



# Mechanical degradation of water-soluble acrylamide copolymer under a turbulent flow: Effect of molecular weight and temperature



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## ABSTRACT

An experimental study of the turbulent drag reduction (DR) performance of water-soluble poly(acrylamide-co-acrylic acid) copolymers with two different molecular weights in a rotating disk apparatus is reported. The DR efficiency of very dilute polymer solutions was measured to relate their DR activity to molecular parameters, such as molecular weight, concentration, temperature, and rotational speed of the disk. Experimental results show that polymers with high molecular weights and high concentrations exhibit a persistent DR activity to mechanical degradation under a turbulent flow.

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## Introduction

The skin frictional drag of a turbulent flow can be remarkably and actively decreased owing to the employment of tiny amounts of additives, such as flexible polymeric chain molecules [1–3] and colloidal surfactants, compared to non-additive passive drag reduction (DR) systems [4]. Among various active drag-reducing materials, linear, long-chain, flexible, and high molecular weight polymers are known to be the most effective drag reducers. Due to the pronounced effect of polymers on the turbulent DR, many commercial and scientific applications have been reported, including those relevant to crude-oil pipelines [5], irrigation systems [6], firefighting [7], and biomedicine [8].

Various theories and mechanisms on turbulent DR have been reported. Specifically an elastic theory of the turbulent DR was introduced to discuss the properties of homogeneous and isotropic three-dimensional turbulence in the presence of polymer additives without any wall effects using a cascade theory [9]. The central idea of this theory, limited to linear flexible chains in a good solvent, is that polymer effects on small scales are not described by the viscosity but by the elastic modulus. The importance of an elastic property for the description of the DR mechanism was also examined by adopting a simple model to study both the turbulence

and dissolved polymer molecules [10]. On the other hand, the turbulent DR was also interpreted using a model in terms of the solvation of polymer chains, and the formation of relatively stable domains, in which the domains partly suppressed the vortex formation and act as energy sinks [11]. It was also found that the flexibility of the polymers helps the domain formation, and that higher molecular weights help to resist turbulence eddies in a better manner. Note that these mechanisms are further related to the idea of polymer threads proposed by Bewersdorff [12], confirming that the central thread provides DR, which is almost equivalent to premixed solutions of the same total polymer concentration flowing in the pipe [13]. Despite the existence of many available theories, note that a full understanding of this theory is still lacking.

Nonetheless, in terms of the experimental drag-reducing polymers, water-soluble poly(ethylene oxide) PEO [14,15], polyacrylamide (PAAM) [16,17], and commercial polysaccharides with high molecular weight were extensively used in aqueous systems, while nonpolar drag reducers, such as polyisobutylene (PIB) and polystyrene (PS), have been used in hydrocarbon-based fluids [18]. Various parameters, including polymer concentration, polymer molecular weight, temperature, Reynolds number, and solvents [14,19], have been found to strongly influence the DR efficacy.

Although the polymers are considered as the most effective drag reducers, they are less resistant to high mechanical shears – such as those generated by pumps – and under turbulent flows

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with high shear and extensional rates. The molecular degradation of the polymers can be caused by skin friction [20] and is very sensitive to temperature variations [6]. Recently, to reduce the polymer susceptibility to bond scission, graft copolymers have attracted much attention. The PAAM grafted on polysaccharides – with a varying number and length of grafted chains [21] – was found to be more stable under shears than linear PAAM [22]. In addition, the copolymers with a small portion of hydrophobic moieties [23] were also investigated and the persistent DR efficiency due to the intermolecular interaction was identified.

In this study, the copolymer of poly(acrylamide-co-acrylic acid) [poly(AM-co-AA)] was chosen as a drag reducer to investigate the influences of various parameters, such as molecular weight, concentration, temperature, and rotational speed, on its turbulent DR in a rotating disk apparatus.

