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Synergistic Effect of Natural and Synthetic Polymers as Drag Reducing Agents in Single Phase Water Flow

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Abstract

One of the most power consuming applications in the process and petroleum industry is the turbulent means of transporting fluid over long distances through pipelines. High pumping energy is required to overcome the occurring frictional pressure drop. An economical approach has been reported where addition of small amount in parts per million (ppm) of high molecular weight polymeric solution can lead to large decrease in pressure drop. Synthetic polymers are more effective in reducing pressure drop (drag reduction, DR), but cannot be degraded easily by biological means. There is the need to combine them with natural polymers to improve DR and reduce their environmental effect. In this study, an experimental flow facility which consists of liquid storage tanks, pumps and unplasticised polyvinylchloride (uPVC) horizontal pipe system with 20 mm ID has been constructed. DR in single phase water flow was investigated using partially hydrolyzed polyacrylamide (HPAM; magnafloc 1011), polyethylene oxide (PEO) and Aloe Vera mucilage (AVM) separately, as well as mixtures of HPAM-AVM and PEO-AVM at Reynolds number from 12,916 to 48,871. The master solutions of 2000 ppm and 20000 ppm for HPAM, PEO and AVM respectively and their respective mixtures were used at different concentrations. The pressure drop was measured using mercury U-tube manometer. Drag reduction of 75.2, 78, 82.5 and 83% was obtained by the polymer mixtures, which was higher than the DR of individual polymer alone at all mixing proportions and Reynolds numbers at the same concentration. The observed synergism in DR may be attributed to the increase in the dimension of the macromolecular coil of the polymer mixture solutions due to the presence and rigidity of other polymer molecules.

Keywords: Drag reduction; synergism; polymers; polymer-polymer mixture.

1. INTRODUCTION

Drag reduction (DR) is defined as the decrease in pressure drop in turbulent flow by addition of small amount of polymers (Bewarsdorff and Gyr, 1995) which had been reported way back in 1940 (Toms, 1940). It was reported that adding small amounts of long-chain polymer in turbulent flow caused large decrease in frictional pressure drop in pipes. Meanwhile turbulent drag had been reported much earlier (Forrest and Grierson, 1948).

These have found wide range of applications in the process industries such as crude oil pipeline trans-portation over long distances, irrigation, floodwater disposal and sewage. Other applications include marine and biomedical system, drilling of oil from reservoir, firefighting, extraction, filtration, heat and mass transfer application (Edomwonyi-Otu and Angeli, 2014; Marmy *et al.*, 2012). It was recently suggested that they could be used for transportation of drinking water because of their harmless properties (Edomwonyi-Otu and Adelakun, 2018).

Synthetic drag reducing polymers such as polyacrylamide (PAM), hydrolyzed polyacrylamide (HPAM) and polyethylene oxide (PEO) are found to be very effective in drag reduction. However, they have high resistance to biological degradation which makes them less environmentally friendly (Nour *et al.*, 2012). Natural drag reducing polymers, which are mainly polysaccharides are less effective on drag reduction, environmentally friendly and are degraded quickly by biological means (Deshmukh and Singh, 1986). Some efforts have been made to improve the drag reduction effectiveness as well as reducing the environmental impact of flexible polymers such as PEO, HPAM and PAM among others. This can be done by combining with a rigid polymers such as xanthan gum (XG), Carboxymethyl cellulose (CMC), Aloe Vera mucilage (AVM) and other plant and animal extract in order to have a synergistic effect on drag reduction and environmental friendliness (Singh, 1995).

Some studies had been carried out to improve the drag reduction effectiveness and shear stability by using polymer-polymer, polymer-fiber and polymer-surfactants mixtures in order to have a synergistic effect on drag reduction (Edomwonyi-Otu *et al.*, 2016; Gustavo and Soares, 2016; Lee *et al.*, 1974; Malhotra *et al.*, 1988; Reddy and Singh, 1985). It also aimed to improve the effect of synthetic polymer degradation (Gustavo and Soares, 2016).

