



Influence of structure and composition of poly(acrylamide-g-propylene oxide) copolymers on drag reduction of aqueous dispersions



Leidiane G. Reis^a, Ingrid P. Oliveira^a, Renata V. Pires^a, Elizabete F. Lucas^{a,b,*}

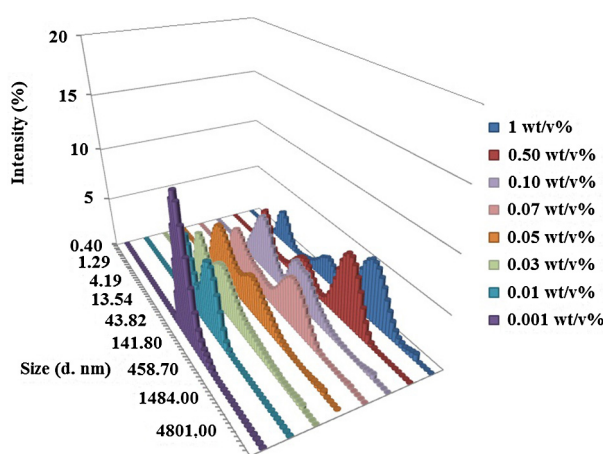
^a Federal University of Rio de Janeiro (UFRJ)—Institute of Macromolecules (IMA), Av. Horácio Macedo, 2030, Ilha do Fundão, 21941-598 Rio de Janeiro, Brazil

^b Federal University of Rio de Janeiro, COPPE, Program of Metallurgy and Materials Engineering, Av. Horácio Macedo, 2030, Block F, Cidade Universitária, 21941598 RJ, Brazil

HIGHLIGHTS

- Polyacrylamide-g-poly(propylene oxide) presents high efficiency as drag reducer.
- Influence of molar mass on drag reduction is overlapped by the intrinsic viscosity.
- Monomodal particle size distribution provides better efficiency in drag reduction.
- Amphiphilic graft copolymer has high resistance to shear degradation.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 16 February 2016
Received in revised form 28 April 2016
Accepted 3 May 2016
Available online 4 May 2016

Keywords:

Amphiphilic copolymer
Polymer dispersion
Surface activity
Drag reduction

ABSTRACT

Linear polymers with high molecular weight are used to increase the flow efficiency of viscous fluids. However, these molecules are susceptible to mechanical degradation, reducing their performance. On the other hand, amphiphilic molecules, such as surfactants, have produced good results as drag reducers and are more resistant to shear due to their capacity for self-regeneration. This paper evaluates the influence of amphiphilic copolymers of acrylamide grafted with poly(propylene oxide), with varied PPO contents and chain lengths, on the reduction of drag in aqueous systems under turbulent flow. The behavior of the amphiphilic copolymers in aqueous solution was evaluated in terms of molecular size and molecular size distribution and intrinsic viscosity. The drag reduction percentage of the polymeric dispersions was obtained by testing the pressure loss in a capillary tube viscometer. The drag reduction results indicated better efficiency of the grafted amphiphilic copolymers in comparison with the polyacrylamide homopolymer, being observed that such performance depends not only on molecular weight but also on intrinsic viscosity, molecular size and molecular size distribution. Moreover, the copolymer containing 5 mass% of PPO presented the best resistance to molecular breakage after 30 flow cycles at high shear rates.

* Corresponding author at: Federal University of Rio de Janeiro, COPPE, Program of Metallurgy and Materials Engineering, Av. Horácio Macedo, 2030, Block F, Cidade Universitária, 21941598 RJ, Brazil.

E-mail addresses: Leidiane.g@ima.ufrj.br (L.G. Reis), elucas@metalmat.ufrj.br, lucaselizabete3@gmail.com (E.F. Lucas).

This behavior can be related to the lower molecular weights and the possibility of interaction between the pendant chains of PPO, causing greater shear resistance.

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1. Introduction

Polymeric additives have been used in drilling fluids since the 1930s, when they were introduced to control infiltration. Since then, their use has increased because of their specificity, so that today they are the leading water-based fluid additives [1–4]. Polymeric additives can cause changes in the rheological, chemical and/or physical properties of the fluid even when used in small quantities, and can also be tailored to have specific properties by combining two or more polymers with different characteristics [3,5–8]. The choice of the polymer to be used for a determined application is associated with the specific characteristics, which in turn depend on the chemical structure and length of the chain, as well as the properties desired of the fluid to achieve the efficiency of drilling oil and gas wells [9]. The energy loss that occurs due to drag during high-speed flow of fluids increases operating costs, so the reduction of drag by the addition of polymer molecules has been widely studied. Various theories have been proposed to explain the drag reduction mechanism of polymeric additives [10–13], but this phenomenon is still not fully understood [9,13–17].