## Experimental

The motor assembly with the motor and loader that was used to operate the disk consists of a servo motor, which can be operated up to a maximum rotational speed of 3000 revolutions per minute (rpm) at a very high rotational acceleration [19]. The rotational speed and motor power control are determined by the loader. The loaded torque on the disk is detected using a rotary shaft torque sensor. It can measure the torque from 0 to 200 oz in. accurately. One shaft of the torque sensor is coupled to the motor using a steel coupling, and the other is joined directly to the hollow disk shaft to monitor the torque. In order to examine the DR efficiency, torque was measured by a rotating disk apparatus (RDA) in which a disk rotates at uniformly set speeds. The vessel of the apparatus was made of stainless steel, and its dimension was 15.9 cm in diameter and 5.6 cm in depth. The disk was also made of stainless steel with dimensions of 14.5 cm in diameter and 0.3 cm in thickness. The volume of the solution required to fill the entire container was approximately 1020 cm<sup>3</sup>. While the rotational velocity of the disk was controlled by a speed controller (Cole Parmer Master Servodyne Unit), the variable torque was measured by a multimeter. The temperature of the system was maintained by a constant temperature circulating apparatus at 25 °C with an error range of 0.5 °C unless the dependence on the temperature was at 40 °C and 80 °C. The rotational velocity of the disk was confirmed by a digital tachometer. However, the temperature increased initially by a few degrees when the rotational speed was increased from 0 rpm to the set value. Therefore, we operated the setting temperature to be lower than the experimental temperature, making sure that the polymer was injected into the chamber at the appropriate temperature of either 40 °C or 80 °C.

Turbulent flows occur in the RDA when  $N_{Re}$  is larger than  $3 \times 10^5$  ( $\omega > 570$  rpm) [24]. Here,  $N_{Re}$  is defined as:

$$N_{Re} = \frac{\rho r^2 \omega}{\mu} \quad (1)$$

where  $\rho$  is the fluid density,  $\mu$  is the fluid viscosity,  $r$  is the radius of the disk, and  $\omega$  is the angular velocity [rad/s,  $2\pi \times$  (revolution per minutes)/60]. Since the DR phenomenon of the polymer occurs only in the turbulent region, all the experiments in this study were undertaken at rotational speeds above this critical rotational velocity of the disk. It can be also noted that a cylindrical double gap rheometer device has also been recently introduced to study polymeric turbulent DR [25,26].

Stock solutions of high molecular weight poly(AM-co-AA) ( $M_w = 5 \times 10^6$  g/mole and  $1.5 \times 10^7$  g/mole, Sigma-Aldrich, USA) at 0.5 wt% were prepared in advance with mild stirring for one week at room temperature, using de-ionized water as a solvent. A desired amount of stock solution for a specific polymer

concentration of the DR measurement was subsequently injected into the RDA for the actual turbulent DR study. Note that the test of the molecular weight was conducted using two different molecular weight copolymers only due to its limit on the commercial availability, and the DR data of  $M_w = 5 \times 10^6$  g/mole was adopted from our previous work for the reanalysis [27].

The percentage DR (%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. To obtain the DR efficiency of the rotating disk apparatus, the torque that was required to rotate the disk in the pure solution at a given speed was measured first. The percent DR was then calculated by measuring – at the same speed – the corresponding torque that was required in the solution with the added polymer, as follows:

$$\%DR(t) = \left( \frac{T_s - T_p(t)}{T_s} \right) \times 100 \quad (2)$$

where  $T_s$  is the torque of the pure solvent and  $T_p$  is the torque of the polymer dilute solution. Unless the rpm was mentioned, the DR test was performed at 1980 rpm, which is corresponding to about  $N_{Re} = 1 \times 10^6$ .

Furthermore, the shear viscosity of the polymer solution with 50 wppm poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) was examined using a rotational rheometer (Physica, MCR300, Germany) with a Couette cell geometry (DG 26.7) at different temperatures (25, 40 and 80 °C). The steady shear viscosity was measured as a function of shear rate in the range of 0.01–100 s<sup>−1</sup>.

