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A novel approach for drag reduction using polymer coating

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Abstract

Polymer drag reduction (DR) remains challenging for marine applications due to the difficulties in introducing the polymer drag-reducer into external flows. We developed a novel coating that bonds drag-reducing polymers to metallic surfaces. The coating consists of a polydopamine (PDA) layer that can attach to any substrate. A layer of anionic polyacrylamide (APAM), which is a drag reducing agent, is then grafted on the PDA-coated surface. Owing to the covalent bond, the long chains of APAM polymer slowly dissolve from the PDA-coated surface into water. The drag of surfaces with 0.1 to 0.8 mg cm⁻² of deposited APAM at three Reynolds numbers, Re , of 8×10^3 , 11×10^3 , and 16×10^3 were investigated in a turbulent channel flow. The maximum amount and duration of DR was achieved for an optimum APAM deposition of 0.4 mg cm⁻² at Re of 16×10^3 . A large DR of 19% occurred in the first 10 min of the tests, and then gradually reduced to zero within an hour. We also observed that increasing Re resulted in a greater initial DR that spanned over a shorter duration, potentially due to a faster dissolving rate of the polymer coating.

Keywords

Drag reduction, polymer coating, wall-bounded turbulent flows

26 **1. Introduction**

27 Reducing the drag associated with turbulent flows has always been an active research area due to its
28 widespread industrial applications. Among the various turbulent drag reduction methods used for liquid
29 flows, the most successful and robust one is the addition of a small amount of a polymer with high molecular
30 weight into the fluid. Toms, (1948) first established this phenomenon, and subsequently its drag-reducing
31 mechanism was investigated by numerous experiments (Ptasinski et al., 2001; White and Mungal, 2008).
32 Polymer drag-reduction typically requires less than 100 weight parts per million (wppm) of the polymer to
33 result in up to 70% drag reduction (Escudier et al., 2009; Ptasinski et al., 2001; Virk et al., 1967). As a
34 result, industry applies polymer drag reduction for reducing pumping costs in various applications including
35 petroleum pipelines, sewage networks, and firefighting systems.

36 In most laboratory-scale experiments and industrial applications, the drag-reducing polymer mixes with
37 the whole fluid to achieve a homogeneous solution. This method is feasible for internal flows where a finite
38 volume of fluid is present, e.g. for pipe flows. In contrast, for external flows, such as the flow over ships
39 and submarines, it is not feasible to introduce a homogeneous polymer solution as freshwater continuously
40 enters the boundary layer. As an alternative, a concentrated solution of the drag-reducing polymer is
41 injected from the surface into the thin turbulent layer that surrounds the surface. The injected polymer
42 gradually mixes and disperses in the surrounding flow. Since the resulting polymer solution is
43 heterogeneous and its concentration decreases with increasing distance from the injection port, the
44 subsequent drag reduction also varies along the surface (Berman, 1986; Hoyer and Gyr, 1996; Tiederman
45 et al., 1985; Wei and Willmarth, 1992; Willmarth et al., 1987). Therefore, to maintain a constant drag
46 reduction, the polymer should be injected uniformly along the surface, or at least from several locations.

47 The injection of polymer solution is typically carried out through several surface penetrations into the
48 turbulent boundary layer (Semenov, 1991). For marine applications, such a modification to the vessel hull
49 is not desirable and it remains impractical as the injection orifices reduce the structural integrity of the hull.
50 As a result, it is of interest to introduce the polymer solution into the flow using a method that is not
51 destructive toward the vessel hull. In a novel technique, Motozawa et al., (2010) dispersed polyethylene

52 oxide (PEO), which is a drag-reducing polymer, into conventional antifouling (AF) paint. To test the
53 coating, they applied it to the inner cylinder of a Taylor-Couette device. They measured the torque using a
54 load cell and compared the drag with a baseline non-coated surface. Their results demonstrated that the
55 PEO dissolved into water and generated an initial drag reduction (DR) of up to 20%. The DR gradually
56 diminished, and after 30 hours, the drag was higher than the non-coated surface. The latter observation was
57 associated with the large roughness of the residual AF paint. It is also important to note that due to the small
58 volume of the Taylor-Couette device (1 liter), a homogeneous polymer solution formed that was also
59 subject to shear-induced degradation. More recently, Yang et al., (2014) also investigated a drag-reducing
60 coating that combined a self-polishing AF paint and PEO. Due to a hydrolysis reaction, the AF coating
61 becomes soluble and gradually erodes, releasing the PEO into the surrounding flow. To reduce the coating
62 roughness, they sieved out the largest PEO particles. The performance of the coating was investigated in
63 several facilities including a Taylor-Couette device, a turbulent channel flow, and two towing facilities.
64 Yang et al., (2014) observed more than 10% DR with respect to the baseline AF paint, while a smaller DR
65 (~2-3%) was observed with respect to a non-coated smooth surface.

66 The previous investigations show that when the drag-reducing polymer is mixed with an AF paint a
67 rough surface is generated, which can contribute to drag increase. In addition, when the drag-reducing
68 polymer is depleted, the remaining rough AF paint may result in drag increase with respect to a smooth
69 surface (Motozawa et al., 2010; Yang et al., 2014). To address this issue, we have developed a novel
70 technique to apply a polymer coating without using an AF paint. The developed coating consists of dual
71 layers of polydopamine (PDA) and a commercial anionic polyacrylamide (APAM). The technique is
72 inspired by mussel adhesive proteins that enable robust adhesion to a variety of substrates under water.
73 Surface modification with dopamine and other catecholamine derivatives has recently created a universal
74 surface modification platform (Lee et al., 2007). Alkali-induced autoxidation of dopamine into PDA
75 coatings was the most interesting single step surface functionalization method to graft a wide range of
76 polymers subsequently (Liu et al., 2016; Ryu et al., 2018). Because of the high reactivity of a PDA coating
77 towards the nucleophiles, it plays the role of an intermediate linker to immobilize other chemical

78 components onto the surface and impart the desired property (Chang et al., 2016). In the subsequent
79 sections, we describe the coating procedure, characterize it, and evaluate its drag-reducing performance.
80 For the latter task, we used a turbulent channel flow and measured the pressure drop of the flow.

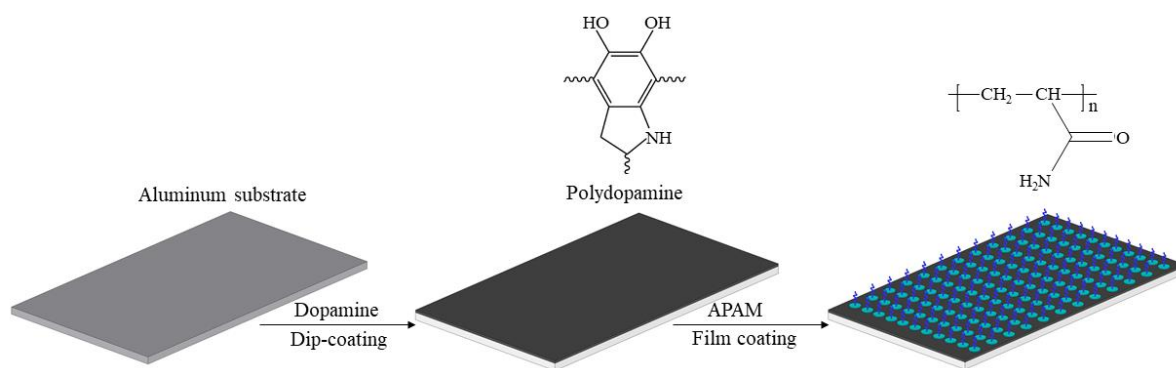
81 **2. Materials and methods**

82 In this section, we first explain the preparation of the polymer coating. Then we briefly describe the flow
83 facility and the measurements used for evaluating the DR performance of the coating.

