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GRAFTED NATURAL POLYMER AS NEW DRAG REDUCING AGENT: AN EXPERIMENTAL APPROACH

The present investigation introduces a new natural drag reducing agent which has the ability to improve the flow in pipelines carrying aqueous or hydrocarbon liquids in turbulent flow. Okra (Abelmoschus esculentus) mucilage drag reduction performance was tested in water and hydrocarbon (gas-oil) media after grafting. The drag reduction test was conducted in a buildup closed loop liquid circulation system consisting of two pipes of inside diameters (ID) 0.0127 and 0.0381 m, four testing sections in each pipe (0.5 to 2.0 m), tank, pump and pressure transmitters. Reynolds number (Re), additive concentration and the transported media type (water and gas-oil), were the major drag reduction variables investigated. The experimental results show that new additive drag reduction ability is high, with maximum achieved percentage of drag reduction (%Dr) up to 60%. The experimental results showed that the drag reduction ability increased by increasing the additive concentration. The %Dr was found to increase by increasing Re by using the water-soluble additive while it was found to decrease by increasing Re when using the oil-soluble additive. The %Dr was higher in the 0.0381 m ID pipe. Finally, the grafted and natural mucilage showed high resistance to shear forces when circulated continuously for 200 s in the closed-loop system.

Keywords: natural drag reduction agent; percentage drag reduction; pipeline system; turbulent flow.

Transporting liquids through pipelines is considered one of the most power consuming applications in the industry due to the turbulent mode these liquids are transported within. Introducing supporting pumping stations along the way of the pipeline was one of the solutions applied. However, increasing the number of supporting pumping stations will increase the production and transportation cost. An alternative technique, suggested later by many authors, was the injection of viscoelastic additives to improve the flow in pipelines (drag reduction). The phenomenon of reducing the drag by the addition of viscoelastic additives was discovered in the early 1940s by Toms [1]. He stated that the addition of small amounts of high molecular weight polymer solvent can significantly reduce frictional pressure drop in turbulent flows leading to maintain the flow energy resulting increment in pipeline capacities. This discovery was applied by many researchers in a wide variety of industrial applications such as oil transportation, heat and mass transfer applications and firefighting [2-3].

Artificial polymers were suggested as drag reduction agents (DRA) by many researchers [4-15]. Yet, most of the polymers used as DRAs are synthetic polymers, which are used as flocculation agents in some applications [16]. Most artificial polymers are not biodegradable and cannot be considered as an environmentally friendly product, and they have low resistance to shear forces exerted by the turbulent structure inside the pipe during the transportation. All this encouraged a large number of researchers to replace the existing artificial polymeric additives by natural biodegradable polymers or insoluble additives. Natural polymers were introduced in several occasions to replace the existing synthetic ones and as

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an environmentally friendly solution [17-19]. Most of the investigated and discovered natural DRAs are water soluble and driven from agricultural sources such as guar gum [20-21], Aloe Vera [22], cocoa husk [23] and xanthan gum [24].

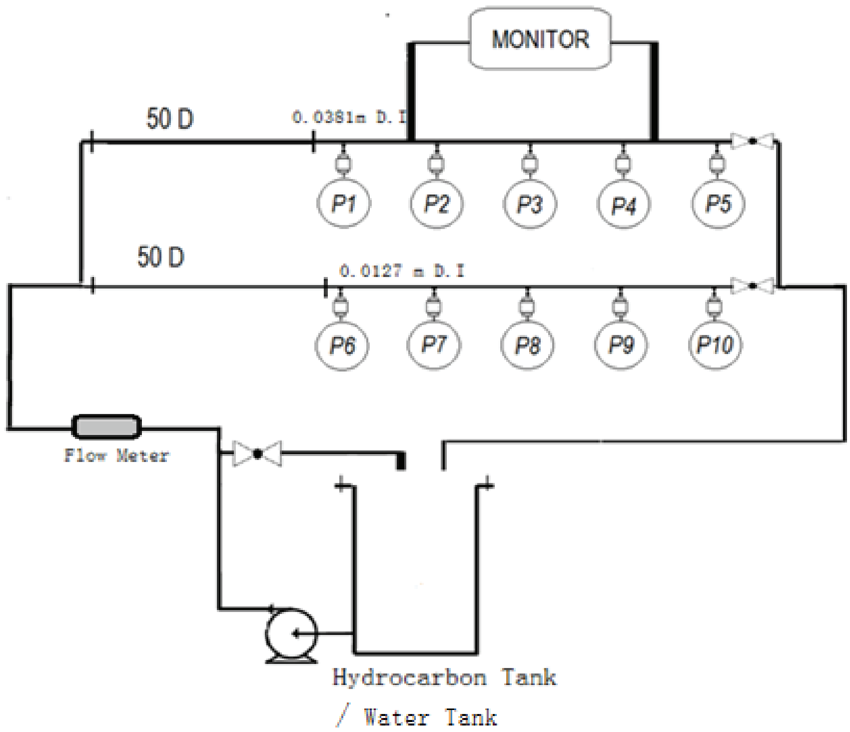
In the present work, a new DRA will be introduced. The new additive is extracted from hibiscus leaves. The extracted mucilage is water soluble. The mucilage solubility will be changed by applying polymer grafting technique to make it soluble in the gas-oil (hydrocarbon media). The effect of solution flow rate, additive concentration, pipe diameter and testing section length on the drag reduction performance will be examined.

MATERIALS AND METHOD

Liquid circulation system

The drag reduction experimental study for four independent variables, namely pipe diameter (*D*), testing section length, volumetric flow rate (with reference to Reynolds number) and the additive concentrations, was carried out in a built up closed loop liquid circulatory system. Figure 1 shows the schematic diagram of the closed loop liquid circulation system used to test the drag reduction phenomenon.

