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Shear-flow rheology and viscoelastic instabilities of ethanol gel fuels

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ABSTRACT

This study presents a rheological investigation to understand the instability mechanism in newly formulated ethanol gel (EG) fuels when subjected to simple shear flow in a cone-plate rheometer (CPR). Shear-thinning EG fuels are formulated using an organic gellant, Hydroxypropyl methylcellulose (HPMC). EG fuels are sub-classified into concentrated, semi-dilute or dilute gels based on the consistency and elasticity number (*El*) that describes an interplay between the elastic effects and the viscous effects. The extent of shear-thinning effect in the zero-shear regime is found out by comparing shear-thinning parameter β, with an elastic parameter . The relation proves the existence of minimal elastic component even in the dilute gels. Ratio of normal stress to shear stress variation with *Re*, shows the existence of critical *Re* only for the semi-dilute and dilute gels (EG1-EG4) that marks the onset of elastic instability, whereas this transition is absent in concentrated EG5. During the shear process, two distinct phenomena are identified. First, in the low *Wi* regime, the presence of Normal stress N_1 induces the secondary flow based elastic instability, which is counterbalanced by the presence of another Normal stress N_2 to stabilize the flow. Secondly, in the high *Wi* regime, the polymer shows the maximum extension beyond which polymer breaks down with increase in the applied shear rate.

1. Introduction

Previous research on polymer-based materials were primarily driven by the need for making them suitable for several applications ranging from food technology, where they can be utilized efficiently in the form of stabilizing agents and preservative coatings [1] to pharmaceutical drug delivery purposes [2–4]. Recently, another critical area that has attracted the attention is the utilization of polymer-additives to the fuel for rocket propulsion systems. However, the polymer-based fuel for the rocket engine that yields comparatively high performance requires a proper formulation methodology followed by the rigorous flow and rheological characterization [5]. These polymeric gellant based fuels or gel fuels were obtained by adding gelling agents to the base fuel, which changed the phase of the fuels from liquid to semi-solid (high viscous, solid like material) [6,7]. Commonly, the gel fuels possess the enhanced rheo-physical property and contain the combined advantages of both liquid and solid rocket fuels. For example, at low or negligible applied shear force, the gel fuels behave like a solid or semi-solid, which allows easy handling and leakage-proof storage. Additionally, the gels in the semi-solid state are considered as the stable state at which the agglomeration of the gellants, aggregation and phase separation can be prevented. Conversely, a liquid-like behaviour at high shear enables easy pumping of the fuel at demand, thereby allowing thrust control and re-ignitability in rocket engines [8]. In this light, an initial step of the present study is to formulate organic gellant (HPMC: Hydroxypropyl methylcellulose) based ethanol gel (EG) fuels and characterize the shear rheological behaviour of EG using cone-plate rheometer (CPR).

Prior studies on gel fuel rheology using the conventional rheometers and elongational viscometers investigated the effects of viscosity as a flow governing parameter. Rahimi et al. [8] explained how the extensional viscosity played a key role in governing the elastic nature of the viscoelastic gels, which is crucial in the spray process in controlling the breakup of the droplets. In addition, the effective concentration of the gelling agent, i.e., the gellant loading rate (GLR) was found to affect the rheological nature of the gel fuels. Yarin et al. [9] focused on the rheological elongational property of the single gel simulant droplet, which forms the sub-grid unit of spray. This study investigated the significance of viscosity in the process of droplet formation, detachment, and droplet breakup. Furthermore, it was concluded that the constitutive power law of the uniaxial elongational flow was analogous to the shear-thinning behaviour exhibited by the simple shear flow. Rahimi et al. [8] and Natan [10] carried out a detailed rheological characterization for large number of gel propellants ranging from

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Nomenclature

Dimensionless groups

Re	Reynolds number, $Re = (\rho\dot{\gamma}R^2)/\eta_0$
Wi	Weissenberg number, $Wi = \lambda_c\dot{\gamma}$
β	shear-thinning parameter, $\beta = \frac{[\eta]\eta_0}{\rho_{gel}R^2}(\eta_0/\tau_0)$
λ^*	normalized relaxation time, $\lambda^* = \lambda_C/(\eta_0/\tau_0)$
ψ	stress ratio ($\psi = -\psi_2/\psi_1$)
El	elasticity number, $El = \frac{\lambda_C\eta_0}{\rho R^2} = \frac{Wi}{Re}$

Rheo-physical parameters

$\dot{\gamma}$	shear rate (s^{-1})
η	shear viscosity (Pa·s)
$[\eta]$	dimensionless intrinsic viscosity
τ,τ_0	shear stress, yield stress (Pa)
λ_c	relaxation time (s)
ψ_1, ψ_2	normal stress coefficients (Pa·s^{-2})
N_1, N_2	normal stresses (Pa)
ρ	density of the fluid (kg·m^{-3})
F_N	normal force (N)

Abbreviations

CPR	cone-plate rheometer
GLR	gellant loading rate
EG	ethanol gels
R	characteristic length