84 **2.1. Formulation and manufacturing of the polymer coating**

85 The polymer coating consists of two layers of polydopamine (PDA) and anionic polyacrylamide
86 (APAM) as shown in Fig. 1. The APAM used has a molecular weight ranging from 12 to 15 Mg/mol and
87 an intrinsic viscosity of 24.29 deciliters per gram, supplied by SNF oil and gas. The PDA layer works as an
88 adhesion layer for the APAM solution. Dopamine hydrochloride (DA), containing both amino and phenolic
89 hydroxyl functional groups, was dissolved into a Tri(hydroxymethyl) amino methane (Tris)-HCl (50 mM,
90 pH 8.5) buffer solution with a concentration of 0.5 mg/mL. pH-induced oxidation of DA changes the
91 solution color to dark brown. Lee et al., (2007) reported that under oxidative conditions dopamine can be
92 self-polymerized to form a single-step thin coating on a wide range of substrates, mimicking the adhesion
93 of *Mytilus edulis* foot protein. Under oxidative conditions, the hydroxyl groups show deprotonation and
94 become dopamine-quinone. Later via intramolecular cyclization this dopamine-quinone subsequently turns
95 into leukodopaminechrome, which subsequently forms 5,6 dihydroxyindole or 5,6 indolequinone through
96 further oxidation and rearrangement (Bernsmann et al., 2011; Jiang et al., 2011). Inter-molecular
97 crosslinking of 5,6 dihydroxyindole or 5,6 indolequinone through branching reactions finally leads to a
98 melanin-like polymer polydopamine (Bernsmann et al., 2011) (see Fig. 1). Thus, a firmly adherent
99 dopamine into PDA layer can be formed on the surface of a substrate that is immersed in the dopamine
100 solution for a certain time. PDA coating thickness depends on the concentration of dopamine in the solution,
101 deposition time, pH, and supplied oxygen in the solution. The PDA-coated surface layers contain amino
102 groups and phenolic groups, which can react with a variety of molecules via Schiff-base and Michael

103 addition chemistries to facilitate immobilize thiol or amine containing molecules (Zeng et al., 2018). At this
104 point, PDA can be tightly attached to any material by covalent or noncovalent interactions (such as π - π
105 interactions, charge transfer interactions). Thus, using this dopamine chemistry, we had subsequently
106 grafted APAM via a possible Schiff-base or Michael-type addition reaction between oxidized catechols of
107 the PDA-coated surface and the nucleophilic amines of APAM (Yang et al., 2016).



108
109 Fig. 1. Scheme of the polymer coating layers starting with the base aluminum substrate on the left side to the APAM film coating
110 on the right side of the Fig..

111 Aluminum substrates with dimensions of $236 \times 36 \text{ mm}^2$ were first ultrasonically cleaned with acetone,
112 ethanol, and deionized water respectively for 30 min each. To achieve a homogeneous thin primary PDA
113 film on the substrates, the cleaned substrates were immersed into a freshly prepared DA solution and shaken
114 at 200 rpm with a mechanical shaker at $25 \text{ }^\circ\text{C}$ for 12 hours. The coated substrates were washed with
115 deionized water several times and then dried. Due to the great adhesion of PDA with a wide range of
116 polymers, one-step PDA coating has been thoroughly explored as a surface modifier. Prior to the grafting
117 of APAM, the freshly-coated PDA surface was sintered at $70 \text{ }^\circ\text{C}$ in an oven for 15 mins to get a homogenous
118 surface coverage of the primary coating of PDA (Gibson et al., 2019). Homogenous APAM solutions at
119 different concentrations were prepared by dissolving APAM into a 50:50 mixture of deionized water and
120 ethanol. We prepared six different concentration: 500, 1000, 1500, 2000, 3000 and 4000 weight parts per
121 million (wppm) of APAM polymer solution. The dissolving process was done via magnetic stirring to avoid
122 any mechanical degradation as suggested by Abu Rowin et al., (2018). Using film coatings, a wet layer of
123 homogeneous APAM solution with a thickness of 2.0 mm was deposited on the sintered PDA. After the

124 film coating process, the coated substrate was dried in an oven and formed a thin PDA/APAM layer. The
125 variation of wppm in the wet-film solution allows us to vary the amount of deposited APAM, D , as shown
126 in Table 1.

127 Table 1. The amount of deposited polymer for different APAM concentrations.

wppm	D (mg cm ⁻²)
500	0.1
1000	0.2
1500	0.3
2000	0.4
3000	0.6
4000	0.8

128 The coating topography and thickness were investigated using a Zeiss Sigma 300/VP- FESEM (Carl
129 Zeiss Microscopy Ltd.). A field emission scanning electron microscopy (FESEM) image of a small
130 10×10 mm² substrate of a silicon wafer was taken as a reference and shown in Fig. 2(a). The FESEM image
131 of the silicon wafer coated with a PDA layer in Fig. 2(b) shows a scatter of small roughness features spread
132 over the surface, which are mainly the aggregates of PDA deposited on the surface. The average diameter
133 of these roughness features has a wide range varying from 0.01 to 2 μm. Most of the PDA roughness
134 features were covered when the APAM layer with 0.8 mg/cm² of polymer was applied, as is shown in Fig.
135 2(c). As can be seen, the PDA/APAM coating does not generate large-scale roughness features that can
136 contribute to an increase in local turbulent drag. The thickness of the PDA/APAM coating was evaluated
137 from a side view FESEM image (e.g., Fig. 2(d)) of a silicon wafer coated with a similar coating protocol to
138 what was described in the previous section. After the drying process, the thickness of the combined PDA
139 and APAM layers was approximately 70 μm. The root-mean-square of coating roughness, estimated from
140 the side view FESEM images, was approximately 5.9±0.4 μm. The change of the polymer deposition
141 density did not alter the coating roughness. At the three Re of the current study, the coatings are considered

142 hydrodynamically smooth since the roughness elements submerge in the viscous sublayer (Wu and
143 Patterson, 1989). For the three Reynolds numbers applied here, the estimated linear viscous sublayer varies
144 from 33 to 59 μm which is an order of magnitude larger than the surface roughness. As it was discussed,
145 the pressure drop measurements is carried out $52H$ downstream of the coated plate. Therefore, the surface
146 topology and roughness of the coating does not have any effect on the measured drag reduction.