Dingilian and Ruckenstein (1974) were the first to use mixtures of PEO, PAM and CMC at different concentrations. They observed a positive synergistic effect on PEO-CMC and PAM-CMC combination and negative effect on PAM-PEO combinations. Drag reduction of the binary polymer system was higher (positive deviation) than the drag reduction caused by each of the polymers when present alone in a solution at the same concentration as in the mixture. It was proposed that, to achieve a synergy, at least one or both of the polymers must have rigid structure.

The investigation of interaction of polyethylenimine with anionic polymers in the solution mixtures of polyethylenimine with PAM, PEO and polyacrylic acid using fluorescence polarization measurements resulted in the loss of their drag reducing properties (Parker and Joyce, 1974). In the study of DR in mixtures of polymers and fibers in pipe diameters of 2.5, 5.0 and 7.0 cm (Dschagarowa and Bochossian, 1978; Lee *et al.,* 1974) and in the polymer-polymer and polymer-fiber mixtures in a flow system of pipe diameter of 25.4 mm and 2 m length of the test section (Reddy and Singh, 1985), synergistic effect had been reported. The mixture of a rigid and flexible polymer was found to give maximum synergy compared to mixture of two flexible polymers (Dingilian and Ruckenstein, 1974) due to increase in polymer coil dimension as a result of the co-presence of molecules of both polymers in the solution and the rigidity of the polymers.

In the investigation of DR by polymer-polymer mixtures in steel capilary tube of diameter 0.1575 cm, concentrations of 20 and 100 ppm and at Reynolds number (Re) of 14000 Malhotra *et al.,* (1988), synergism was not achieved at lower concentration and Re. It was shown that the interaction between the polymer molecules at higher concentration and flow rate increased the extension of the polymer molecules which resulted in the expected synergy in the DR.

In a recent, study of mixtures of PAM-XG and PEO-XG in a straight tubes (Gustavo and Soares, 2016) a clear synergy in DR was observed for the two different polymer combinations suggesting that the co-presence of different polymer molecules may have caused the formation of aggregates which led to the increase in DR efficacy (Shetty and Solomon, 2009; Wyatt *et al*., 2011; Mohsenipour and Pal, 2013; Pereira *et al.,* 2012; Steele *et al*., 2014; and Andrade *et al.*, 2016).

Despite the works done in understanding the interaction between natural and synthetic polymer solutions, the literature is still scanty compared to synthetic polymers. There still remained the need to develop robust models for the accurate prediction of drag reduction in synergistic systems which require a lot more data than is currently available. Hence, this work aims to provide more data on the study of the synergistic effect of aloe vera mucilage (AVM), polyethylene oxide (PEO) and hydrolyzed polyacrylamide (HPAM) in a horizontal water flow system.

2. EXPERIMENTAL SET-UP

Description of the flow facility.

The schematic diagram of the experimental set-up is as shown in Figure 1. The flow facility was divided into three sections which are: the handling section, pumping or regulating section and test section. The handling section consists of water and separator tank, for water draining, with capacity of 200 and 220 liters where the water was stored. In the regulating or pumping section, 20 mm ID uPVC pipes were each connected to water and separator tanks. The centrifugal pumps (model Jet 102M/N.31227) were used to circulate the test fluids into the test section. The globe valves were used to regulate the flow rates which were measured with variable area flow meters (LZM-20J; ±5% accuracy). The valves were located between the pumping section and the test section while the globe valves also regulate the recycle and bypass flows. The calibrated water flow meter had maximum flow rate of 24 GPM or 100 LPM while the injection port for the polymer master solution was located by the side of the water pipeline before Y-junction. The New Era-programmable peristaltic pump (model NE-9000; ±2% accuracy) was used to inject the polymer master solution into the water phase. The test section was made up of straight acrylic pipe of 20 mm ID and 140 times the diameter of the pipe (140D) long from the Y-junction to the second pressure port. The pressure taps were created by making small holes at the bottom of the acrylic pipe walls at the distance of 140D which provided fully developed flow in the test section. U-tube manometer (Pyrex) and Fann viscometers (model 35A; ±1% accuracy) were used for the measurement of pressure drop and viscosity of the polymer solutions respectively.