The main way of increasing flow efficiency involves adding small quantities of linear polymers with high molecular weight to the drilling fluid [2,14–18]. The difficulty of using these molecules is that they are susceptible to mechanical degradation, which reduces their efficiency. An alternative indicated in the literature is the use of amphiphilic molecules, mainly surfactant molecules [15,19–21]. In comparison with the linear polymers with high molecular weight used to reduce drag, surfactants are not susceptible to irreversible degradation, since they reassociate quickly after mechanical degradation [15,19–21]. Surfactant molecules have the capacity for self-regeneration during strong shear, making them highly efficient at reducing friction during the flow of fluids [22–39]. The drag reduction caused by adding a surfactant has therefore been considered adequate for applications involving reuse/recirculation of fluids [14,35]. The molecular self-organization characteristic is also seen in polymer-surfactant systems [25,27]. In recent years, amphiphilic polymer molecules have been widely investigated as drag reducers due to their ability to form self-organized structures in solution and their potential to be more resistant to molecular degradation by mechanical action than linear molecules with high molecular weight [29,40].

The drag reduction phenomenon has been demonstrated by use of polymers in solvents, such as poly(methyl methacrylate) (PMMA) in toluene and carboxymethyl cellulose (CMC) in water. Homopolymers based on polyacrylamide (PAAM) with poly(acrylic acid) (PAA) or with poly(ethylene oxide) (PEO) in water are increasingly being used [40–42]. The copolymer PAAM-PEO has become the most common due to its low production cost and easy handling, besides its efficacy in reducing pressure loss of fluids in pipes. The drag reduction caused by PAAM-PEO in relation to pure water was found to be 33% [43]. A study comparing the effect of a PAAM homopolymer with a copolymer of poly[acrylamide-co-*N*-acryloyl(L) vanilla] (90/10) on drag reduction found that the copolymer had virtually constant efficiency over time, explained by the conformation of the isolated macromolecule and its self-association in solution. The slight decrease in the efficiency of this copolymer is caused by the disaggregation of the aggregates. Besides this, the disaggregation rates are different due to the

competition between the polymer–polymer and polymer–solvent interactions. This explains the difficulty of obtaining reproducible results when measurements are made in concentrated solutions [44].

Studies conducted over the past 10 years into the efficiency of polymers as drag reducer have often investigated copolymers based on polyacrylamide (PAAM), poly(ethylene oxide) (PEO), poly(propylene oxide) (PPO) and tridecyl acrylate (C13), obtaining copolymers of the PAAM-g-PEO, PAAM-g-PPO, PAAM-g-PEO-g-C13 and PAAM-g-PPO-g-C13 types [29]. According to the authors, the results lead to the conclusion that the architecture of the additives analyzed, in the working conditions studied, is a parameter that governs the drag reduction of aqueous fluids. Better drag reduction performance is achieved with the use of copolymers containing: (i) lower contents and lengths of grafted hydrophilic chains; and (ii) higher content of grafted hydrophobic chains, with molecular weight in the range of 300–1000 g/mol. However, such results have not been correlated to solution properties.

This paper analyzes the performance of grafted amphiphilic copolymers based on hydrophobically modified polyacrylamide with poly(propylene oxide) in reducing drag (%DR) of aqueous solutions under turbulent flow. We varied the pendant chain lengths (300 and 1000 g/mol) and their mass content in the copolymer (from 5 to 35%). Besides this, we measured the size and size distribution of the particles and the intrinsic viscosity of the aqueous solutions, and correlated the results with the drag reduction performance.

2. Experimental

2.1. Reagents

Samples of polyacrylamide grafted with poly(propylene oxide) (PAAM-g-PPO) were previously synthesized and characterized (Table 1) [45]. Methanol P.A., supplied by Vetec Química Fina (Xerém, RJ, Brazil), was used as received.

2.2. Purification of the PAAM-g-PPO copolymers

To characterize the samples regarding intrinsic viscosity, molecular size and molecular size distribution, the copolymers were purified by redissolution in distilled water followed by precipitation in methanol. In a previous study, it was observed that precipitation of the polymer directly from the reaction medium is not sufficient to remove the sodium dodecyl sulfate (SDS) used in the micellar polymerization, so a second precipitation is necessary to eliminate all traces of surfactants from synthesized copolymers [29]. The purification of the copolymers after this second precipitation step was confirmed by nuclear magnetic resonance (¹³C NMR), with the disappearance of signals referring to the structure of the surfactant used.

2.3. Determination of the intrinsic viscosity [η] of the PAAM-g-PPO dispersions

The intrinsic viscosity [η] indicates the behavior of a single molecule in a solvent medium, which is related to its hydrodynamic volume [46]. The intrinsic viscosities of the copolymer dispersions

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