## Results and discussion

Fig. 1 shows the shear viscosity as a function of three different temperatures (25, 40, and 80 °C) for 50 wppm of poly(AM-co-AA). Severe shear-thinning behavior was clearly observed for this copolymer solution ( $M_w = 1.5 \times 10^7$  g/mole) [28]. It is also confirmed that the shear viscosity decreased with increasing temperature as a function shear rate. And the approximate shear rate in the DR test condition could be estimated using diameter and thickness of the disk in our RDA geometry and a rotational speed of revolutions per minute (rpm) of 1980 rpm with a shear rate of about 570 s<sup>−1</sup>. The infinite shear viscosity of the polymer solution thereby could be considered as a polymer solution viscosity for estimating the rotational Reynold number. Furthermore, the overlap concentration could be also estimated to be 200 ppm based on PAAM of  $M_w = 1.8 \times 10^7$  g/mole [29]. Therefore our

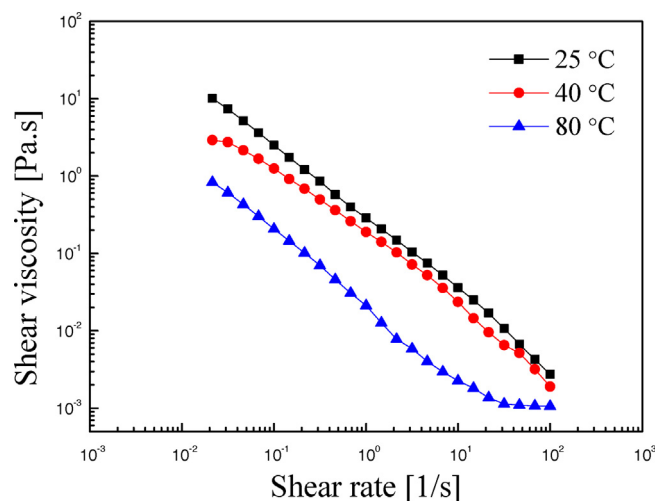
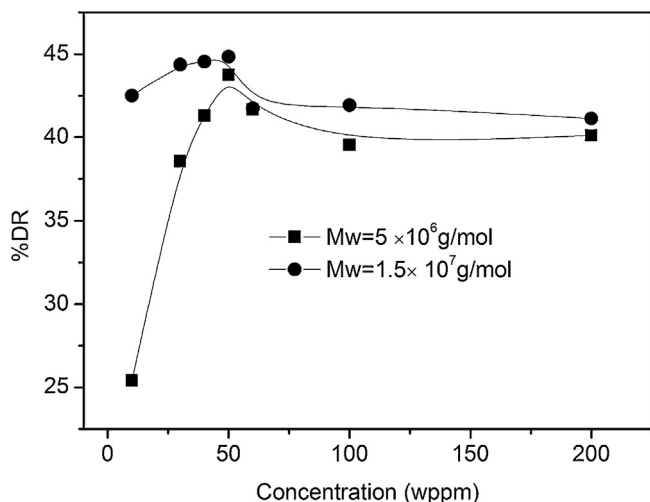


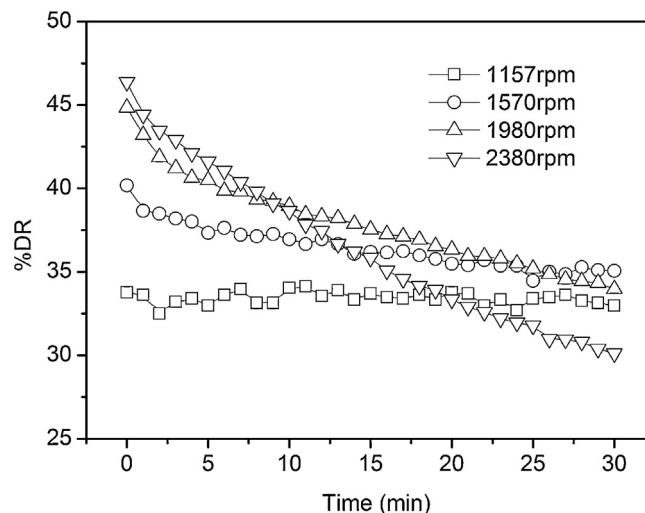
Fig. 1. Shear viscosity as a function of shear rate for different temperatures of poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) solution at a concentration of 50 wppm.