Polymer preparation

The polymers used were polyethylene oxide (PEO) manufacture by Sigma-Aldrich with average molecular weight of 8×10^6 g/mol, partially hydrolysed polyacrylamide, HPAM (Magnafloc 1011) manufacture by BASF with molecular weight of chemicals 10×10^6 g/mol, and aloe vera mucilage (AVM) extracted from aloe vera plant. All the polymers, were first prepared individually before the polymer mixture solutions, were water soluble and were used without further purification.

A master solution of 2000 ppm of each of the synthetic polymer was prepared as follows: 10 g of each of the polymer powder was measured (Kerro, BLC 3002) and gently spread over 5 liters of water surface and stirred for 3 hours with a mechanical stirrer (Gilverson, L28) at low speed (to avoid degradation of the polymer) for the mixture to be completely homogenized. The stirred solution was left for 12 hours, mostly overnight, to ensure complete dissolution of the polymer particles and removal of trapped gas bubbles to form the master solution (Abubakar *et al.*, 2014a; Edomwonyi-Otu *et al.*, 2015).

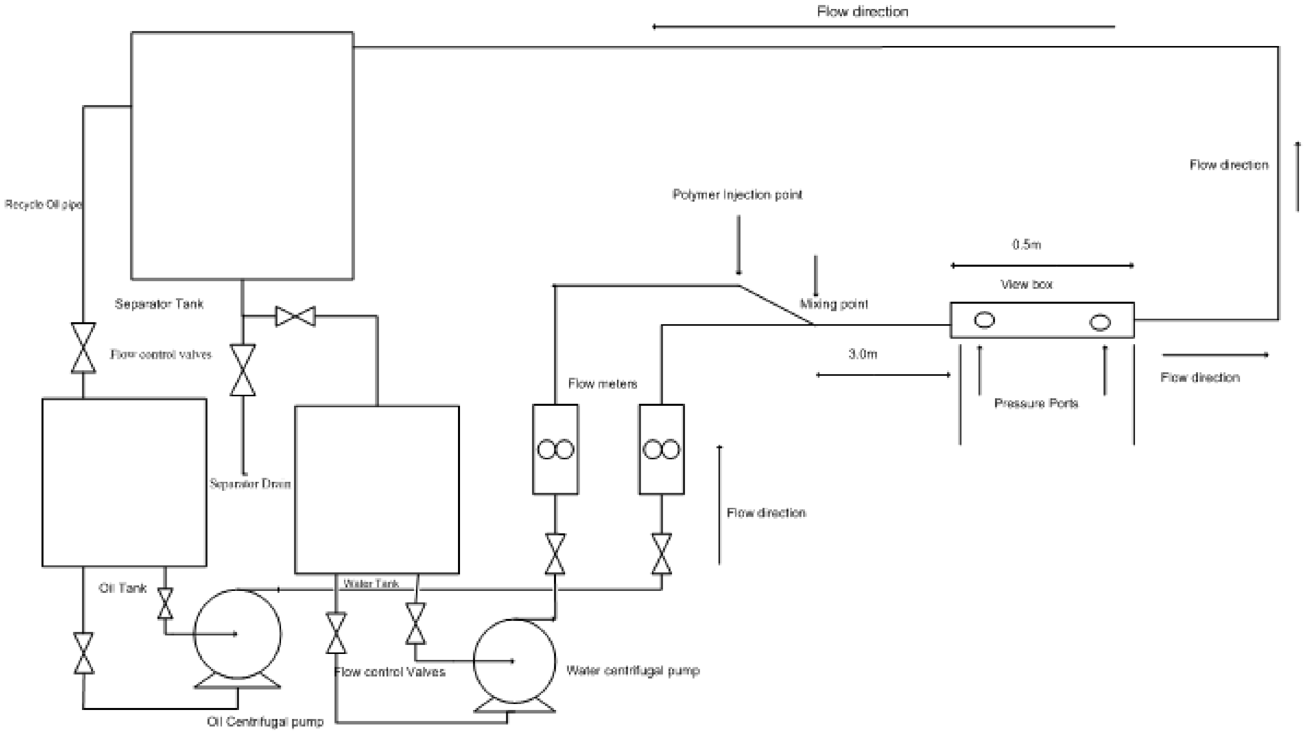

Figure 1: Schematics of experimental flow facility.

Aloe vera leaves were harvested from a garden, washed, cut vertically on both sides and soaked in water for 10 minutes to remove the Aloin within them. The leaves were then peeled and the aloe vera mucilage (AVM) was extracted by scraping and sieving the gel from the aloe leaves. Aloe vera leaf contains about 98% water while the remaining 2 % is the AVM (Bozzi *et al.,* 2007; Davis, 1997). 20,000 ppm master solution of AVM was prepared. AVM suffers biological degradation in 24 hours, its solution was aged for 4 hours for thorough mixing (Abdulbari et al., 2011). The master solution was injected into the water-phase at specific flow rate of the injection pump in order to achieve the required concentration in the water flow line. The master solutions of each of the polymer mixture were prepared by mixing the solutions of each of the polymers prepared separately in required quantities.

After the preliminary experiments with each of the polymer solutions in single phase water flow, the total concentration (TC) for any mixture was chosen based on at least one of the polymers in the mixture giving maximum DR at that concentration (Reddy and Singh, 1985).