The rig mainly consists of a liquid tank, Precision pump model PPM-158 with maximum load 10.5 m^3/h and pipes of diameters of 0.0381 and 0.0127 m. The tank connected with the recirculation pipe used to control the liquid flow rate entering the system. Each testing section is supported with a ball valve. These valves are used to close and open the testing section stream and not to control the flow rate entering to the section in order not to disturb the developed turbulent flow entering to each testing section. In each pipe, the first testing point starts after 50*D* (*D* = diameter of pipe) to ensure the flow is constant turbulent. After 50*D* is where the first transmitter sensor is located, *i.e.*, 1.91 and 0.635 m for pipe with ID of 0.0381 and 0.0127 m, respectively.



Instruments	
P1-P5	Pressure sensor for 0.0381 m id
P6-P10	Pressure sensor for 0.0127 m id
	Valve
	Centrifugal pump

Figure 1. Schematic diagram of closed loop circulation system.

The maximum capacity of the liquid tank is 180 l (0.5 m×0.6 m×0.6 m). The main piping system starts by connecting the liquid tank to the pump by 0.0381 m ID pipe as the first delivering exit. Then the piping system delivers the liquid pump out through 0.55 m horizontal pipe with 0.0381 m ID. This is where the ultrasonic flow meter located before the liquids transferred through vertical pipe of 0.0381 m ID. Flow rates were measured using a Burket 8035 minisonic flow meter. The power supply for this detector is 12 to 30 V and can measure point pressure within the range of 9.65-15.86 bar (140-230 psi) with fluid viscosity less than 3×10^{-4} m^2/s (300 cSt). From this vertical pipe, liquids are delivered into two different pipes each with ID of 0.0381 and 0.0127 m, respectively. The testing section is made of four parts, each 0.5 m in length. Each pipe is supported with five pressure measurement holes. A pressure transmitter sensor is located at each pressure testing hole. All the pressure transmitters are connected to an interface that gives computational pressure readings straight to the computer. The pressure measurements are taken every two seconds. SCADA interface was designed and installed on the computer for the pressure reading visualization in each testing point.

Material Used

Raw material

The scientific name of okra is *Abelmoschus esculentus* and it is occasionally referred to as *Hibiscus esculentus* L. The type of okra used was Clemson Spineless okra which is usually found in Malaysia or other equatorial climates countries. Okra mucilage is a polysaccharide derived from plant exudates that give gelling and adhesive effects. Ganji *et al.* [25] reported that okra mucilage is water soluble, consisting of 1.7 million molecular weight of glycoprotein which produces viscous, shear thinning and visco-elastic solutions in water. Figure 2 shows the molecular structure of the polysaccharide in the okra mucilage.

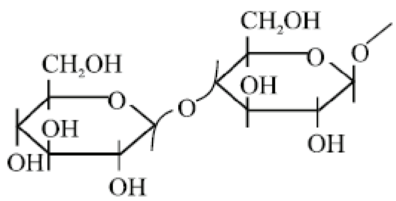


Figure 2. Molecular structure of okra mucilage (monosaccaride).

Monomer

Acrylonitrile is a monomer used to graft with free radical polysaccharide obtained from the okra mucilage. Acrylonitrile can be intact with polysaccharide free radical backbone by the presence of a vinyl group in acrylonitrile [26]. Acrylonitrile is widely used in plasticizing manufacturing processes. The other regulatory name for acrylonitrile is 2-Propenenitrile, molecular structure of C_3H_3N. The density of acrylonitrile is 0.81 g/cm^3 at 25 °C with molecular weight of 53.1 g/mol. The boiling and melting points of this chemical compound are 77.3 and -82 °C, respectively. The vapor pressure is 100 Torr at 23 °C with a conversion factor of 2.17 mg/m^3 at 25 °C for 1 ppm concentration. This compound is colorless, has a pungent onion-garlic smell and is soluble in isopropanol, ethanol, ether, acetone, and benzene. Figure 3 shows the molecular structure of acrylonitrile.

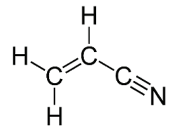


Figure 3. Molecular structure of acrylonitrile.

Solvent

N,*N*-Dimethyl formamide was used to dissolve the resulting grafted polymeric powder and make it soluble in the hydrocarbon media. The other regulatory name for *N*,*N*-dimethyl formamide is *N*,*N*-dimethylmethanamide, molecular formula of C_3H_7NO. The compound density is 0.944 g/cm^3 in liquid form at 25 °C and the viscosity is 0.92 cP (9.2×10^{-4} kg/m.s) at 20 °C. The boiling and melting points of this chemical compound are 153 and -61 °C, respectively. The vapor pressure of this compound is 0.3 kPa at 20 °C, with a conversion factor of 9.44 mg/m^3 at 25 °C for 1 ppm concentration. This solvent is colorless and odorless, except for fishy smell due to impurities of dimethylamine. Figure 4 shows the molecular structure of the N, N dimethyl formamide.

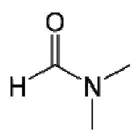


Figure 4. Molecular structure of N,N-dimethyl formamide.

Methodology

Extraction method

Extraction can be done by immersing chopped okra (1:3 by weight) into distilled water. This okra is soaked overnight to mangle okra mucilage out from the okra pod. The clear water will turns into a very viscous thick solution, indicating that the mucilage is ready to be used. For water as the transported liquid, this mucilage will be used as drag reduction agent.

However, when hydrocarbon is used as transported liquid, okra mucilage will be grafted in order to be inverted into hydrocarbon soluble products.