147

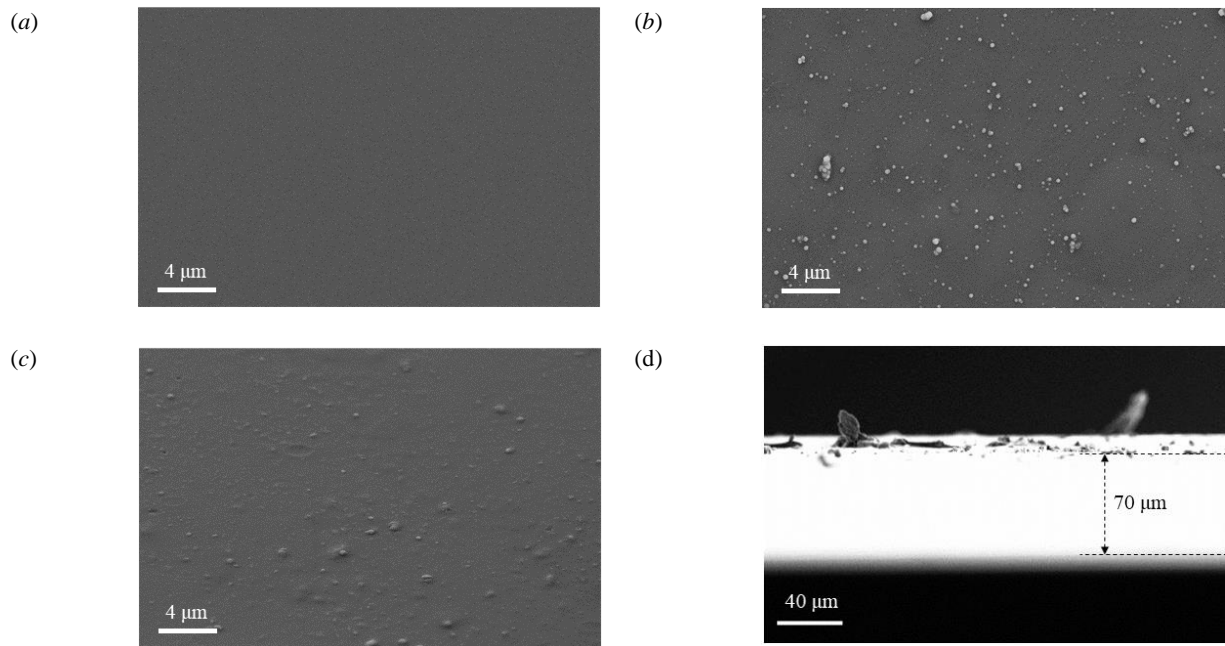


Fig. 2. FESEM images with a top-down view of the (a) silicon wafer, (b) silicon wafer coated with PDA, and (c) silicon wafer coated with PDA and APAM. (d) Side view of the PDA/APAM coating after drying in the oven.

148

149 2.2. Flow facility

150 The experiments were conducted in a closed-loop turbulent channel flow facility. A schematic diagram
151 of the experiment appears in Fig. 3. The rectangular test section of the channel had a height, H , of 6 mm
152 and a width, W , of 40 mm. The relatively large aspect ratio of $W/H = 6.7$ ensures a two-dimensional flow
153 in the center of the channel (Dean, 1978; Vinuesa et al., 2014). The total length of the rectangular test
154 section was $200H$ (1.2 m). A replaceable test plate with dimensions of $240 \times 40 \text{ mm}^2$ in the streamwise and
155 spanwise directions was installed $42H$ (250 mm) downstream of the channel entrance. A groove with

156 dimensions of $236 \times 36 \text{ mm}^2$ was machined on the test-plate to hold the aluminum substrates flush to the
 157 inner surface of the channel. The coated plates were placed close to the entrance to allow the released
 158 polymer to mix with the incoming flow, and form a fully developed flow farther downstream, where the
 159 pressure drop is measured. A bare aluminum substrate was used as the baseline smooth surface. A
 160 centrifugal pump controlled using a variable frequency driver, circulated tap water in the loop. The total
 161 volume of water was 50 liters.

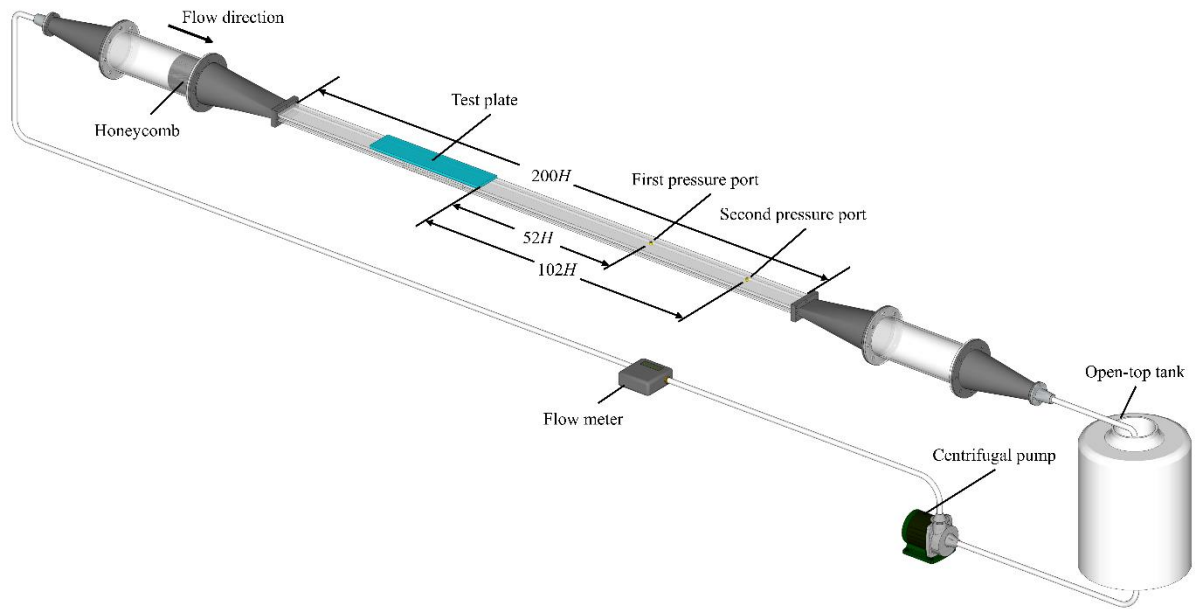


Fig. 3. Schematic diagram of the experimental setup showing the pressure ports and the test plate.

162 To measure the turbulent drag, we incorporated measurements of pressure drop downstream of the test
 163 plate. The two pressure ports were at $130H$ (800 mm) and $180H$ (1100 mm) downstream of the channel
 164 entrance. In wall-bounded experiments with polymer injection, there is an initial region where the injected
 165 polymer mixes with the incoming flow and drag-reduction significantly varies with streamwise distance.
 166 According to the measurement of Tiederman et al., (1985), this region with large variation of drag reduction
 167 is limited to a maximum distance of $30H$ downstream of the polymer injection slot. Beyond the streamwise
 168 location, the variation in drag reduction and the pressure drop are negligible. In the current investigation,
 169 the distance between the downstream edge of the coated surface and the first pressure port is $52H$, which is

170 sufficient to measure the drag-reduction. The second pressure port is also $102H$ downstream of the coated
171 surface. A differential-pressure transducer (P15, Validyne Engineering Corp., Northridge, CA) was
172 connected to a sine wave demodulator (CD15, Validyne Engineering Corp., Northridge, CA) to measure
173 the pressure difference between the two pressure ports. The percentage of DR downstream of the coated
174 surfaces was calculated following

$$\text{DR} = \frac{\Delta P_0 - \Delta P}{\Delta P_0}, \quad (1)$$

175 where ΔP_0 and ΔP are the pressure drops over the baseline and the coated surface, respectively. The
176 average velocity across the cross-section, U , was set to 1.2, 1.6, and 2.3 m s^{-1} , equivalent to Reynolds
177 numbers of 8×10^3 , 11×10^3 , and 16×10^3 . The Reynolds number, Re , is defined as $U H/\nu$. Here, ν is the
178 kinematic viscosity of the water. All three Re correspond to the turbulent regime. To remove any trapped
179 air, the water was circulated in the facility for 15 min. Then the coated surfaces were installed on the test-
180 plate, and water was circulated for an additional minute before recording the pressure data.