In case of this study, 30 ppm and 400 ppm were selected as the total concentration for the mixture of HPAM-AVM and PEO-AVM. The mixture master solutions of 2000 ppm and 20,000 ppm were prepared to achieve the required proportions. The mixing of the polymers can also be guided using Equation 1 (Gustavo and Soares, 2016)..

$$C_1 + C_2 = 1$$

where, C_1 and C_2 are concentrations of each of the polymers..

Experimental procedure

After the preparation of the polymer mixture, a sample was collected for viscosity measurement as a criterion for biodegradability test using a viscometer (Fann 35; rotor-bob-torsion spring combination, R1-B1-F1 and shear rate of 511 sec^{-1} at ambient conditions). The criterion used for the biodegradation test is the measurement of viscosity of the polymer master solution as a function of time as described by Singh *et al.,* (2000).

The master solution of 2000 ppm (HPAM and PEO) and 20,000 ppm (AVM) of the polymers as well as their mixtures were tested for their biodegradability at mixing proportions for period of 10 days. After the experiment, one more sample of solution was collected for the same purpose. The average of these two measurements was taken. The flow meters and injection pump was tested before running the experiments to ensure accurate delivery of the required amounts of water and polymer concentration into the test section.

The experiment was carried out in horizontal pipe of test section of 20 mm ID at ambient conditions (25 °C, 1 atm). The pressure drop was measured using the U-tube manometer. Each experimental run was repeated three times and the average of the pressure drop measured before and after the addition of the drag reducing agents of the master solutions. It was observed that, within 24 hours of the preparation of the AVM, it started degrading and at the end of 240 hours the solutions had shown considerable loss of viscosity, which shows similar behavior with xanthan gum. Altering the structure of the AVM molecules with HPAM and PEO made it less susceptible to bacteria attack and improved DR. It had been reported (Deshmukh and Singh, 1987; 1991 and Singh *et al.,* 2000) xanthan gum degraded within 24 hours, but grafting it with polyacrylamide showed no loss in viscosity up to 10 days making it to be less susceptible to biodegradation.

