found in the distilled water experiment with DNA shown in Fig. 3. Since a ssDNA and a dsDNA will be extended to more or less the same length before scission, the DR measurement of Fig. 3 suggests that, in the case of a dsDNA vs ssDNA, the length of the polymer is not as important as the stiffness and flexibility of the molecule in drag reducing power. Figure 3 also shows that the DR power of DNA in buffer solution is about twice that in distilled water before degradation. Since one dsDNA will give rise to two ssDNA, the concentration in the ssDNA solution will be twice that of the dsDNA solution. As %DR is proportional to polymer concentration at low concentration, our measurement suggests that %DR is about 4 times stronger for a dsDNA than a ssDNA.

From the above discussions, it is clear that dsDNA is a better drag reducer than ssDNA in the sense that not only does dsDNA give a higher %DR efficiency for the same concentration but also possesses a greater resistence to degradation. However, it is difficult to compare the %DR between dsDNA and the common long-chain polymers such as the PEO used in our experiment as their lengths are so different. It is difficult to find a PEO sample with molecular length similar to that of λ-DNA. But for concentrations similar to those depicted in Fig. 1, it can be seen that the degradation rate of dsDNA is smaller than that of PEO although they have similar initial drag-reduction

efficiencies. However, it has to be mentioned that the stronger resistance of dsDNA to MMD does not mean that dsDNA is a more efficient drag reducer than other normal linear polymers. DR efficiency and resistance to MMD can be two different issues.

In the cascade picture of Kolmogorov, the smallest length scale in the flow is given by $l^* = l_0 \mathrm{Re}^{-3/4}$ [15]. With l_0 being the dimension of our rotating disk and $\mathrm{Re} \sim 1.2 \times 10^6$, this l^* is of the order of a few microns which is comparable to the size of a DNA. The shear rate γ at this length scale is given by $\gamma^* = (\epsilon/l^{*2})^{1/3}$ (highest shear rate) where ϵ is the power per unit mass transferred to the flow from large scales to small scales. If we consider a molecule with length L exposed to a simple shear flow, the molecule will be stretched because of the drag of the medium. The tension along the length of the molecule will be maximum at the midpoint of the molecule and can be calculated by assuming that the highly stretched chain is free draining [16] to give $F_{\max} \sim \eta\gamma L^2$ where η is the viscosity of the fluid. If γ is given by the cascade picture at length scale L, then in terms of ϵ, $F_{\max} \sim \eta\epsilon^{1/3}L^{4/3}$. Thus, it can be seen that $F_{\max}$ decreases with shorter chain length. Since the bond breaking strength in a dsDNA is fixed, there will be a length scale below which the DNA will not be degraded by the flow. In order to break a dsDNA by stretching, $F_{\max}$ must be of the order of 500 pN [17]. For an order of magnitude estimation, $\epsilon \sim 0.1$ W/g [18] (as estimated by the power needed to maintain the rotation of the disk) and $L \approx 16$ μm, one gets $F_{\max} \sim 10^3$ pN, which is sufficient to break the dsDNA. For dsDNA of length $L/2$, $F_{\max}$ is less than the breaking strength of dsDNA and hence not degraded as confirmed by the 8 h degradation experiment mentioned above.

However, the above degradation picture will predict, contrary to experimental results, that the PEO shown in Fig. 1 will not be broken by the turbulence at all because of its shorter length. Usually, the length of polymers used in DR experiments are smaller than the microscales of the turbulence, but they all degrade under turbulence. A possible explanation for their degradation is that they are not degraded by tensile extension as in the case of DNA. Recently, it has been reported that there can be strong deformation in the conformation of the flexible polymer under the passive advection of a turbulent flow in the length scale of the polymer [19]. If the bending rigidity of the polymer is not strong enough, degradation can be induced by the advection of the flow. Presumably, the longer persistence length in the double helix of the dsDNA gives a stronger bending rigidity for the DNA. However, validity of this argument remains to be tested by future experiments.

This work has been supported by research grants from the KOSEF through the ARC at Korea University and the National Science Council of Republic of China under Grants No. NSC 90-2112-M008-037 and No. NSC 89-2112-M001-058.

*To whom all correspondence should be addressed.
Email address: phckchan@ccvax.sinica.edu.tw

[1] W.-M. Kulicke, M. Kötter, and H. Gräger, Adv. Polym. Sci. **89**, 1 (1989).
[2] N. S. Berman, Annu. Rev. Fluid Mech. **10**, 47 (1978); H. J. Choi, C. A. Kim, J. I. Sohn, and M. S. Jhon, Polym. Degrad. Stabil. **69**, 341 (2000).
[3] See, for example, K. R. Sreenivasan and C. M. White, J. Fluid Mech. **409**, 149 (2000); E. Balkovsky, A. Fouxon, and V. Lebedev, Phys. Rev. E **64**, 056301 (2001), and references therein.
[4] A. Groisman and V. Steinberg, Nature (London) **405**, 53 (2000); Phys. Rev. Lett. **86**, 934 (2001).
[5] T. Perkins, D. Smith, and S. Chu, Science **276**, 2016 (1997).
[6] N. Sasaki, Y. Maki, and M. Nakata, J. Appl. Polym. Sci. **83**, 1357 (2002).
[7] M. E. Cowan, C. Garner, R. D. Hester, and C. L. McCormick, J. Appl. Polym. Sci. **82**, 1222 (2001).
[8] J. H. Hand and M. C. Williams, Nature (London) **227**, 369 (1970).
[9] C. A. Kim, D. S. Jo, H. J. Choi, C. B. Kim, and M. S. Jhon, Polymer Testing **20**, 43 (2001).
[10] It is known that DNA will be affected by ions. Since the container is made of aluminum, there will be a layer of Al_2O_3 which can insulate the metal from the buffer solution. Furthermore, the buffer will neutralize ions from the container if there are any. Also, we have performed experiments at moderate Reynolds number such that there is DR but nearly no MMD; demonstrating that properties of the DNA are not affected by the materials of our apparatus.
[11] H. Schlichting, *Boundary Layer Theory* (McGraw-Hill, New York, 1979), 7th ed.
[12] Briefly, the length distribution of the concentrated sample of degraded DNA is analyzed in an electrophoresis device against a known sample of length markers. The length of degraded DNA can be measured by comparison of the positions of electrophoresis bands produced by the length markers. It is found that our degraded DNA sample produces only two bands during electrophoresis, one at 48 kbp and the other at 23.1 kbp, indicating that some of the 48 kbp DNA is being cut by half.
[13] The reason why half of the 48 kbp DNA appears to be 23.1 kbp instead of 24 kbp is due to the specific sequence in the λ-DNA to be cut by a specific enzyme for the marker for electrophoresis measurement. Hence, one can consider 23.1 kbp as a half of the 48 kbp DNA.
[14] K. R. Sreenivasan and C. Meneveau, Phys. Rev. A **38**, 6287 (1988).
[15] H. Tennekes and J. L. Lumley, *A First Course in Turbulence* (MIT Press, Cambridge, 1972).
[16] P.-G. de Gennes, J. Chem. Phys. **60**, 5030 (1974).
[17] D. Bensimon, A. J. Simon, V. Croquette, and A. Bensimon, Phys. Rev. Lett. **74**, 4754 (1995).
[18] The electric power needed to maintain disk rotation is about 300 W, and we assumed that turbulent dissipation is of the order of 100 W.
[19] M. Chertkov, Phys. Rev. Lett. **84**, 4761 (2000); E. Balkovsky, A. Fouxon, and V. Lebedev, Phys. Rev. Lett. **84**, 4765 (2000).