Grafting method

The grafting method [25] in this research involved okra mucilage as natural polymer and acrylonitrile. The initiator in this free-radical polymerization is nitric acid. The reaction occurs in nitrogen conditions. 800 m of distilled water solution of okra mucilage are flushed with nitrogen for 20 min. This is to remove oxygen from free radical sites instigated at the backbone of the okra mucilage polymer. 0.23 mol of acrylonitrile is added, and stirring is required for 30 min before the solution can be bubbled with nitrogen for another 20 min. The last step before the start of the reaction is the injection of 15 ml of 1 mol of nitric acid containing ceric ions. The reaction takes place at 70 °C for 24 h in a water bath. The reaction is stopped by adding 8 ml of concentrated hydroquinone. The product is washed with acetone and precipitate. This step is required to remove excess and unbound acrylonitrile since acetone can be soluble into this monomer. Then, a cake is formed and slurred by isopropanol for a while before precipitating again for reducing time required for the drying process. This step also is important to remove unbound polysaccharide free radicals in order to ensure the free radical site and backbone of polymer are not associated with oxygen that can cause biodegradation. This filtrated cake is dried in an oven at 40 °C. This powder is dissolved for 24 h in dimethyl formamide to become liquid. This is used as new formulated polymeric DRA.

Transported liquid

The transported liquid used in the present investigation was diesel obtained from Shell and pure water. The tested physical properties of hydrocarbon liquid and water were viscosity and density. The viscosities of hydrocarbon liquid and water were 3×10^{-3} and 8.6×10^{-4} Pa s, and the densities were 853.2 and 996 kg/m^3, respectively.

Experiment progress

All the experiments were carried in a constructed liquid circulation system, testing different variables:

- Concentrations: 50, 100, 200, 400, 600, 800 and 1000 ppm.
- Liquid flow rate: 5 to 8.5 m^3/h.
- Pipe length: 0.5, 1.0, 1.5 and 2.0 m.
- Pipe diameter: 0.0127 and 0.0381 m ID.

The experimental procedure starts by testing every pipe length and pipe diameter, the operation begins when the pump starts delivering the solution through the testing section. The solution flow rate is fixed at the certain value by controlling it from the bypass section. Pressure readings are taken according to testing section. Changing the solution flow rate to another fixed point, pressure readings are taken again until certain pressure limit. This procedure is repeated for each pipe diameter and concentration to test its effect on the drag reduction operation.

Experimental calculation

Percentage dag reduction calculations. Pressure drop readings through testing sections, before and after drag reducer addition, were needed to calculate the percentage drag reduction, %Dr, as:

$$\%DR = \frac{\Delta p_b - \Delta p_a}{\Delta p_b} \quad (1)$$

where Δp_b and Δp_a are pressure drops before and after the addition of DRA, respectively.

RESULT AND DISCUSSION

In order to verify the built up experimental closed-loop system used, the friction factor data for the flow of the additive-free solutions (hydrocarbon liquid) was calculated and compared with the Blasius asymptote equation. The data obtained are plotted in Figure 5. The Blasius correlation is defined by the following equation:

$$f = 0.0791\mathrm{Re}^{-0.25} \quad (2)$$

Furthermore, the maximum *%Dr* asymptote suggested by Virk [27] is presented. Virk's correlation can be defined by the following equation:

$$f = 0.59\mathrm{Re}^{-0.58} \quad (3)$$

For determination of laminar flow, friction factor for laminar flow in wall bounded channel was defined as:

$$f = \frac{16}{\mathrm{Re}} \quad (4)$$

Figures 5 and 6 show the verification of the transport media. It can be noticed that the values of friction factor against fluid Reynolds number of water are close to the Blasius asymptote which proves that the built up closed loop circulation system is appropriate for the purpose of this study.

Figures 7 and 8 show the drag reduction performance of grafted mucilage with gas-oil and water media, respectively. The additive showed good drag reduction ability within the operating conditions investigated. Generally, Figure 7 shows that a maximum *%Dr* of 55% was achieved by the addition of 1000 ppm of the grafted mucilage to the diesel. Also,

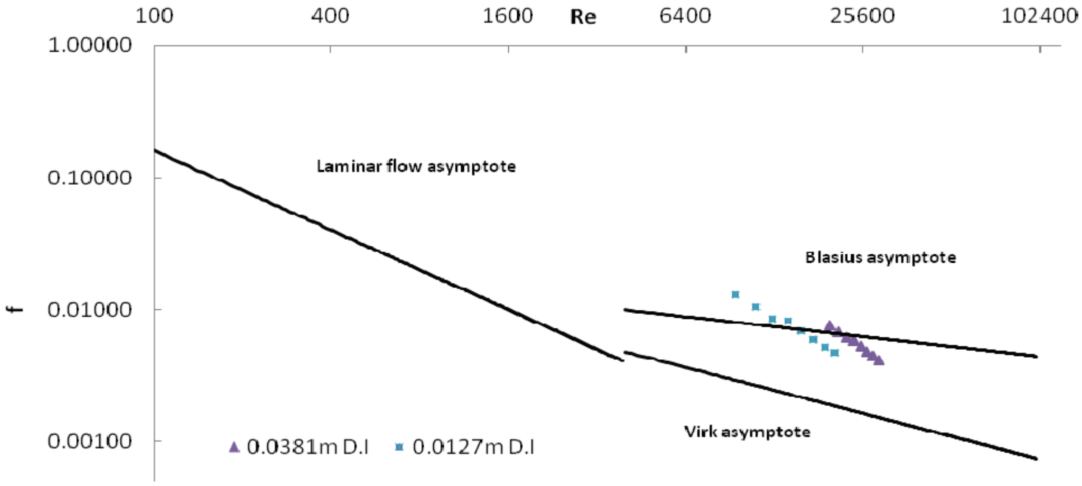


Figure 5. Data verification for hydrocarbon liquid at L/D = 39.37.