HPAM also showed no loss in viscosity up to ten days owing to its high resistance to bacteria attack which made it less susceptible to biodegradation. PEO showed no loss in viscosity for 72 hours but degradation

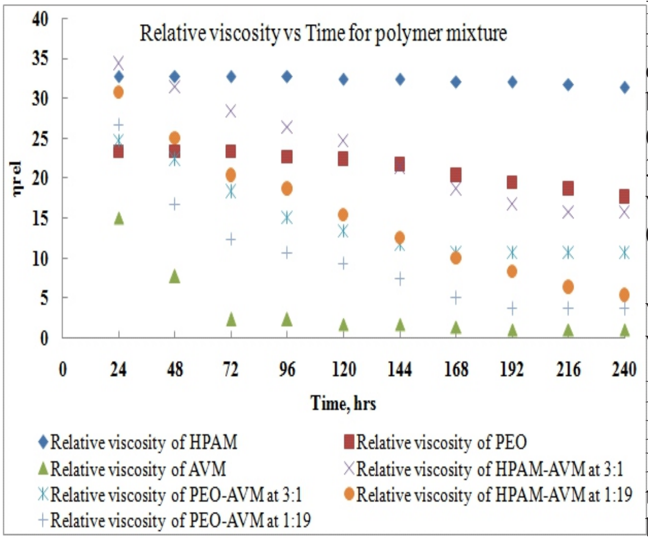

Figure 2: η_{rel} vs time for HPAM, PEO and AVM as well as their mixtures at 3:1 and 1:19 proportion for biodegradation test.

(DRAs). HPAM, PEO, AVM, HPAM-AVM and PEO-AVM were tested at different concentration and Reynolds numbers. The concentrations of HPAM and PEO ranged from 2.5-100 ppm while the concentration time progressed which may be attributed to its flexibility (Deshmukh and Singh, 1987; 1991 and Singh *et al.,* 2000).

It also observed that the viscosity of the polymer mixture of HPAM-AVM and PEO-AVM at mixing ratio of 3:1 gradually decreased with time. The altering of the AVM structure by the synthetic polymers (HPAM and PEO) made it less susceptible to bacteria attack which minimized the biodegradation as well as improved drag reduction effectiveness. However, in mixing ratio of 1:19, greater loss in viscosity was observed as time progressed compared to that of ratio 3:1 which may be due to high proportion of the AVM in the polymer mixture solution (Deshmukh and Singh, 1987 and 1991).

HPAM, PEO and AVM Mixtures

Figures 3 - 5 show the plots of drag reduction against concentration at different Reynolds numbers for HPAM (Figure 3), PEO (Figure 4) and AVM (Figure 5) in single-phase water flow (SPF). It was observed that DR increased with increase in the polymer concentration at different Re due to the increase in the number of HPAM, PEO and AVM molecules which increased the viscosity of the transported liquid. The increase in polymer concentration of aloe vera mucilage (AVM) ranged from 5-500 ppm at flow rates of 10 l/min, 20 l/min, 30 l/min and 40 l/min (Re from 12,916-48,871). The optimal polymer concentration for this flow system was found to be 30 ppm for HPAM and PEO and 400 ppm for AVM from our single phase preliminary experiments. The optimal concentration was selected to be the total concentration (TC) of the polymer mixture (Reddy and Singh, 1985). The Re of 12,916, 25,339, 37,773 and 48,871 were investigated. The pressure drop was recorded and used for calculation of drag reduction (Equation 2);

$$DR = (\Delta P_{wo} - \Delta P_{w})/\Delta P_{wo} \times 100\% \quad \ldots\ldots\ldots(2)$$

where ΔP_{wo} and ΔP_{w} are pressure drop of the fluid without and with DRAs.

RESULTS AND DISCUSSION

Biodegradation test

Figure 2 shows the plots of relative viscosity against time for polymers and their mixtures indicating biodegradation increased the interference of the additive molecule with the turbulent eddies in the pipeline flow.