**Fig. 2.** %DR versus concentration of poly(AM-co-AA) copolymer for two different molecular weights ( $5 \times 10^6$  g/mole [27] and  $1.5 \times 10^7$  g/mole) at 1980 rpm.

polymer concentration for the DR tests is considered to be in the dilute polymer solution condition.

Fig. 2 shows the dependence of the DR efficiency on the polymer concentration for two different molecular weights at 1980 rpm measured at room temperature. For higher polymer concentration, the higher shear viscosity could be expected. However at such a high rpm of 1980 rpm corresponding to about 570(1/s) shear rate, the shear viscosity difference could become narrower as similar to Fig. 1 without significantly affecting the rotational Reynold number. Even though different polymer concentrations could slightly affect the rotational Reynold number, we kept the rpm constant for experimental convenience. The %DR efficiency increases with a polymer concentration at lower concentrations, and it then slightly declines before it becomes steady at a high concentration. The same trend applies to both polymer solutions, while the polymer with a higher molecular weight ( $M_w = 1.5 \times 10^7$  g/mole) shows a larger DR efficiency than that of the lower molecular weight polymer ( $M_w = 5 \times 10^6$  g/mole) over the entire polymer concentration range. Here, specifically, at 10 ppm, the DR efficiency of the higher molecular weight polymer (42.5%) is nearly twice as large as that of the lower molecular weight polymer (25%). In addition, the maximum DRs can be found at increasing polymer concentrations in both polymer solutions. Specifically, the polymer concentration for the maximum DR appeared at 50 wppm for the lower molecular weight polymer and shifted to a slightly lower concentration for the higher molecular weight polymer. Note that the DR efficiency of the same copolymer with lower molecular weight polymer ( $M_w = 5 \times 10^6$  g/mole) was previously tested and then interpreted using the Brostow equation [27]. Our group [30,31] had reported that the maximum DR efficacy of PEO (50wppm) is approximately 35%. Additionally, the degradation rate for PEO and poly(AM-co-AA) is 25% and 13% after 30 min at 25 °C, suggesting that poly(AM-co-AA) is less degradable. In addition, the degradation rate for 50 wppm guar gum [32] is 43% after 30 min, indicating that guar gum is more degradable when compared with poly(AM-co-AA). On the other hand, the maximum DR efficiency as a function of concentration observed in Fig. 2 could be explained based on assumption that the turbulent DR is caused by a sum of contribution of individual polymer molecules, the concentration of polymer solution have a positive effect on the increment of drag reduction. As the concentration of polymer increases, the shear viscosity of the polymer solution is increased leading to the increase of drag reduction, while the shear viscosity becomes increasingly significant at higher concentrations



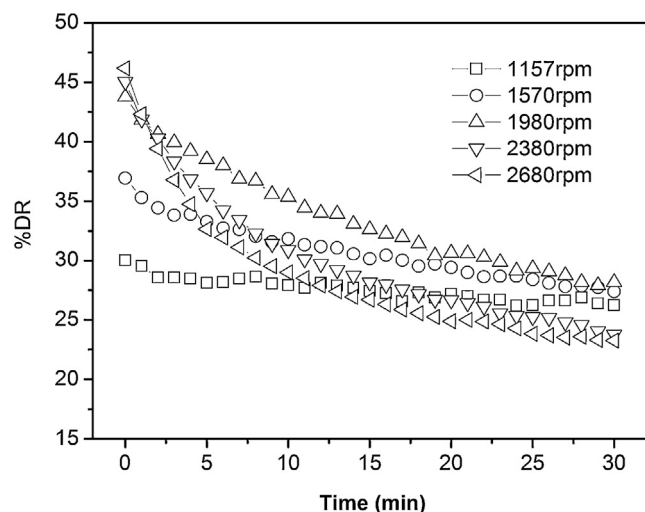
**Fig. 3.** %DR for 50 wppm of poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) under different rotational speed at 80 °C.