181 In the experiments, the measurements of pressure drop indicate the effect of the locally dissolved
182 polymer, not the polymer solution that has circulated back into the channel. This is mainly due to the small
183 amount of dissolved polymer in the bulk fluid and high mechanical degradation of the pump. If the
184 maximum amount of deposited polymer ($\sim 70 \text{ mg}$ based on 0.8 mg cm^{-2}) is dissolved in the loop, the final
185 concentration would be 1.4 wppm, which is small to result in a significant DR. In addition, we used a small
186 centrifugal pump at high rotational speeds to ensure that the polymer solution degrades after circulating
187 through the pump (Den Toonder, 1995).

188 **3. Results**

189 In this section, we first characterize the surface before and after the DR tests using a Fourier transform
190 infrared (FTIR) spectrophotometer. Then we evaluate the drag measurements for a non-coated surface and
191 discuss the drag measurements for the coated surfaces. The error bars in this section indicate the
192 measurement range (minimum to maximum) estimated from at least two independent experiments for each
193 case.

194 **3.1. Surface characterization.**

195 To confirm the polymer grafted to the surface, the coating was characterized by FTIR using an Agilent
196 Technologies Cary 600 Series FTIR spectrometer (ATR mode) between 600 and 3900 cm^{-1} . The ATR-
197 FTIR analysis displayed in Fig. 4(a, b, and c) are for the surface of a pristine aluminum substrate with the
198 PDA coating only, a PDA-coated aluminum surface with a freshly-grafted APAM coating (PDA/APAM),
199 and a PDA/APAM-coated aluminum surface after the drag reduction test, respectively. In the latter case,
200 the surface was exposed to water flow for a long time. As shown in Fig. 4(a) and Fig. 4(b), broad absorbance
201 appears between 3610 and 3720 cm^{-1} , ascribed to the N – H/O – H stretching vibration of the PDA coating.
202 The absorption peak at 1409 cm^{-1} represents the phenolic C – O – H bending vibration. The presence of this
203 characteristic absorbance peak confirmed the presence of a thin PDA layer on the surface (Shah et al.,
204 2019). Based on the previous research of Wang et al., (2017) the absorbance peak at 1257 cm^{-1} was assigned
205 to the N – H scissoring vibrations, while the peak at 1510 cm^{-1} was attributed to the C – O stretching from
206 phenolic moieties. Compared to the PDA coating, the new appearance of an absorbance peak at 1664 cm^{-1}
207 in the PDA/APAM spectrum in Fig. 4(b) corresponds to C = O stretching in amide groups of
208 polyacrylamide which confirms the successful grafting of APAM to the PDA coated surface (Yang et al.,
209 2010). The absorption peaks at 2890 and 2973 cm^{-1} represent C – H stretching of CH_2 . The new absorbance
210 peaks at 1648 and 3439 cm^{-1} in the spectrum of the PDA/APAM after drag reduction, in Fig. 4(c), were
211 attributed to hydration which proves the successful interaction of the APAM with water in the flow channel
212 (Feng et al., 2017). The presence of broad peaks at 3000 and 3600 cm^{-1} in Fig. 4(c) confirms that PDA
213 coating can remain intact after a DR test and creates an opportunity to reuse it to graft APAM again.

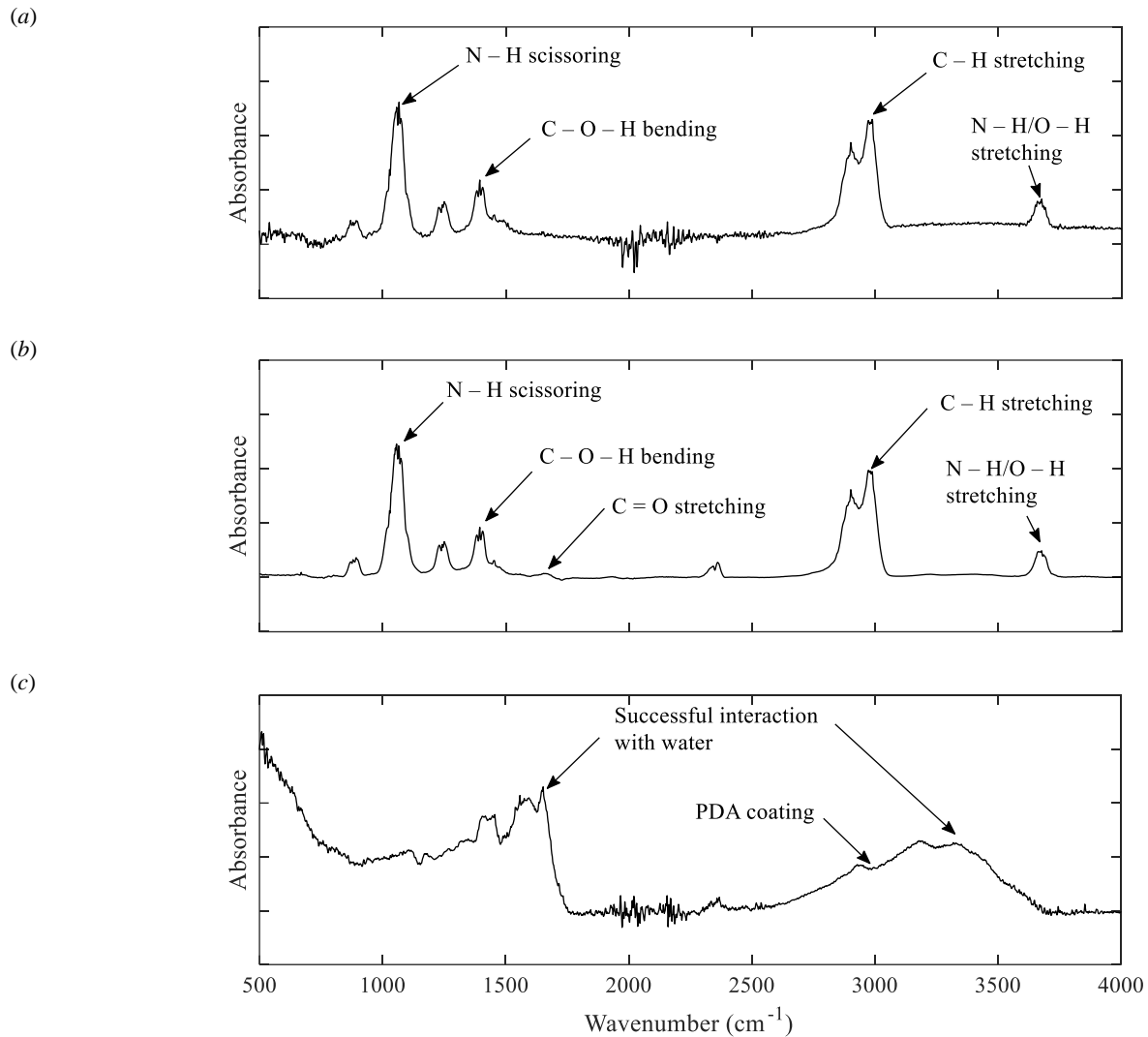


Fig. 4. FTIR measurements of the (a) PDA coating, (b) PDA/APAM coating, and (c) PDA/APAM coating after the DR test.