Subscripts

0	zero-shear rate
∞	infinite-shear rate
SS	stock solution

hydrazine-based fuels to kerosene using the rotational rheometer and globally classified the gel propellants as viscoelastic solid/liquid, yield thixotropic fuels based on yield stress (τ_0), thixotropy, and viscoelasticity. Arnold et al. [11] carried out the rheological characterization of inorganic gellant (fumed silica, GLR of 4–7 wt.%) loaded hydrocarbon fuels such as RP1 and JP-8, which possess definite yield stress values. It was observed that the shear-thinning index varied with GLR and Herschel-Bulkley law was used as the constitutive law for the yield stress fuels during the shear flow process. The study revealed that the physical stability of the gel fuels was a linear function of the GLR. In a separate study, rocket propellant like simulants were employed for investigating the effect of gellant type and the critical concentration of the GLR during the shear process using torsional flow device [12]. The rheological observation through visual photographic techniques reported the occurrence of the flow irregularities such as slip, shear localization, and fracture. The frequency of occurrence of flow irregularities was found to decrease with an increase in the gellant concentration of gel simulants. Similarly, when the viscoelastic fluids undergo high shear rates, it experiences the flow irregularities that can be considered as the flow instabilities. Here, the resultant flow instability depends majorly on the bulk rheological properties of the complex fluid. Likewise, it is anticipated for the viscoelastic fluids to exhibit the flow instability such as fluid expulsion from CPR due to the presence of secondary flows and the radially outward acting centrifugal force. However, the viscoelastic gels inherently feature the ability to counteract the flow irregularities because of the coupled viscosity and elastic normal stresses that restrict the sample from expelling out. The occurrence of instabilities are undesirable both in rheological studies in terms of shear localization and other practical phenomena such as drop impact (DI) (medium shear process) and atomization (high shear process). In comparison to the liquids, gel fuels behave markedly different during DI and atomization, as they tend to endure various ranges of shear. Particularly, during drop impact of Newtonian fluids, as a droplet hits the surface, it undergoes a sequential process of kinematic spreading, receding and rebounding that depend on fluid viscosity, surface tension, and other surface properties [13,14]. For the viscoelastic non-Newtonian fluids, in addition to these parameters, the droplet deformation also depends on the rheo-physical parameters such as shear viscosity (η), shear stress (τ), and yield stress (τ_0) [15,16], which make the droplet-surface interaction phenomenon complex to predict. Similarly, in an atomization process of non-Newtonian fluids, intricacies lie in selecting the appropriate gellant loading rate (GLR), mass flow rate, momentum ratios, and shear viscosity that are indeed the critical factors to govern the atomization efficiency [17–19].

The flow instabilities in gels can be categorized as viscous instability, elastic instability, viscoelastic instability and are determined based on the relative dominance of coupled competing forces of viscosity variation (due to the shear rate) and elastic effects (due to the polymer network behaviour in shear regimes). Although, rheometric flow studies provide the shear viscosity values, additional parameters such as elastic normal stresses N_1 and N_2 are also required to comprehend the elastic effects. The magnitude of τ and N_1 can be determined experimentally through direct rheological measurements. The real complexity lies in determining the N_2 values experimentally [20]. However, according to the molecular theories for dilute polymer solutions [21,22], the ratio of elastic normal stresses (N_2/N_1) can be equalled to zero or negative at the low shear rate regimes, thereby concluding that effects of N_2 can be ignored. The molecular theories did not consider the N_2 effect as the counteracting phenomenon to the elastic instability offered by N_1. Nevertheless, in the practical application, the presence of N_2 is inevitable as it stabilizes the flow during the shear flow. These normal stresses are shear dependent, it also infers that the onset of instabilities depend on the magnitude of the applied shear in addition to the rheo-physical property. The coupled stabilizing and destabilizing effect of normal stresses during the flow and shear process is the primary intent to get insight into the instabilities that occur in the yield-stress gel fuels using the conventional rheometer.

In this paper, we first present the formulation of eco-friendly, HPMC loaded ethanol gel fuels (EG). A sub-classification of the gel fuels vital in distinguishing the consistency is proposed based on the dimensionless parameters. This sub-classification is devised based on the flow parameter to detail the suitability of the gel for specific applications. We show that shear rheological interpretations in the CPR can aid in interpreting the onset of flow instabilities in gel fuel. The degree of shear-thinning of the gel fuels by varying the GLR in the low to medium range of shear is rheologically investigated. During the shear flow process, the following key factors are observed for detailing (1) elastic effect of the viscoelastic gels in low shear regimes, and (2) viscoelastic instabilities and the stabilizing mechanism during the high shear flows.

2. Materials and methods

2.1. Materials

The gel fuel samples were prepared using three components: Research grade ethanol (Purity of 99.5%; CAS No. 64-17-5) as the base fuel, Macromolecular Hydroxypropyl Methylcellulose (HPMC; CAS No. 9004-65-3) with the bulk density of $\rho_B = 689.18$ kg/m^3 as the organic gellant, and deionized water. All the chemicals were procured from Sigma-Aldrich Co. and were used without any modification.