Also as the Re increased the extent of the turbulence in the water pipeline flow increased, which created sufficient environment for the additive-turbulent eddies interaction which brought about drag reduction effectiveness (Abubakar *et al*., 2014b; Edomwonyi-Otu *et al*., 2015; Virk, 1975). It was also observed that as the Re increased from 37,773 to 48,871, there was high level of instability in the flow. However, for the aloe vera mucilage (AVM), DR decreased at higher Re of 48,871 due to the effect of polymer entanglement (Bhambri and Fleck, 2016).

Among the three polymers studied, PEO was found to be more effective (with 72% DR at 30 ppm) compared to the remaining two probably due to the flexibility of the PEO (Abubakar *et al.*, 2014a; Bewardoff and Gyr, 1995; Eshrati *et al.*, 2015; Japper-Jaafar *et al.*, 2009). The drag reduction effectiveness of AVM, which has the least maximum DR (50% at 400

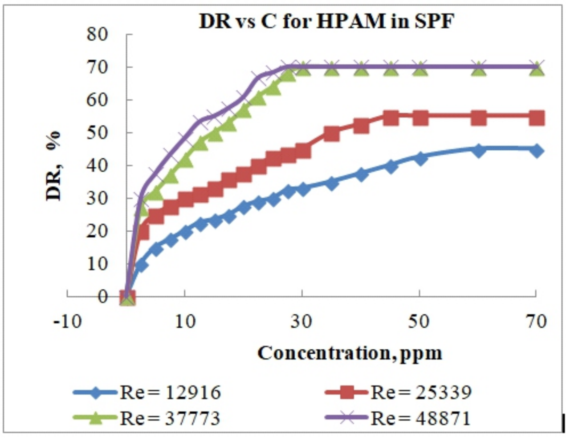

Figure 3: Plot of drag reduction against concentration for single-phase water flow at different Reynolds number for HPAM in 20 mm pipe diameter.

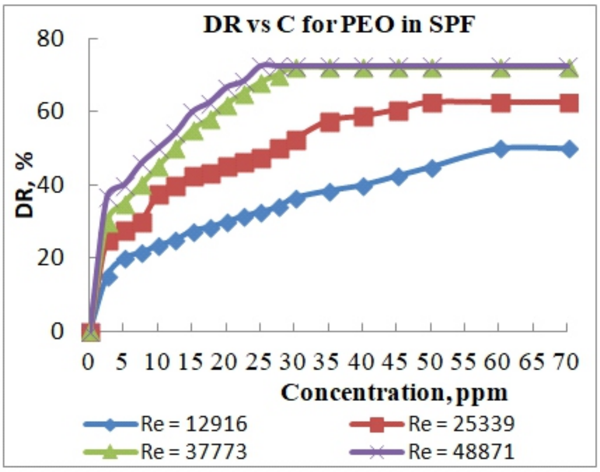

Figure 4: Plot of drag reduction against concentration for single-phase water flow at different Reynolds number for PEO in 20 mm pipe diameter.

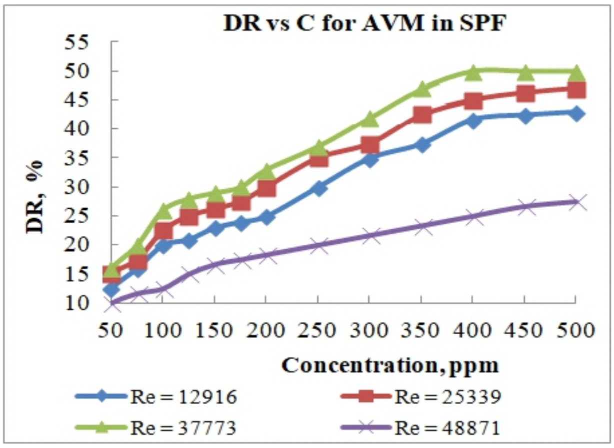

Figure 5: Plot of drag reduction against concentration for single-phase water flow at different Reynolds number for AVM in 20 mm pipe diameter.

ppm) among the three polymer used, was improved by blending it with flexible HPAM and PEO.