214

215 The elemental contents on the coated substrate were tested using an energy-dispersive X-ray
 216 spectroscopy (EDX), performed using Bruker XFlash. The EDX analysis in Table 2 shows that after
 217 grafting APAM on the PDA-coated surface, the nitrogen (N) and oxygen (O) content increased significantly
 218 compared to that of the PDA-coated-only surface. The increase of these characteristic contents from APAM
 219 also proves the successful grafting of APAM on the PDA surface.

220

Table 2. Surface elemental composition of the coated surfaces using EDX.

Sample Name	C	O	N	Si
Control (Si wafer)	37.85	-	-	62.15
PDA	73.65	21.68	4.66	-
PDA/APAM	50.53	37.95	11.52	-

221

222 3.2. Drag of the non-coated surface.

223 To evaluate the measurement system and to obtain the baseline drag for comparison with the coated
 224 surfaces, pressure drop measurements were carried out downstream of the non-coated surface. The
 225 measurements are compared with Dean’s correlation for turbulent flow expressed as $f = 0.073(Re)^{-0.25}$. Here
 226 f is the fanning friction factor. As shown in Fig. 5, the friction factors at the three tested Re of 8×10^3 , 11×10^3 ,
 227 and 16×10^3 are comparable to Dean’s correlation. The maximum discrepancy between the current
 228 measurement and Dean’s correlation is about 5% and is mainly associated with the smaller aspect ratio of
 229 the channel ($W/H = 6.7$). The small discrepancy indicates the validity of the current measurements and
 230 confirms that a fully developed turbulent flow was present at the measurement location. The repeatability
 231 of the measurement, shown by error bars, varies by no more than 7%.

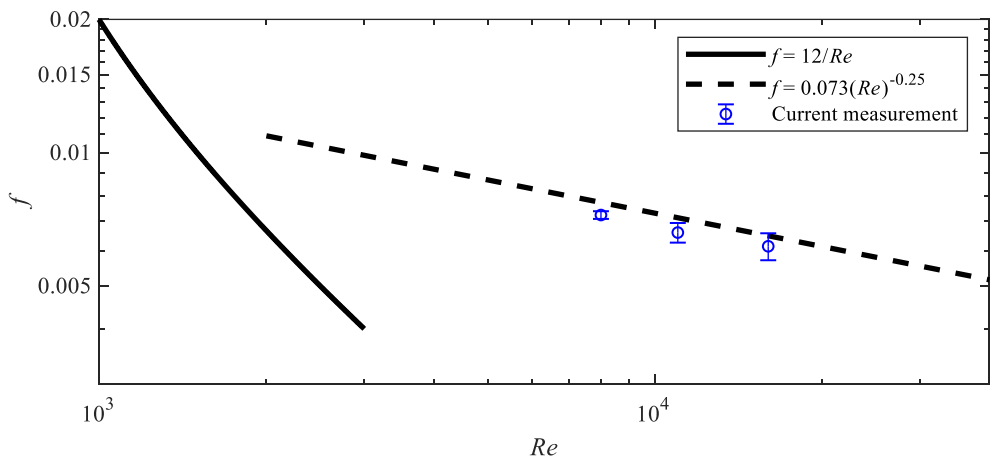


Fig. 5. Friction factor against Reynolds number for the non-coated surface. The Poiseuille equation for the laminar flow ($f=12/Re$) and the Dean’s correlation ($f = 0.073Re^{-0.25}$) are also shown.

232

233 3.3. Drag of the polymer-coated surfaces.

234 The effect of varying the amount of deposited polymer is shown in Fig. 6. In this figure, DR is shown
235 as a function of time, T , during which the coated surfaces was exposed to flow at Re of 16×10^3 . For a
236 polymer deposition of 0.1 mg cm^{-2} , the DR remains negligible, potentially due to the small amount of
237 released polymer. For 0.2 mg cm^{-2} , the DR starts at 5% and quickly reduces to zero after 10 min. For higher
238 deposition densities of 0.3 and 0.4 mg cm^{-2} , DR remains constant at about 15-20% for the first 20 min of
239 the experiments. The DR gradually reduces and after about 50 min approaches zero. In contrast, a different
240 trend is observed for higher deposition densities of 0.6 and 0.8 mg cm^{-2} . At 0.6 mg cm^{-2} , DR starts at about
241 18%, but immediately reduces with increasing time, and reaches zero after 45 min. For the maximum
242 deposition density of 0.8 mg cm^{-2} , DR also starts at about 15% and diminishes rapidly within 10 min. The
243 measurements show that the deposition rate of 0.4 mg cm^{-2} results in the maximum DR percentage and
244 duration. At the end of the tests, the APAM coating was completely dissolved in water, while the PDA
245 layer was still available for re-grafting a new layer of APAM if necessary.

246 Three observations are made from Fig. 6. First, the variation of DR with the amount of deposited polymer
247 indicates that the deposited amount affects the release rate of the polymer into the water. The polymer
248 release rate affects the local polymer concentration and therefore the DR. Second, an optimum amount of
249 deposited polymer can result in a polymer release rate that maximizes DR amount and duration. If a larger
250 amount of polymer is deposited in the coating, DR and its duration decrease. It is hypothesized that for
251 larger amounts of deposited polymer ($D = 0.6$ and 0.8 mg/cm^2), the adhesion of the APAM chains to the
252 PDA layer is weaker. Therefore, the *loose* APAM polymers that are not bonded to the PDA layer, quickly
253 release into water and deplete the polymer supply. This results in the larger initial DR followed by a fast
254 decrease in DR observed for $D = 0.6$ and 0.8 mg/cm^2 cases. Third, the decrease in DR with time indicates
255 that the release rate of the polymer coating also varies in time. The trends show that as the polymer coating
256 erodes and dissolves into the flow, the release rate decreases. It is conjectured that the top APAM layers
257 release quickly, while the bottom APAM layers that are closer to the PDA layer release slower. In general,
258 the deposited amount of APAM controls both the DR percentage and its duration.

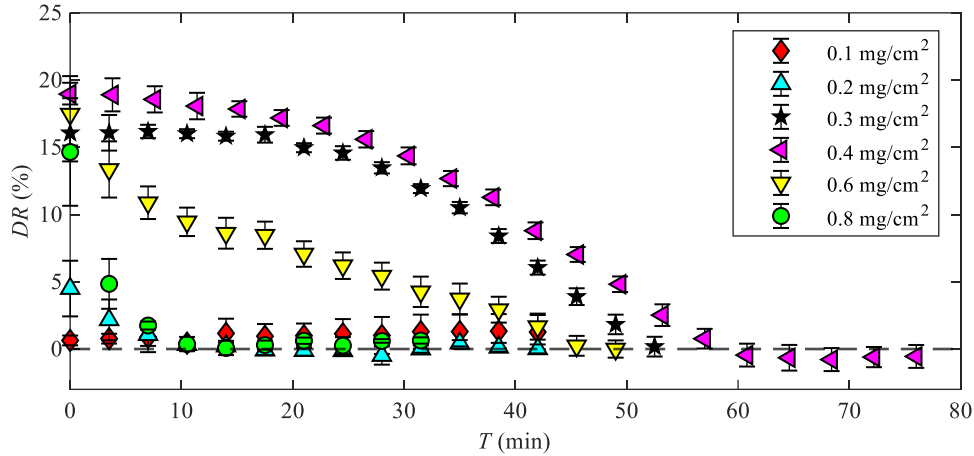


Fig. 6. Effect of APAM deposition on DR at $Re = 16 \times 10^3$.