of polymer, resulting in a lower drag reduction efficiency [33]. Such behavior has been also reported for a pipe flow [34].

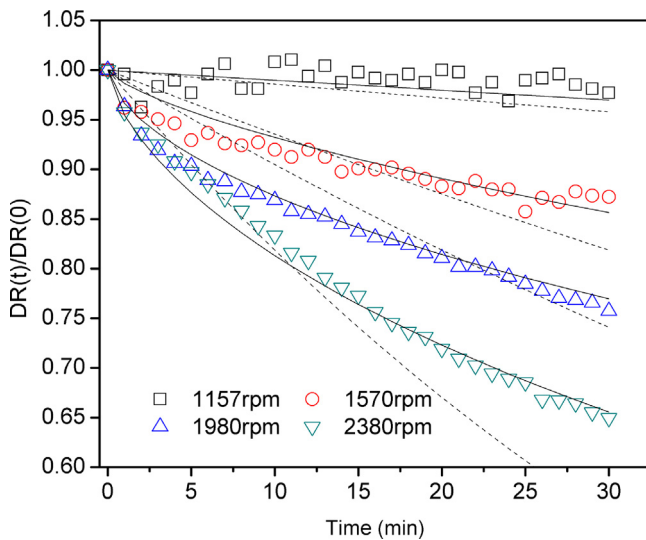
Figs. 3 and 4 depict the influence of the rotational speed on the DR efficiency of a 50 wppm copolymer at 80 °C. For all the examined rotational speeds, the DR efficiency decreased with time, a finding that is attributed to the mechanical degradation of the polymer chains induced by a shear flow. At high rotational speeds, the DR efficiency decreased more rapidly than at low rotational speeds, indicating that the polymer was susceptible to the mechanical degradation because of the increase in the mechanical energy of the violent turbulent flow. In comparison to the mechanical degradation behavior of the higher molecular weight copolymer (Fig. 3), the polymer with lower molecular weight (Fig. 4) was more easily and rapidly degraded with time at the same rotational speed.

$$\frac{DR(t)}{DR_0} = \exp\left(-\frac{t}{\lambda_1}\right) \quad (3)$$

$$\frac{DR(t)}{DR_0} = \exp\left[-\left(\frac{t}{\lambda_2}\right)^{1-n}\right] \quad (4)$$



**Fig. 4.** %DR for 50 wppm of poly(AM-co-AA) ( $M_w = 5 \times 10^6$  g/mole) under different rotational speed at 80 °C [27].



**Fig. 5.**  $DR(t)/DR(0)$  versus time for poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) under different rotational speed at 80 °C. Dotted lines are obtained from Eq. (3) and solid lines are obtained from Eq. (4).

Eqs. (3) and (4) [32] were used to quantitatively describe the degradation profiles for the copolymers in Figs. 3 and 4, respectively, which indicates that the degradation rates increase with increasing rotational speeds. The relative DR =  $DR(t)/DR(0)$  of the copolymer at the higher molecular weight of  $1.5 \times 10^7$  g/mole (Fig. 5) shows a more persistent DR activity than that of the copolymer with the lower molecular weight ( $5 \times 10^6$  g/mole) (Fig. 6), suggesting that the polymer with the higher molecular weight is less susceptible to mechanical degradation under a turbulent flow. From Fig. 6, we can see that the DR percentage declines by about 10% at every rpm when compared with that of Fig. 5. In addition, when compared to the single-relaxation process (Eq. (3)), it can be seen that the stretched exponential model (Eq. (4)) fits the degradation data well for both copolymers at different rotational speeds, as shown in Figs. 5 and 6. The fitting of the parameters is implemented over a long time range, as listed in Tables 1 and 2. The values of  $\lambda_2$ s for the copolymer poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) at 1157 rpm, 1570 rpm, 1980 rpm,

**Table 1**

Optimal parameters in Eqs. (3) and (4) for poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) (50 wppm) under different rotational speed at 80 °C.

