

A novel method for improving the physicochemical properties of diesel and jet fuel using polyaspartate polymer additives

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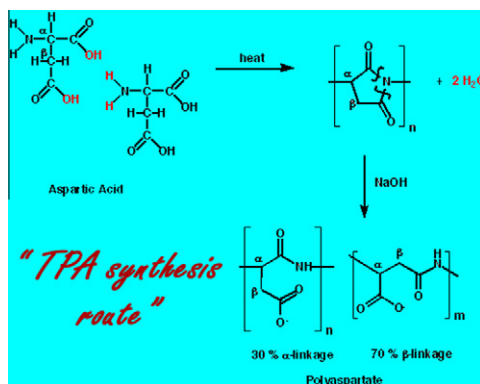
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HIGHLIGHTS

- ▶ The proposed technique improves the properties of both diesel and jet fuels.
- ▶ The used techniques proclaim important conclusions for the H₂O removal mechanism.
- ▶ Yet the technique is simple enough to be of interest in the fuel community.

GRAPHICAL ABSTRACT



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ABSTRACT

Thermal Polyaspartate Anion, a derivative biopolymer of aspartic acid, was used to remove water residues of both diesel and jet liquid hydrocarbons fuels. In this work a novel and simple method for improving the properties of fuel is reported. Different masses of TPA polymer have been mixed with a constant volume of fuel and the humidity concentration, the heat of combustion, the flash point as well the cetane index were measured according to the ASTM standard protocols. For the evaluation of the effect of the polymer additive in the physicochemical properties of the fuel, a number of ASTM standard measurements were followed. In addition, Fourier transform infrared spectroscopy (FTIR), temperature-modulated differential scanning calorimetry (TM-DSC) and thermogravimetric analysis (TGA) were used in order to evaluate the interactions between water and polymer. The removal of the humidity/water is requisite in fuel technology because of the numerous problems related to the presence of water, such as: corrosion of engine parts, pipelines and storage reservoirs, ice formation at very low temperatures and turbidity growth due to impurities. Water removal improved the physicochemical properties of both studied fuels, diesel and jet, up to 463 j/g and 1040 j/g for the heat of combustion, 28.2% and 71.4% for the total acid number and up to 39.4% and 25.7% for humidity, for diesel fuel and jet fuel respectively. Overall, the proposed method can be used in a simple fuel cleaning process using a metal mesh vessel of Thermal Polyaspartate Anion (TPA) polymer. The polymer can be replaced and re-generated as often needed.

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1. Introduction

Energy, water, food, environment, poverty, terrorism and war, disease, education, democracy and population will be the top 10 problems of our world for the next 100 years [1]. Energy policy is

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a strategy in which government decides to address the issues of energy development along with the development of the energy industry to sustain its growth; including energy production, distribution and consumption. The attributes of energy policy may include legislation, international treaties and incentives to investment [2]. In any case, nowadays petroleum and natural gas are indisputably the most important energy sources for western societies. Therefore, both for economic and environmental reasons, a very important point in current technology is the development of new processes/methods for improving the physicochemical properties of mineral-based and also bio-based fuels. According to the degree of improvement, the environmental benefits will be ipso facto; a better fuel implies fewer pollutants.

The quality of automotive fuels in the European Union is specified by standards developed by the European Committee for Standardization (CEN). The first set of CEN-ratified standards (March 1993) for automotive fuels was adopted by all member states in September 1993. Three standards cover automotive fuels quality: the EN 590 for diesel fuel, the EN 228 for gasoline and EN 589 for automotive LPG. The standards are periodically updated to reflect changes in specifications, such as the mandatory reductions in sulfur content [3]. Studying the environmental impact of petroleum combustion is one of the major tasks in oil distillation technology. Knowledge of the concentration of contaminations such as water and sulfur components is important for evaluating fuel quality and behavior during the combustion. The low combustion efficiency and the high pollutant contents of liquid hydrocarbon fuels (gasoline, diesel fuel and jet fuel) are responsible for air pollution and as a result, many investigations have focused on improvement of diesel fuel properties [4–6].

Alhamed and Bamufleh [7] reported that granular activated carbon (GAC) samples produced from date's stones by chemical activation using $ZnCl_2$, can be used in desulfurization of a model diesel fuel composed of $n-C_{10}H_{34}$ and dibenzothiophene (DBT). With GAC as adsorbate, up to 92.6% of DBT was removed in 48 h. Duarte et al. [8] reported an ultrasound-assisted oxidative desulfurization process applied in diesel oil and petroleum products. In optimum conditions, up to 99% of sulfur was removed followed by extraction with methanol. On the other hand, sulfur removal without ultrasound under the same conditions, was lower than 82% for model compounds and 55% for diesel oil samples; ultrasound improved the efficiency of oxidative desulfurization. Many other researchers reported different methods and processes for improving fuel purity and properties of both mineral based and bio-fuels [9–12].