259 To demonstrate the effect of the deposited APAM amount on the initial DR, the DR averaged over the
 260 first 10 min, DR_0 , is shown in Fig. 7 for various values of D . As can be seen, there is a clear trend of
 261 increasing DR_0 with increasing deposition reaching up to $\sim 19\%$ at 0.4 mg cm^{-2} . After this point, higher
 262 deposition does not increase DR_0 , and it reduces to $\sim 5\%$ at 0.8 mg cm^{-2} . As we discussed above, this trend
 263 is potentially associated with a quick depletion of the APAM layer due to the release of the loose polymers
 264 that are present in the thicker layers. Another potential cause of this trend can be the saturation phenomenon
 265 reported by Lumley, (1973). Winkel et al., (2009) reported that an excessive polymer concentration
 266 increases the drag owing to greater solution viscosity. For example, Warholic et al., (1999) observed 69%
 267 DR for an injected polymer concentration of 500 wppm, while a smaller DR of 52% was observed for
 268 1000 wppm. In the current experiment, the excess polymer concentration may not be a factor since the
 269 overall DR is smaller than 20%, suggesting that the local viscosity has not reached the saturation limit. In
 270 an experimental investigation using a similar polymer, Shaban et al. (2018) measured DR of 25, 43, 51, and
 271 57% for homogeneous polymer solutions with concentrations of 10, 20, 90, and 150 ppm, respectively.
 272 Therefore, for the smaller DR of the current experiment, the local polymer concentration is expected to be
 273 smaller than 10 ppm.

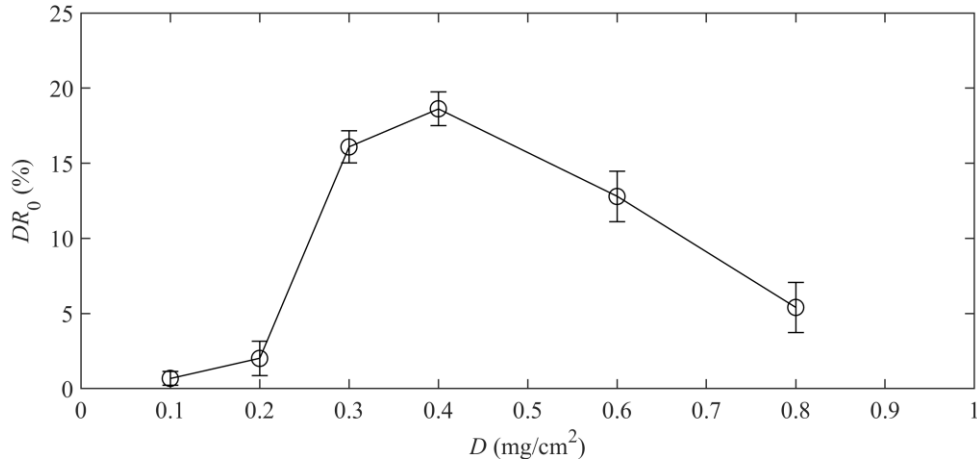


Fig. 7. Drag reduction averaged over the first 10 min.

274 The variation of Re is expected to affect the release and dissolution rates of the polymer layer by
 275 changing the shear stress applied to the polymer coating. To investigate the effect of Re , the coatings were
 276 tested at $Re = 8 \times 10^3$, 11×10^3 and 16×10^3 for an initial APAM deposition of 0.6 mg cm^{-2} . It is worth noting
 277 here that the polymer chain scission (polymer degradation) due to the greater shear rate of the high Re flows
 278 can decrease the DR (Feng et al., 2017). Based on the analysis of Vanapalli et al., (2005), at the wall-shear-
 279 rate of the largest Re of 16×10^3 , the molar mass of the polymers reduces to approximately 8 Mg/mol. At
 280 the two smaller Re , the wall-shear-rate does not reduce the molecular mass. As seen in Fig. 8, for all the
 281 three Re , the DR gradually decreases in time. For $Re = 8 \times 10^3$, the DR started at 13% and reduces to zero
 282 after 35 min. Based on visual inspection of the substrate, we observed that a residual layer of APAM
 283 remained attached to the substrate at $T = 35$ min. Further exposure to the flow at $Re = 8 \times 10^3$ resulted in a
 284 slow loss of the APAM layer without any considerable DR. Therefore, the shear of the flow was not
 285 adequate to efficiently release the APAM coating into the flow. At $Re = 8 \times 10^3$, visual inspection of the
 286 surface showed that after 100 min the entire APAM layer was removed from the surface. However, the DR
 287 was negligible beyond the initial 35 min, potentially due to the small polymer release rate.

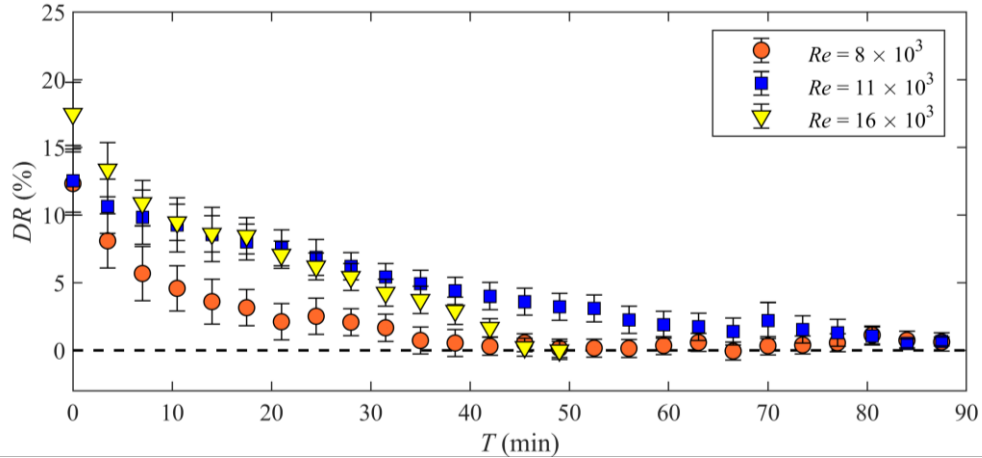


Fig. 8. The effect of Re on the DR over time for a surface coated with 0.6 mg cm^{-2} of APAM.

288 The initial DR for $Re = 11 \times 10^3$ and $Re = 8 \times 10^3$ at $T = 0$ are similar and about 12%. In contrast, at
 289 $Re = 11 \times 10^3$, DR persists for a longer time and extends up to approximately 80 min. It is conjectured that
 290 the DR is larger for $Re = 11 \times 10^3$ relative to $Re = 8 \times 10^3$ due to the higher polymer release rate; the higher
 291 flow shear increases the release rate of the APAM coating. Another possibility that may explain the increase
 292 in DR with increasing Re is reduction of local viscosity due to two mechanisms: (a) the local viscosity at
 293 the larger Re is smaller since the released polymer mixes with more fluid, (b) the higher shear-rate results
 294 in a smaller local viscosity due to the shear thinning behavior of polymeric solutions. However, as it was
 295 discussed, the local viscosity of the fluid is expected to be small due to the low DR. The maximum initial
 296 DR is obtained at $Re = 16 \times 10^3$. However, after 8 min of testing, the DR at $Re = 16 \times 10^3$ approaches that of
 297 $Re = 11 \times 10^3$ and remains comparable until 25 min. Visual inspection of the substrate showed that, after
 298 50 min, the APAM layer was completely removed from the coated substrate, which is consistent with the
 299 observed zero DR. The results indicate that the maximum DR percentage and duration are obtained when
 300 the flow releases APAM at an optimum rate. For the current experimental condition, $Re = 16 \times 10^3$ results
 301 in a larger initial DR while $Re = 11 \times 10^3$ extends the DR over a longer period for the surface with $D = 0.6$
 302 mg/cm^2 .