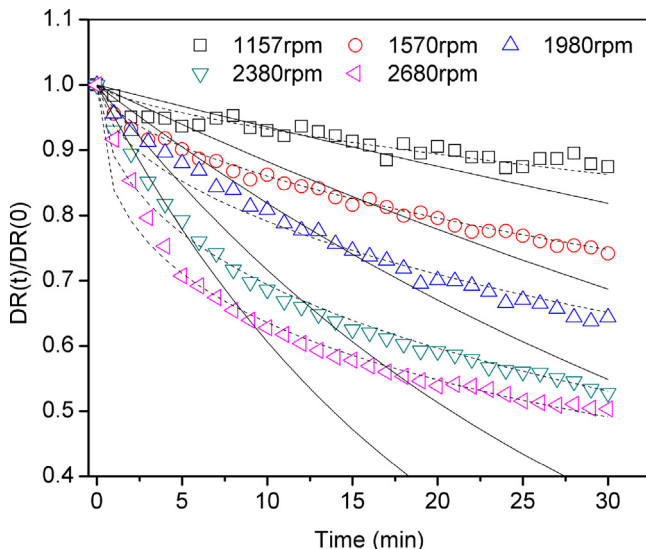
Rotational speed	$\lambda_1$	$\lambda_2$	$N$
1157 rpm	700.07	1000.10	0.005
1570 rpm	150.22	400.09	0.28
1980 rpm	100.56	280.99	0.40
2380 rpm	50.04	113.13	0.35

**Table 2**

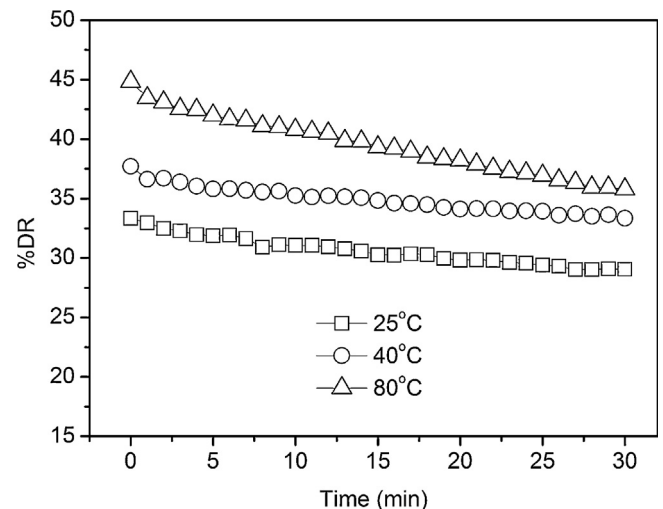
Optimal parameters in Eqs. (3) and (4) for poly(AM-co-AA) ( $M_w = 5 \times 10^6$  g/mole) (50 wppm) under different rotational speed at 80 °C.