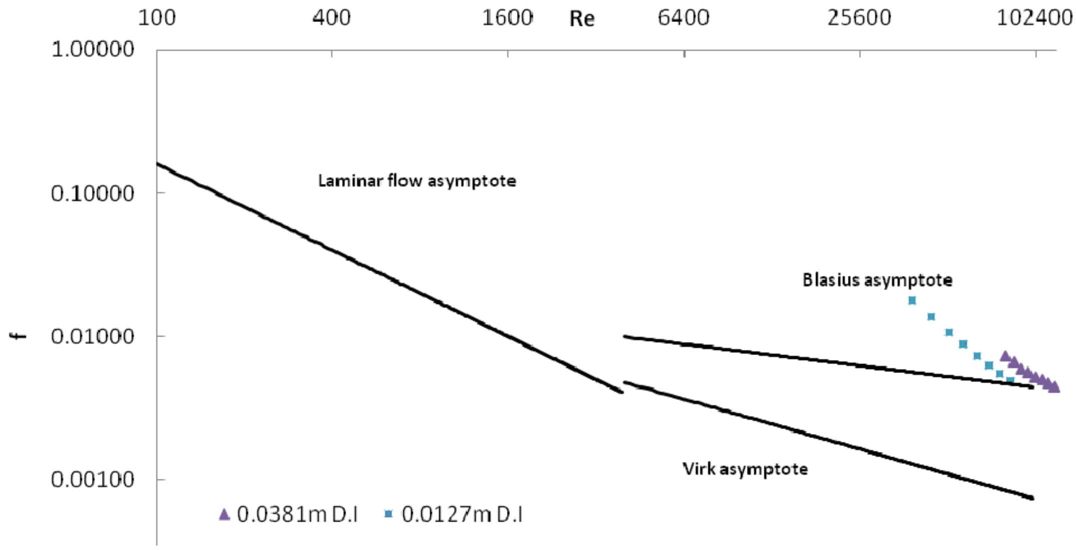


Figure 6. Data verification for water at L/D = 39.37.

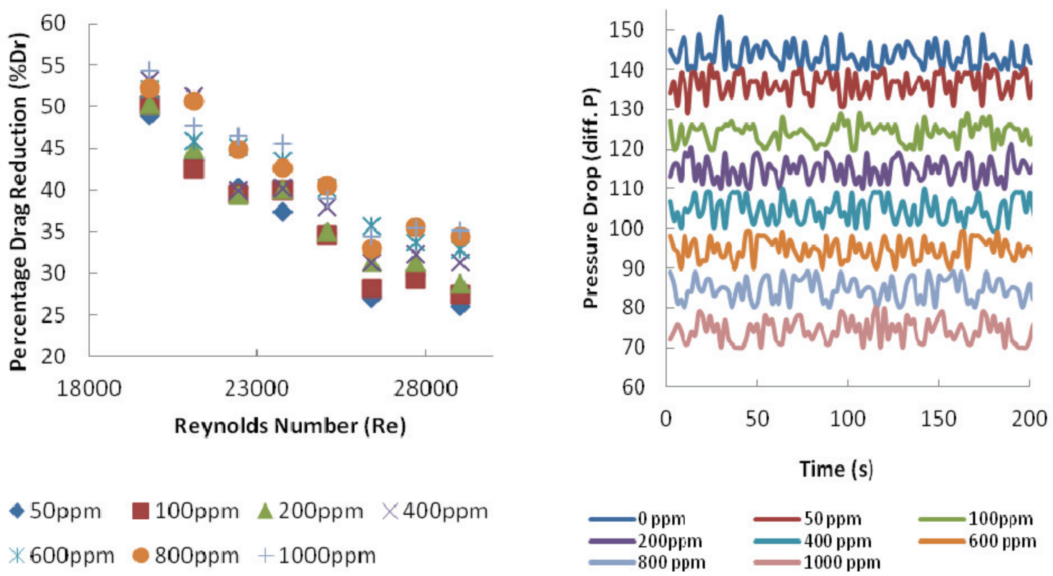


Figure 7. Effect of concentration (ppm) on %Dr in hydrocarbon liquid by increasing Reynolds number with different addition of grafted polymer trough 0.0381 m ID and 2.0 m pipe length and pressure drop data for Re = 29017.

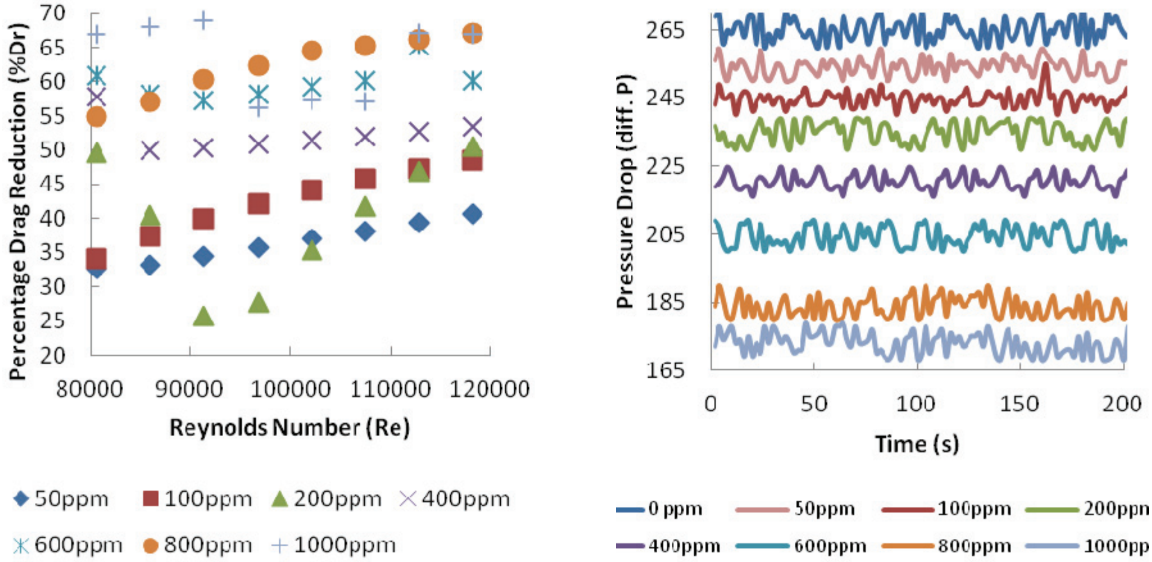


Figure 8. Effect of concentration (ppm) on %Dr in water by increasing Reynolds number with different addition of natural polymer trough 0.0381 m ID and 2.0 m pipe length and pressure drop data for Re = 118235.4.