Single-phase flow with polymer mixtures

Figures 6-9 show the effect of polymer mixture concentration and Reynolds number on drag reduction in single-phase water flow (SPF) at different Reynolds number and concentration for mixture of HPAM-AVM and PEO-AVM. It was observed that DR increased rapidly with increase in the proportion of HPAM and PEO in the mixtures and Re, where maximum DR of 75.2, 78, 82.5 and 83% were obtained at the mixing proportion of 3:1 and 1:19 for a TC of 30 ppm and 400 ppm. It was observed that, at lower Reynolds number (12,916 and 25,339) the DR (31, 32, 43, 50%) obtained by polymer mixtures is less than the additive values (summation of DR of HPAM & AVM and PEO & AVM; 35, 37.5, 47.5, 53.73%) (negative deviation) of each of the polymer DR, but greater than the DR of each of the single polymer alone, implying that, there was no synergy at lower flow rate (lower Re) at all mixture concentration (Malhotra *et al.*, 1988).

Synergy in DR was observed at Reynolds number of 37,773 and 48,871 at all mixture proportion for both polymer mixtures (HPAM-PEO and PEO-AVM) at total concentration of 30 ppm showing a positive deviation. The %DR of 75.2 and 78% obtained at Re of 37,773, 48,871 and mixing proportion of 3:1 was higher than the additive values (68.8 and 72.8%) of each of the polymer DR (Dingilian and Ruckenstein, 1974; Reddy and Singh, 1985; Gustavo and Soares, 2016). DR synergy may be due to the interaction amongst the polymer molecules, which influence the extension of the molecules. It may also be due to increase in the polymer coil dimension and their rigidity. The increase in flow rate increased the stretching of polymer molecules which may have resulted in the observed DR synergy. It was also observed that above the Re of 37,773 there was high level of instability in the flow for this system while DR increased at all polymer mixture proportion due to increase in Re (Gustavo and Soares, 2016; Malhotra *et al.*, 1988; Reddy and Singh, 1985).

At the total mixture concentration of 400 ppm, it showed that, the %DR observed in the polymer mixture (52.5, 65, 83 and 83.5% at 1:19) was greater than that for each of the polymer solution (30 and 41%, 45 and 42.5%, 52 and 43.33% and 54.17 and 20.83% at 1:19) at all mixture proportions. As the HPAM and PEO proportions increased in the mixtures, the drag reduction increased due to the increase in the extension of their molecules in the mixture when compared with in their individual solution, bringing greater interactions of the polymer mixture molecules with the turbulent eddies.

However, a positive deviation from additivity was observed at Reynolds number of 49000 and at some

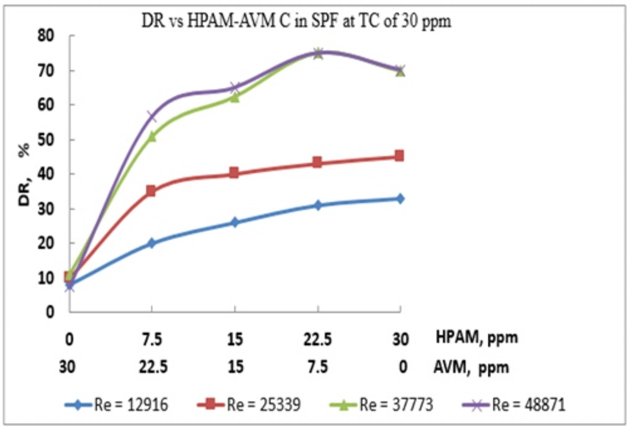

Figure 6: Drag Reduction against mixture concentration (HPAM-AVM) at different Reynolds numbers (Re) in 20 mm ID pipe and total concentration of 30 ppm.