Rotational speed	$\lambda_1$	$\lambda_2$	$N$
1157 rpm	150.56	162.11	0.004
1570 rpm	80.82	100.93	0.13
1980 rpm	50.03	80.67	0.29
2380 rpm	30.41	45.34	0.35
2680 rpm	20.50	35.02	0.45

and 2380 rpm, are estimated to be 1000.10 min, 400.09 min, 280.99 min, and 113.13 min, respectively. On the other hand, the  $\lambda_2$ s values for the copolymer of poly(AM-co-AA) ( $M_w = 5 \times 10^6$  g/mole) at 1157 rpm, 1570 rpm, 1980 rpm, 2380 rpm, and 2680 rpm are estimated to be 162.11 min, 100.93 min, 80.67 min, 45.34 min, and 35.02 min, respectively. The effect of temperature on the DR efficiency was investigated with a 50 wppm copolymer at different temperatures (25 °C, 40 °C, and 80 °C) for a period of 30 min, as shown in Fig. 7. The initial %DR of the copolymer solution increased at increasing temperatures, similar to the results obtained for the linear polymer of PAAM [35]. Note that the viscosity of water also decreases with increasing temperature [36], but the values are obviously smaller than that of the poly(AM-co-AA) solution as shown in Fig. 1. Considering no shear thinning behavior appearing in the Newtonian fluid, it suggests that the viscosity of a polymer solution could be increased a lot with small concentration compared with Newtonian fluid (pure water), leading to higher DR phenomena at higher temperature. In addition, while the shear viscosity difference becomes much narrower for different temperatures at a high shear rate, the temperature increase could cause the reduction of viscosity, resulting in the increase of Reynolds number slightly, finally helping the increase of drag reduction. However, at a

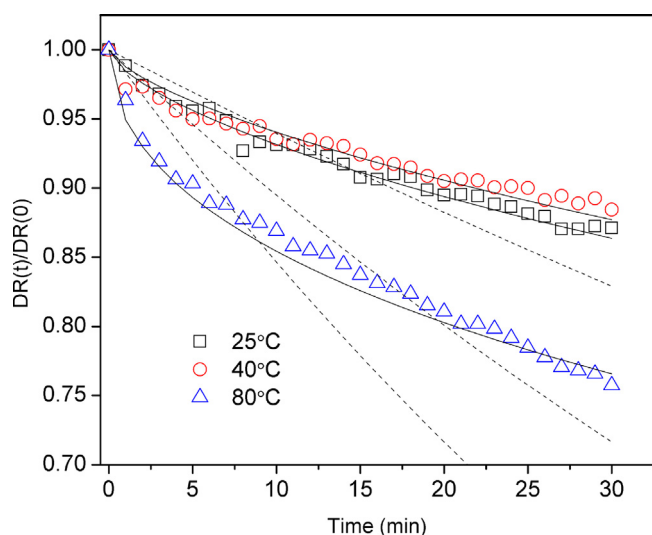


**Fig. 6.**  $DR(t)/DR(0)$  versus time for poly(AM-co-AA) ( $M_w = 5 \times 10^6$  g/mole) under different rotational speed at 80 °C. Solid lines are obtained from Eq. (3) and dotted lines are obtained from Eq. (4).



**Fig. 7.** %DR versus time of copolymer solution ( $M_w = 1.5 \times 10^7$  g/mole) for different temperatures (25 °C, 40 °C and 80 °C) at 1980 rpm.





**Fig. 8.**  $DR(t)/DR(0)$  versus time for copolymer solution ( $M_w = 1.5 \times 10^7$  g/mole) under different temperatures (25 °C, 40 °C and 80 °C) at 1980 rpm. Dotted lines are obtained from Eq. (3) and solid lines are obtained from Eq. (4).

given temperature, the DR efficiency decreased with time rapidly, especially at a temperature of 80 °C.

The relative  $DR = DR(t)/DR(0)$  of the copolymer with the higher molecular weight of  $1.5 \times 10^7$  g/mole was also observed under different temperatures, as shown in Fig. 8. It can be seen that the degradation rates are less than 15% at 25 °C and 40 °C, but changes to a value close to 25% at 80 °C at 30 min. Similarly, Eq. (4) fitted the data of the ratio of  $DR(t)/DR(0)$  well under different temperatures, compared to Eq. (3). The parameter fitting was performed for a period of 30 min, and the outcomes are listed in Table 3. The  $\lambda_{2s}$  values for the copolymer poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) at 25 °C, 40 °C, and 80 °C were estimated to be 550.04 min, 570.82 min, and 470.37 min, respectively.