303 4. Conclusion

304 We developed and tested a novel polymer coating strategy utilizing dual layers of polydopamine (PDA)
305 and anionic polyacrylamide (APAM). The layer of PDA acted as a primary coating on the surface, allowing
306 to subsequently graft a layer of APAM over it. The PDA layer was obtained by immersing the cleaned
307 substrate in a container filled with dopamine hydrochloride solution for 12 hours. A layer of APAM solution
308 was then applied over a freshly prepared and sintered PDA-coated surface using a film coating technique.
309 The coated surface was dried in oven before any characterization or testing was carried out. To confirm the
310 presence of the dual polymer layers on the substrate, the coated surfaces were first characterized with ATR-
311 FTIR. The results proved the presence of both the PDA and APAM coatings, revealing their characteristic
312 absorbance peaks. Field emission scanning electron microscopy images were then taken to analyze the
313 coating topography and thickness. The images showed that the coating did not introduce any large
314 roughness features relative to the base surface. The root-mean-square roughness of the coating was
315 approximately $5.9\pm 0.4\ \mu\text{m}$ for different amounts of deposited APAM. The thickness of the two-layer
316 coating after drying was $\sim 70\ \mu\text{m}$ for maximum APAM deposition of $0.8\ \text{mg cm}^{-2}$.

317 The drag of a turbulent channel flow downstream of the coated surfaces was monitored using pressure
318 drop measurements carried out at streamwise distances of $52H$ and $102H$ downstream of the coated surface.
319 We varied the amount of deposited APAM from 0.1 to $0.8\ \text{mg cm}^{-2}$, and operated the channel at Reynolds
320 numbers, Re , of 8×10^3 , 11×10^3 , and 16×10^3 . The results showed that $0.4\ \text{mg cm}^{-2}$ of APAM resulted in the
321 highest initial drag reduction (DR) of 19%, which gradually reduced to zero over an hour. For smaller
322 APAM depositions of 0.1 and $0.2\ \text{mg cm}^{-2}$, the initial DR was less than 5% and diminished within 10 min.
323 An initial DR of approximately 15% was observed for larger deposition amounts of 0.6 and $0.8\ \text{mg cm}^{-2}$,
324 but the DR diminished faster relative to the $0.4\ \text{mg cm}^{-2}$ case. The shorter duration of DR for larger polymer
325 depositions is mainly associated with a weaker adhesion between APAM and PDA layers, which resulted
326 in a fast deposition of the APAM layer. Therefore, the results show that optimum polymer release-rate can
327 be obtained by controlling the amount of deposited polymer. The polymer release rate also varied with Re .
328 At the lowest Re of 8×10^3 , a sufficient amount of APAM was not released into the flow and the resulting
329 DR was small. At the highest Re of 16×10^3 , a large initial DR was observed, but it quickly diminished as

330 the deposited polymer depleted. A slightly smaller initial DR was observed for a Re of 11×10^3 , however,
331 the DR lasted longer. In general, the results show that the release rate of the polymer, and therefore the DR,
332 is a function of the amount of deposited polymer and the flow Re . The deposited polymer amount should
333 be adjusted to obtain the optimum release rate that results in maximum DR.

334 The developed polymer coating technique showed a considerable DR, offering a cost-effective
335 alternative to the injection of water-soluble polymers. In the current investigation, the DR lasted for about
336 an hour; therefore, the application of this coating is currently limited to systems in which a large DR is
337 sought for a short time-period. Further optimization of the coating is needed to increase its longevity and
338 extend its application. Moreover, due to the dynamic nature of the PDA, some catechol or hydroxyl group
339 will always be available for the re-grafting of APAM or any other polymer onto the PDA-coated surface,
340 which makes this approach a quick and easy way to re-engineer the surface if required.

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344 **6. References**

- 345 Abu Rowin, W., Sean Sanders, R., Ghaemi, S., 2018. A Recipe for Optimum Mixing of Polymer Drag
346 Reducers. *J. Fluids Eng.* 140, 1–10. <https://doi.org/10.1115/1.4040109>
- 347 Berman, N.S., 1986. Velocity fluctuations in non-homogeneous drag reduction. *Chem. Eng. Commun.* 42,
348 37–51. <https://doi.org/10.1080/00986448608911735>
- 349 Bernsmann, F., Ball, V., Addiego, F., Ponche, A., Michel, M., Gracio, J.J.D.A., Toniazzo, V., Ruch, D.,
350 2011. Dopamine-melanin film deposition depends on the used oxidant and buffer solution. *Langmuir*
351 27, 2819–2825. <https://doi.org/10.1021/la104981s>
- 352 Chang, C.C., Kolewe, K.W., Li, Y., Kosif, I., Freeman, B.D., Carter, K.R., Schiffman, J.D., Emrick, T.,
353 2016. Underwater Superoleophobic Surfaces Prepared from Polymer Zwitterion/Dopamine Composite
354 Coatings. *Adv. Mater. Interfaces* 3. <https://doi.org/10.1002/admi.201500521>
- 355 Dean, R.B., 1978. Reynolds Number Dependence of Skin Friction and Other Bulk Flow Variables in Two-
356 Dimensional Rectangular Duct Flow. *J. Fluids Eng.* 100, 215–223. <https://doi.org/10.1115/1.3448633>
- 357 Den Toonder, J.M.J., 1995. Degradation Effects of Dilute Polymer Solutions on Turbulent Drag Reduction

358 in Pipe Flows 63–82.

359 Escudier, M.P., Nickson, A.K., Poole, R.J., 2009. Turbulent flow of viscoelastic shear-thinning liquids
360 through a rectangular duct: Quantification of turbulence anisotropy. *J. Nonnewton. Fluid Mech.* 160,
361 2–10. <https://doi.org/10.1016/j.jnnfm.2009.01.002>

362 Feng, L., Zheng, H., Gao, B., Zhang, S., Zhao, C., Zhou, Y., Xu, B., 2017. Fabricating an anionic
363 polyacrylamide (APAM) with an anionic block structure for high turbidity water separation and
364 purification. *RSC Adv.* 7, 28918–28930. <https://doi.org/10.1039/C7RA05151D>

365 Gibson, C.T., Ridings, C.R., Blok, A.J., Shearer, C.J., Andersson, G.G., Ellis, A. V., 2019. Morphological
366 changes of sintered polydopamine coatings. *Surf. Topogr. Metrol. Prop.* 7.
367 <https://doi.org/10.1088/2051-672X/ab06eb>

368 Hoyer, K., Gyr, A., 1996. Turbulent velocity field in heterogeneously drag reduced pipe flow. *J.*
369 *Nonnewton. Fluid Mech.* 65, 221–240. [https://doi.org/10.1016/0377-0257\(96\)01460-7](https://doi.org/10.1016/0377-0257(96)01460-7)

370 Jiang, J., Zhu, Liping, Zhu, Lijing, Zhu, B., Xu, Y., 2011. Surface characteristics of a self-polymerized
371 dopamine coating deposited on hydrophobic polymer films. *Langmuir* 27, 14180–14187.
372 <https://doi.org/10.1021/la202877k>

373 Lee, H., Dellatore, S.M., Miller, W.M., Messersmith, P.B., 2007. Mussel-inspired surface chemistry for
374 multifunctional coatings. *Science.* 318, 426–430. <https://doi.org/10.1126/science.1147241>

375 Liu, M., Zeng, G., Wang, K., Wan, Q., Tao, L., Zhang, X., Wei, Y., 2016. Recent developments in
376 polydopamine: An emerging soft matter for surface modification and biomedical applications.
377 *Nanoscale.* <https://doi.org/10.1039/c5nr09078d>

378 Lumley, J.L., 1973. Drag Reduction in Turbulent Flow by Polymer Additives. *J. Polym. Sci. Macromol.*
379 *Rev.* 7, 263–290.