The plurality of referred works proposes methods for desulfurization of liquids hydrocarbons fuels. Water is another major additive in liquids fuels, and is mainly responsible for corrosion, both in engines, and storage and transport reservoirs. Furthermore, ice formed at low temperatures results in fuel flow blockages, which is a major problem, especially in jet fuels. Concretely, the presence of water in fuels, causes corrosion, can cause ice blockage in the power grid that can be formed in low temperatures and last it can be divided during the warming-up and cause flame extinguishing from the produced steam (water vapor) and also the total acid number (TAN) can be decreased. The water concentration is also an indication of the degree of oxidation and is a facile method for monitoring fuel quality. The high solubility of inorganic impurities in water is another reason why the removal of water from fuels is imperative. These impurities can become nuclei for bacteria and oxide growth, both a factor in filter blocking. Turbidity is encountered with chemical additives and increases with increasing humidity [13]. Unwanted water can be added in fuel during production processes as well as in all transport procedures, therefore the best way for removal is on the transport vehicle or in any other place where fuel is combusted (burner).

Our work focused on the development of a promising novel technique for enhancement of fuel properties and can be employed as an antifouling technique. The possibility of removal of humidity from liquid fuels via hydrophilic polymers was studied and is reported here. Hydrophilic polymers (prepared by amino-acids) were used, replacing the existing mechanical methodology. In particular, according to the mechanism where charged groups of amino acids can modify the coefficient of hydration in aqueous solutions, we propose the possibility of a similar mechanism where the hydrophilicity of the TPA polymer will enhance our effort to improve petroleum distillation products. Thus, the TPA's synthesis was chosen. The TPA is a polymer with structural unit aspartic acid, that brings charged groups ($-COOH$ and $-NH_2$). The synthesis characterized as a simple route giving high purity product. In addition, the TPA is a biodegradable polymer and therefore non-polluting in use. In our study the TPA was added to diesel and jet fuel several different times and was removed using a paper filter (diameter 47 mm, pore size 0.45 μm). The humidity of treatment fuel samples was estimated directly.

2. Experimental setup and measurement

2.1. Synthesis of Thermal Polyaspartate Anion (TPA)

Thermal Polyaspartate Anion is a biopolymer synthesized from aspartic acid, a natural amino acid [14–16]. The TPA is synthesized by first heating the aspartic acid at temperatures above 180 °C and a polysuccinimide is produced. This reaction is a step of growth polymerization and water is eliminated as the only byproduct (Fig. 1). Using a catalyst, the reaction is shifted to lower temperatures and shorter reaction times. The yield of the polysuccinimide is about 97%. In the second step of the process, aqueous sodium hydroxide is added at 60 °C in order to open the polysuccinimide ring of the polyaspartate. Because the ring can open in two possible ways, two different polymer linkages can be observed, the *a*-linkage and the *b*-linkage.

2.2. Characterization of the polyaspartate samples

Both, fresh [17] (raw) and post-treatment polyaspartate polymer samples were characterized using Fourier transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA).

2.2.1. Determination of TPA molecular weight and surface properties

Gel permeation chromatography (GPC) was carried out at 25 °C using a Polymer Lab chromatographer with two Ultra Styragel linear columns (104 and 500 Å), a UV detector, PEO standards, and pure water as eluent. The BET area was evaluated by N_2 adsorption at 77 K using a commercial volumetric porosimeter (Quantachrome Autosorb-1, with Krypton upgrade). Before measurement, the sample was degassed overnight at 403 K under high vacuum ($\sim 10^{-7}$ mbar). After the mechanical grinding the average grain size evaluated using standard metal sieves.

2.2.2. Fourier transform infrared (FTIR) spectra analysis

Methods such as FTIR spectroscopy and X-ray diffraction (XRD) have been used to study many types of materials, from metals to proteins, with respect to the different vibration adsorption of spectroscopy or the interaction with electromagnetic waves. Specially, the popularity of the FTIR technique, which is mainly due to the availability of the instrumentation, has enriched the literature with every possible characteristic vibrant group in the infrared absorption frequency scale.

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