On the other hand, the DR efficacy was found to increase with increasing concentration within the entire measurement range. In addition, the polymer degraded within a few minutes of the initial injection. After the degradation, the DR efficacy leveled off and was maintained at the same value for a long time. At the same concentration, the copolymers with a higher molecular weight had a hydraulic radius of the chain that was larger in deionized water than polymers with a low molecular weight. The polymer with a high molecular weight was considered to be sufficiently shielded against the mechanical degradation in the solvent [37], resulting in the higher drag efficacy. Note that Pereira and Soares [25] recently reported polymer degradation in terms of DR as a function of concentration, molecular weight, Re and temperature all combined.

As the solution concentration was increased, an increase in the polymer concentration shifted the start point of the logarithmic region to a higher value. Beyond the optimum concentration, further increases of the concentration led to a leveling-off or a slight decrease in the DR. This tendency can be explained as follows: the initial increase in the DR was presumably due to the

increasing number of molecules present that caused the decay of turbulent eddies, while at higher concentrations and with an increasing polymer concentration, the polymer domains began to compete for solvent molecules in an effort to solvate them, resulting in a lower DR [31]. Meanwhile, it is well known that a maximum %DR efficacy of 80% can be achieved in the pipe flow, while only approximately 50% of the maximum DR efficacy can be obtained for this RDA flow which is one of the external flows [38]. In the internal flow, the total drag is the summation of the form drag and the skin drag while the drag in the inner flow such as a pipe flow is a skin drag. Because the turbulent drag reduction comes only from the skin drag, the DR efficiency of the external flow is limited compared to the inner flow.

The DR behavior in a turbulent flow appeared to be related to elongation straining as well as to shearing force. Both of these factors caused the breakage of the main chain of the polymer based on the mechanical energy. The higher the rotational Reynolds number with a higher rotational speed, the more degradation has been observed [35]. In this study, note that the copolymer with a higher molecular weight used as the effective drag reducer is more resistant to mechanical degradation than that of the lower molecular weight at the same rotational speed. In addition, the degree of mechanical degradation in poor solvents was found to be higher than those in good solvents [39,40], and vice versa, in regard to the DR efficacy.

Moreover, the mechanical degradation shown in Fig. 8 becomes more obvious (showing a decrease in DR efficiency with time) as temperature increased from 25 °C to 80 °C. Note that all these DR efficiency for different temperature based on the polymer solution at each temperature at high shear rate. The variation in the DR efficacy is related to the destruction of intramolecular hydrogen bonding due to the increase in the thermal motion of molecules at 80 °C. This is considered to be the reason for the observed increased degradation in the DR of copolymer poly(AM-co-AA) at elevated temperatures. Besides we found that shear viscosity of poly(AM-co-AA) solution decreases with increasing temperature as shown in Fig. 1, which is probably the reason of increase of Reynolds number, finally helping to increase of DR efficiency.

## Conclusion

We investigated DR efficacies induced by the copolymer of poly(AM-co-AA) in RDA under different situations by varying the molecular parameters, such as the molecular weight, concentration, temperature, and rotational speed. It is confirmed that the polymers at a high molecular weight and concentration exhibit highly efficient and shear resistible DR characteristics. In addition, the persistent DR activity is found to decrease with increasing temperature that is related to the increase of the thermal motion of molecules at elevated temperatures, leading to the breakage of the intermolecular interaction.

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**Table 3**

Optimal parameters in Eqs. (3) and (4) for poly(AM-co-AA) ( $M_w = 1.5 \times 10^7$  g/mole) (50 wppm) under different temperatures (25 °C, 40 °C and 80 °C) at 1980 rpm.

Temperature	$\lambda_1$	$\lambda_2$	$N$
25 °C	160.01	550.04	0.34
40 °C	90.45	570.82	0.31
80 °C	60.22	470.37	0.52

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