it can be noticed that the *%Dr* decreases by increasing the Re (within the investigated range 18×10^3 until 3×10^4), which is due to the complicated relationship between the additive properties (especially the viscoelasticity and degradation resistance), degree of turbulence and concentration. Increasing Re means increasing the degree of turbulence inside the pipeline and that will increase the shear stress force applied by the main flow to the polymeric molecules interfering within the turbulent structures (eddies), which will lead to the reduction of the drag reducing agent performance. Figure 8 shows the opposite behaviour when applying the water soluble mucilage, where the *%Dr* increases by increasing Re. To explain this, it is very important to know that the Re range investigated for the water solution is less that that investigated with the diesel solution due to the limited capacity of the experimental rig for the two different liquids. For the case of water solution, the *%Dr* increases by increasing Re. Increasing Re means increasing the degree of turbulence inside the pipe, which will bring a more suitable environment for the additive to interfere within the turbulent structures and its drag reduction effectiveness will increase. For the case or water solution, the Re range was lower than the diesel and that means the degree of turbulence is lower also and the balance between the additive physical properties (viscosity and viscoelsticity) and the degree of turbulence (shear force), still holds and the drag reduction performance is increasing. For the case of the diesel solution (Figure 4), the Re range was higher and the shear forces were higher also and that led to the appearance of the declining part of the *%Dr*-Re curve only. Maximum *%Dr* of 55% was achieved by the addition of 1000 ppm for diesel flowing through 2.0 m pipe length and 0.0381 m ID at Re range of 18×10^3 to 3×10^4. While, 70% drag reduction was achieved when adding 1000 ppm of the water soluble additive to the water flowing through 2.0 meter pipe length and 0.0381 m ID at Reynolds number range of 65×10^3-75×10^4. Most of the experimental results agree well with the previous work published by many authors such as Mowla and Naderi [6], Shetty and Solomon [7], Dubief *et al.* [10], Abdul Bari *et al.* [13] and Choi *et al.* [17].

One the interesting findings in the present work is that the %Dr increases by increasing the addition concentration of the additive as shown in Figures 7 and 8. Increasing the addition concentration means increasing the number of the additive molecules involved in the drag reduction process. Increasing the concentration will increase the interference of the additive molecules with the turbulent structures (eddies) inside the main flow system and that will lead to two results. The first result is increasing the apparent viscosity of the transported liquid due to the noticeable difference in viscosity between the transported liquid and the additive, which leads to the reduction of the number of eddies formed in the turbulence core. The second result is reaching the optimum additive-liquid balanced concentration without changing the viscosity of the transported liquid and at the same time the additive will perform as DRA within the concentration range and that is what was achieved in the present work. Figure 5 shows the apparent viscosity measurements for all the solutions investigated. It is clear that the viscosity value changed by 0.4% only when the concentration of the additive was increased up to 1000 ppm.

One of the key factors influencing and controlling the DRA value is its effect on the transported liquid apparent physical properties (especially the viscosity). Commercially, it is important to highlight that the addition concentration of the DRA must be within the range that shows slight effects on the physical properties of the transported liquid. In the present work, the addition concentration ranged between 50 to 1000 ppm. To make sure that the investigated concentration range was within the acceptable limits (viscosity and density change limits), the apparent dynamic viscosity and density of the transported liquids was measured and the results are presented graphically in Figure 9. The figure shows that the maximum change in the gas-oil dynamic viscosity was 11.0% by the addition of 1000 ppm compared with the additive-free liquid viscosity (Figure 9a). The density change was not more than 2% (Figure 9b). For the water solutions, the maximum change in the viscosity and density was 7 and 2%, respectively.