380 Motozawa, M., Ito, T., Matsumoto, A., Ando, H., Ashida, T., Senda, T., Kawaguchi, Y., 2010. Turbulent
381 Drag Reduction by Polymer Containing Paint: Simultaneous Measurement of Skin Friction and
382 Release Rate 1–9.

383 Ptasinski, P.K., Nieuwstadt, F.T.M., van den Brule, B.H.A.A., Hulsen, M.A., 2001. Experiments in
384 turbulent pipe flow with polymer additives at maximum drag reduction. *Flow, Turbul. Combust.* 66,
385 159–182. <https://doi.org/10.1023/A:1017985826227>

386 Ryu, J.H., Messersmith, P.B., Lee, H., 2018. Polydopamine Surface Chemistry: A Decade of Discovery.
387 *ACS Appl. Mater. Interfaces.* <https://doi.org/10.1021/acsami.7b19865>

388 Semenov, B.N., 1991. The pulseless injection of polymeric additives into near-wall flow and perspectives
389 of drag reduction. pp. 293–308. https://doi.org/10.1007/978-94-011-3526-9_15

390 Shaban, S., Azad, M., Trivedi, J., & Ghaemi, S. (2018). Investigation of near-wall turbulence in relation to
391 polymer rheology. *Physics of Fluids*, 30(12), 125111.

392 Shah, A.A., Cho, Y.H., Choi, H. gyu, Nam, S.E., Kim, J.F., Kim, Y., Park, Y.I., Park, H., 2019. Facile
393 integration of halloysite nanotubes with bioadhesive as highly permeable interlayer in forward osmosis
394 membranes. *J. Ind. Eng. Chem.* 73, 276–285. <https://doi.org/10.1016/j.jiec.2019.01.039>

395 Tiederman, W.G., Luchik, T.S., Bogard, D.G., 1985. Wall-layer structure and drag reduction. *J. Fluid*
396 *Mech.* 156, 419–437. <https://doi.org/10.1017/S0022112085002178>

397 Toms, B.A., 1948. Some observations on the flow of linear polymer solutions through straight tubes at large
398 Reynolds numbers. *Proc. 1st Int. Congr. Rheol.* 2, 135–141.

399 Vanapalli, S.A., Islam, M.T., Solomon, M.J., 2005. Scission-induced bounds on maximum polymer drag
400 reduction in turbulent flow 095108. <https://doi.org/10.1063/1.2042489>

401 Vinuesa, R., Noorani, A., Lozano-durán, A., Khoury, G.K. El, Schlatter, P., Fischer, P.F., Nagib, H.M.,
402 Vinuesa, R., Noorani, A., Lozano-durán, A., Khoury, G.K. El, 2014. Aspect ratio effects in turbulent
403 duct flows studied through direct numerical simulation 5248.
404 <https://doi.org/10.1080/14685248.2014.925623>

405 Virk, P.S., Merrill, E.W., Mickley, H.S., Smith, K.A., Mollo-Christensen, E.L., 1967. The Toms
406 phenomenon: Turbulent pipe flow of dilute polymer solutions. *J. Fluid Mech.* 30, 305–328.
407 <https://doi.org/10.1017/S0022112067001442>

408 Wang, K., Dong, Y., Yan, Y., Zhang, S., Li, J., 2017. Mussel-inspired chemistry for preparation of
409 superhydrophobic surfaces on porous substrates. *RSC Adv.* <https://doi.org/10.1039/c7ra04790h>

410 Warholic, M.D., Massah, H., Hanratty, T.J., 1999. Influence of drag-reducing polymers on turbulence:
411 effects of Reynolds number, concentration and mixing. *Exp. Fluids* 27, 461–472.
412 <https://doi.org/10.1007/s003480050371>

413 Wei, T., Willmarth, W.W., 1992. Modifying turbulent structure with drag-reducing polymer additives in
414 turbulent channel flows. *J. Fluid Mech.* 245, 619. <https://doi.org/10.1017/S0022112092000600>

415 White, C.M., Mungal, M.G., 2008. Mechanics and Prediction of Turbulent Drag Reduction with Polymer
416 Additives. *Annu. Rev. Fluid Mech.* 40, 235–256.
417 <https://doi.org/10.1146/annurev.fluid.40.111406.102156>

418 Willmarth, W.W., Wei, T., Lee, C.O., 1987. Laser anemometer measurements of Reynolds stress in a
419 turbulent channel flow with drag reducing polymer additives. *Phys. Fluids* 30, 933.
420 <https://doi.org/10.1063/1.866278>

421 Winkel, E.S., Oweis, G.F., Vanapalli, S.A., Dowling, D.R., Perlin, M., Solomon, M.J., Ceccio, S.L., 2009.
422 High-Reynolds-number turbulent boundary layer friction drag reduction from wall-injected polymer
423 solutions. *J. Fluid Mech.* 621, 259–288. <https://doi.org/10.1017/S0022112008004874>

424 Wu, H., Patterson, G.K., 1989. Laser-Doppler Measurements Of Turbulent-Flow Parameters In A Stirred
425 Mixer. *Chem. Eng. Sci.* 44, 2207–2221.

- 426 Yang, J., Saggiomo, V., Velders, A.H., Stuart, M.A.C., Kamperman, M., 2016. Reaction pathways in
427 catechol/primary amine mixtures: A window on crosslinking chemistry. *PLoS One* 11.
428 <https://doi.org/10.1371/journal.pone.0166490>
- 429 Yang, W.J., Park, H., Hwan Chun, H., Ceccio, S.L., Perlin, M., Lee, I., 2014. Development and performance
430 at high Reynolds number of a skin-friction reducing marine paint using polymer additives. *Ocean Eng.*
431 84, 183–193. <https://doi.org/10.1016/j.oceaneng.2014.04.009>
- 432 Yang, Z., Wang, J., Luo, R., Maitz, M.F., Jing, F., Sun, H., Huang, N., 2010. The covalent immobilization
433 of heparin to pulsed-plasma polymeric allylamine films on 316L stainless steel and the resulting effects
434 on hemocompatibility. *Biomaterials*. <https://doi.org/10.1016/j.biomaterials.2009.11.091>
- 435 Zeng, Y., Liu, W., Wang, Z., Singamaneni, S., Wang, R., 2018. Multifunctional Surface Modification of
436 Nanodiamonds Based on Dopamine Polymerization. *Langmuir* 34, 4036–4042.
437 <https://doi.org/10.1021/acs.langmuir.8b00509>