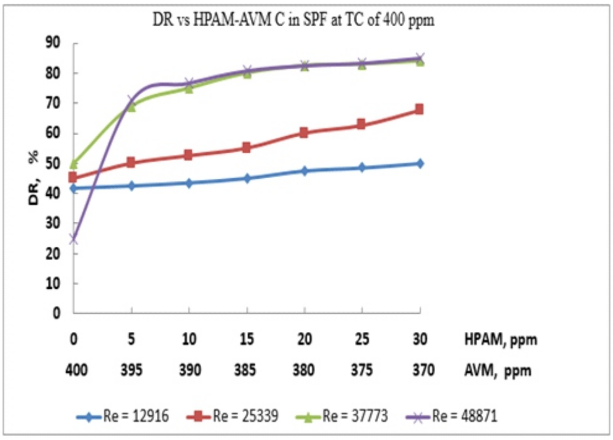

Figure 8: Drag Reduction against mixture concentration (HPAM-AVM) at different Reynolds numbers (Re) in 20 mm ID pipe and total concentration of 400 ppm.

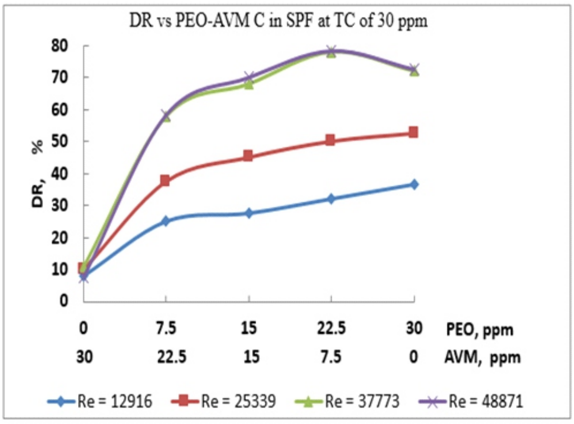

Figure 7: Drag Reduction against mixture concentration (PEO-AVM) at different Reynolds numbers (Re) in 20 mm ID pipe and total concentration of 30 ppm.

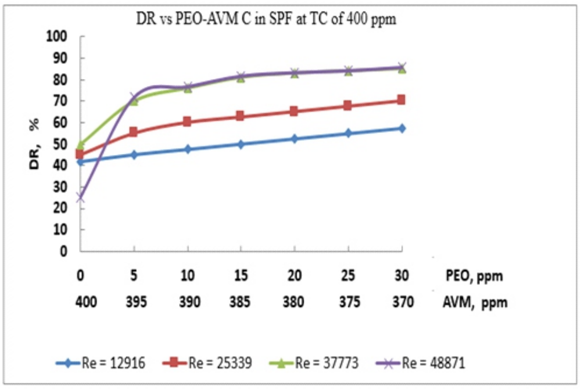

Figure 9: Drag Reduction against mixture concentration (PEO-AVM) at different Reynolds numbers (Re) in 20 mm ID pipe and total concentration of 400 ppm.

specific polymer mixture proportion which may be due to increase in the stretching of the polymer chains (Dschagarowa and .

Of the two polymer mixtures investigated, mixture involving PEO was found to be more effective (with 75.5% DR at mixture proportion of 22.5 ppm PEO-7.5 ppm AVM and 82.5% at mixture proportion of 20 ppm PEO-380 ppm AVM) than mixture using HPAM, which may be attributed to the flexibility of the mixture of PEO (Reddy and Singh, 1985; Singh, 1995).

4. CONCLUSIONS

The synergistic effect of natural and synthetic polymers as drag reducing agents in single-phase water flow in 20 mm horizontal pipe diameter has been studied. The AVM as polysaccharides can be made less susceptible to biodegradation by combining it with the synthetic polymers such as HPAM and PEO reducing environmental effect of the synthetic polymers. Grafting HPAM and PEO onto AVM enhanced effectiveness of drag reduction of the AVM. %DR of PEO and its mixture was higher than that of the HPAM and its mixture due to the degree of flexibility of the polymer molecules. DR and synergism in DR for the polymer mixtures depended very much on concentration and Reynolds number.

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