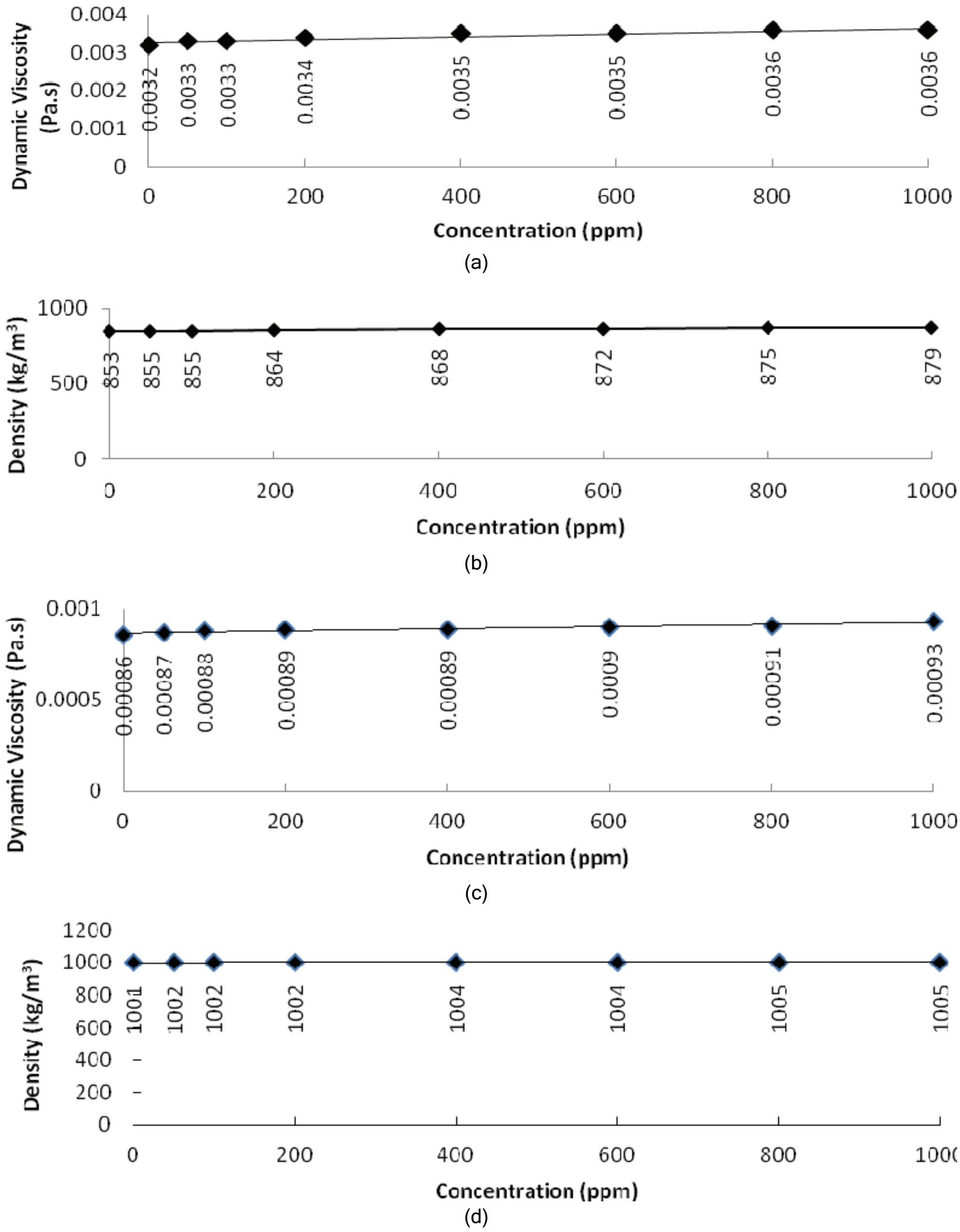


Figure 9. Changes in: a) kinematic viscosity of diesel by adding of DRA concentration; b) density of diesel by addition of DRA concentration; c) kinematic viscosity of water by addition of DRA concentration; (d) density of water by addition of DRA concentration.

The effects of viscosity and density are shown in Figures 9a-9d.

Figures 7 and 8 also show the pressure drop readings before and after addition of polymer to the hydrocarbon liquid. Frequency and pressure drop of the data become lower in same period of time when adding minute's quantity of polymer. Lowering pressure drop means improving the flow. The fluctuation shows the frequency been reduced by increasing of concentration. Turbulent flows are usually associated with unstable flow with fluctuating pressure drops. As known, pressure drop can similarly represent eddies formation by losing the main flow energy. By addition of polymer, it is proven that the flow becomes smoother, which can maintain the energy inside pipe, and the turbulence can be smoother and "laminarize".

On the other hand, this can prove that this natural polymer can maintain the effectiveness as DRA in certain period of time and pump cycles. This is one important factor to be considered because it shows degradation rate of polymer as DRA when injecting into pipeline system. The possibility of polymer degradation due to pump circulation gets higher with time. The pump shear stress due to the rotation is high and can cause the polymer to break into shorter polymers. This is the major factor why pressure drop slightly increases with time. These factors prove the findings of Choi *et al.* [18] that polymers will be degrade within time and pump cycles.

One of the main factors influencing the drag reduction performance is the effect of transported pipe geometry. In the present work, two pipe diameters (0.0381 and 0.0127 m ID) were investigated within all other experimental variables. Figures 10 and 11 show selected samples of the experimental data for the two diameters investigated. Although the Reynolds num-

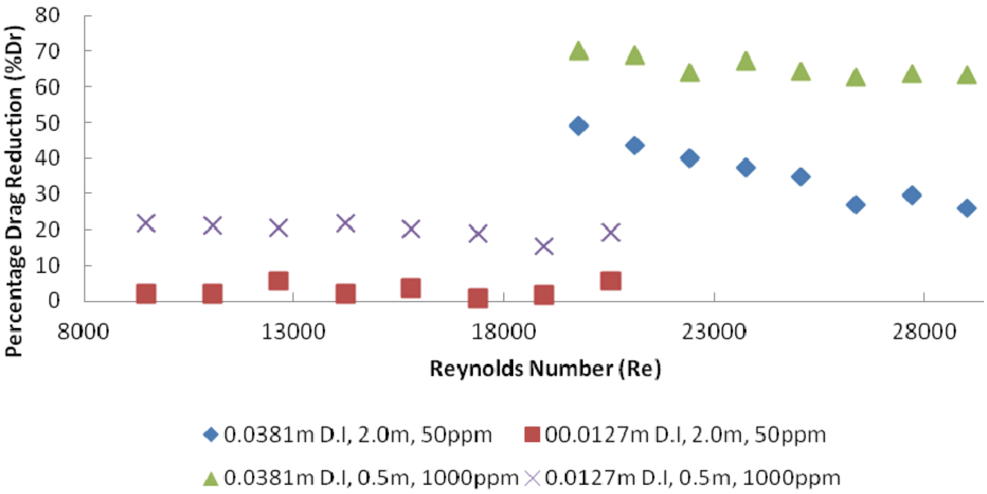


Figure 10. Effect of different conditions on %Dr with transporting hydrocarbon liquid.

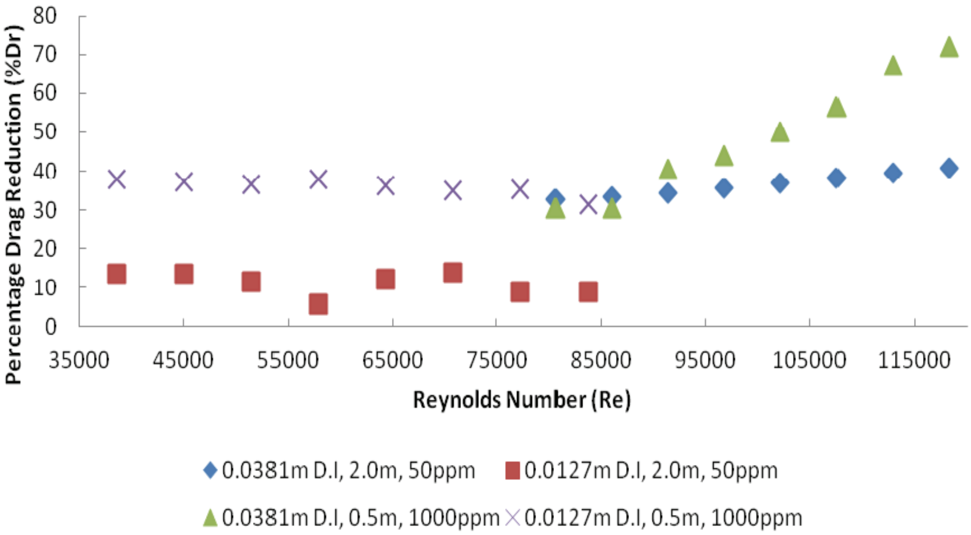


Figure 11. Effect of with different condition on %Dr with transporting water.

ber range is not the same for the two pipes investigated due to the flow capacity of the rig, there is a wide difference between the *%Dr* for the two pipes. It is very clear that the *%Dr* will be higher for the larger pipe (0.0381 m ID) and that maybe due to the change in the space available for the eddies to stretch which will lead to the fact that larger and slower eddies will appear in the turbulent flow. It is believed that such eddies are much easier to be suppressed by the additive due to its low shear intensity. The present work results disagree with the results presented previously by Bari *et al.* [23] who suggested that the drag reduction performance would be higher for lower pipe diameter.

Figures 12-15 show the effect of testing section length on the drag reduction performance of the new additive. Figure 14 shows that the %Dr increases by increasing the testing section length while figures 12, 13 and 15 shows that the %Dr will be higher when the testing section length decreases. Figure 13 showing increment at the first stage before reducing %Dr at higher Re. To explain that, it is very important to relate the effect of the pipe diameter to the degree of turbulence inside the pipe. Figures 14 and 15 show the effect of the testing section length for the 0.0381 m ID and 0.0127 m ID pipes, respectively. Decreasing the pipe diameter will increase the degree of turbulence inside the pipe and that will increase the shear force each molecule of the eddies is exposed to. It is believed that for smaller pipe diameter, the additive will lose its effect when the transportation distance increases due to the continues effect of the high shear forces of the turbulence while the opposite effect will appear for the large pipe diameter because the eddies shear force is lower due to the large space inside the pipe.

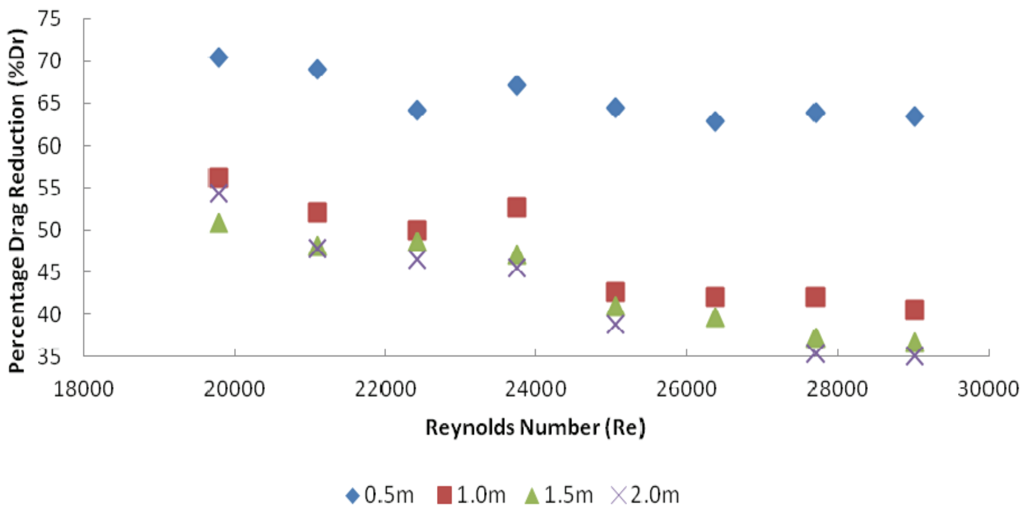


Figure 12. Effect of pipe length on %Dr for 0.0381 m ID carrying hydrocarbon liquid with 1000 ppm DRA addition.

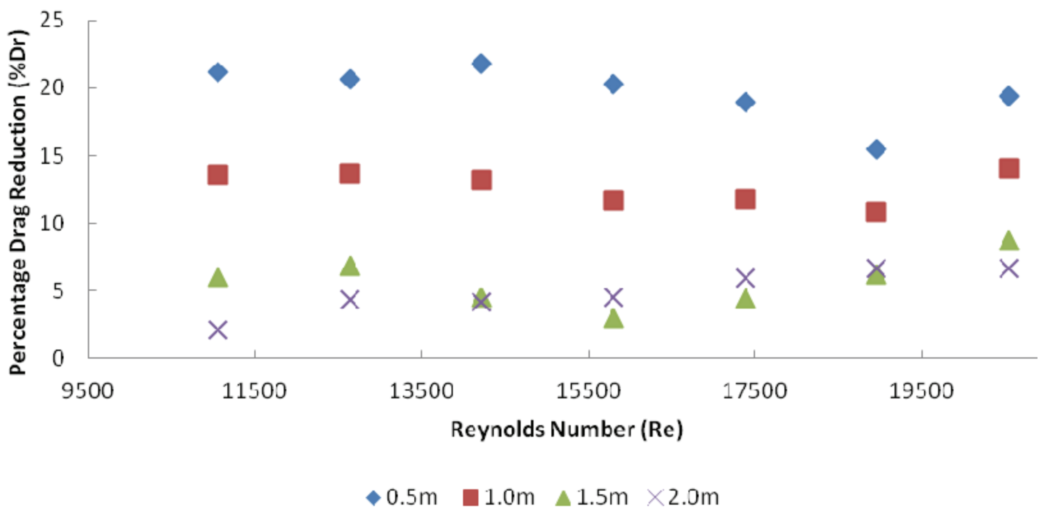


Figure 13. Effect of pipe length on %Dr for 0.0127 m ID carrying hydrocarbon liquid with 1000 ppm DRA addition.

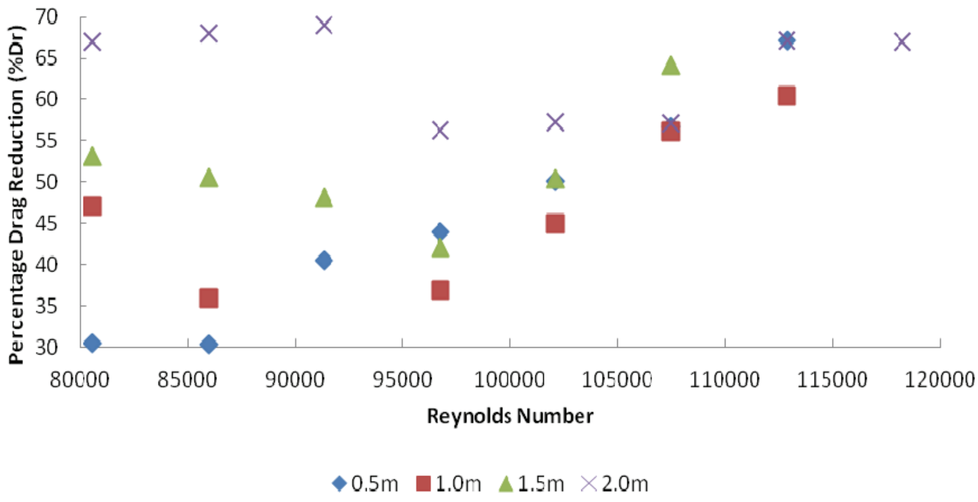


Figure 14. Effect of pipe length on %Dr for 0.0381 m ID carrying water with 1000 ppm DRA addition.

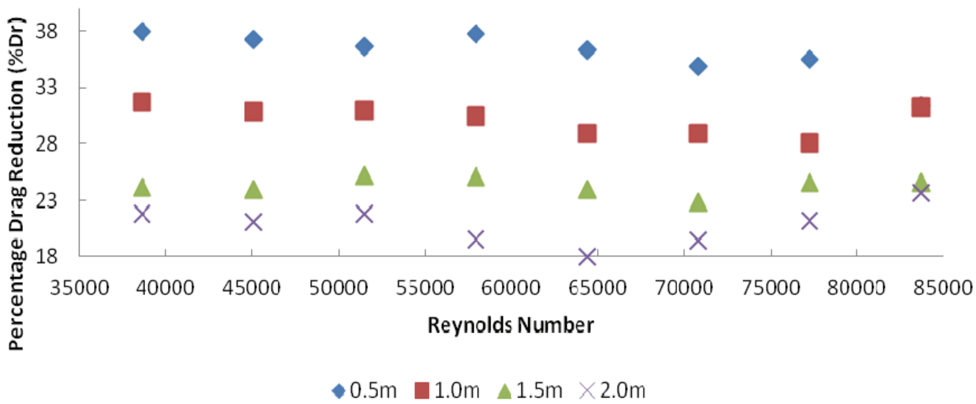


Figure 15. Effect of pipe length on %Dr for 0.0127 m ID carrying water with 1000 ppm DRA addition.

CONCLUSION

It is concluded that okra mucilage-acrylonitrile grafted polymer is applicable as diesel soluble DRA since it is biodegradable, highly effective in water and hydrocarbon liquid. Okra mucilage-acrylonitrile grafted polymer drag reduction performance was found to increase by increasing fluid velocity, pipe length and internal pipe diameter. The drag reduction effectiveness of the new additive was found to decrease with time due to the mechanical degradation of the grafted polymer molecules when exposed to high shear forces exerted by the pump and the turbulent eddies inside the pipe.

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NAUČNI RAD

GRAFTOVANI PRIRODNI POLIMER KAO NOVI AGENS ZA SMANJENJE OTPORA: EKSPERIMENTALNI PRISTUP

U ovom radu preporučen je novi prirodni agens za smanjenje otpora koji poboljšava tok fluida u cevima kroz koje turbulentno struje vodeni rastvori ili ugljovodonične tečnosti. Svojstva okre (Abelmoschus esculentus), kao agensa za smanjenje otpora strujenju, ispitivana su u vodenim i ugljovodoničnim (gas-ulje) medijima nakon graftovanja. Test smanjenja otpora izvođen je u sistemu za cirkulaciju tečnosti sa zatvorenom petljom, koji se sastojao od dve cevi unutrašnjih prečnika 0,0127 i 0,0381 m, sa četiri merna mesta na obe cevi (0,5-2,0 m, rezervoara, pumpe i transmitera pritiska. Rejnoldsov broj (Re), koncentracija aditiva i tip fluida koji struji (voda i gas-ulje) su bile glavne promenljive čiji je uticaj ispitivan na smanjenje otpora strujanju. Eksperimentalni rezultati su pokazali da novi agens može smanjiti otpor strujanju čak do 60%. Sposobnost agensa da smanji otpor strujanju raste sa njegovom koncentracijom. Smanjenje otpora raste sa povećanjem Re-broja kod vodorastvornih aditiva, a opada sa porastom Re-broja kod aditiva rastvornih u ulju. Smanjenje otpora bilo je izraženije kod cevi unutrašnjeg prečnika 0,0381 m. Konačno, graftovane i prirodne biljne sluzi su pokazale veliku otpornost na smicajne sile pri kontinualnoj cirkulaciji u toku 200 s u zatvorenom sistemu.

Ključne reči: prirodni agens za smanjenje otpora strujanju; procenat smanjenja otpora; cevovod; turbulentni tok.