Evolution in Polymer Technology Journal
ISSN: 2642-0864

Mini-Review *Volume 2; Issue 4*

Current State of Synthesis and Use of Oxygen Generating Additives

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Received Date: December 02, 2019; **Published Date:** December 24, 2019

Abstract

The article is an overview of current methods of synthesis and use of monomeric and oligomeric oxygen-containing additives for various products including for motor fuels. Among the aliphatic alcohols, ethanol is the most commonly used oxygen-containing additive. Due to its biological origin and biochemical synthesis with high selectivity, yield and purity of the product, ethanol has a positive effect on the quality characteristics of polymer composites and the performance properties of motor fuels. Ethanol purity (99.9%) plays a crucial role in the stabilization of various composite materials including gasoline-ethanol fuels and biodiesel. Transesterified forms of natural oligomer oils (biodiesel), are widely used as additives to polymers and petroleum diesel fuels, as well as motor fuels. Based on the work of the authors, it has been shown that glycerol waste after biodiesel production can also be esterified and used as oligomeric oxygen-generating additives to various compositions and motor fuels.

Keywords: Oxygen; Content; Additive; Ethanol; Oligomers; Natural; Oils; Biodiesel; Glycerol; Transesterification; Motor Fuel

Abbreviations: MTBE: Methyl Tert-Butyl Ether; RON: Research Octane Number; MON: Motor Octane Number

Introduction

Oxygen-containing and oxygen-generating additives are widely used in various composite materials, polymers and motor fuels. They increase the rate of oxidation of different compositions during and after their use and biodegradation. In connection with the decrease in resourses of mineral oil, gas and other hydrocarbons, the question of their at least partial replacement of fuels from renewable natural raw materials - vegetable oils and bioethanol is urgent. Additives to composite materials and motor fuels, which include oxygen atoms, are widely used because they increase the product oxidation, including during motor fuel combustion. These include several classes of organic compounds such as aliphatic alcohols, ethers and esters of glycerol and higher fatty acids. An example of the use of alcohols is the additives to numerous composite materials, including mixed motor fuels, which are widely used in American and European countries. Methyl tertbutyl ether and bioethanol have already gained global application [1]. Blends of diesel and biodiesel, which is a transesterified higher fatty acid and glycerol, are well known in Europe. They are called blended or composite fuels because they are obtained by blending petroleum fuels with oxygen-containing and oxygen-generating additives based on alcohols or oligomers of vegetable oils, biodiesel or other additives.

Alcohols and Esters as Components of Blended Fuels

Methyl, ethyl, isopropyl and butyl alcohols are commonly used as oxygen-containing and oxygen-generating additives and as components of alcohol fuels. Due to the high oxygen index and octane number of alcohols, they are often introduced into oligomeric and polymeric compositions for accelerating oxidation, including low octane gasoline to improve the anti-knock properties of the latter [2] and increase combustion efficiency [3]. Table 1 shows some characteristics of alcohols and gasoline.

S No	Properties	Gasoline	Methanol	Ethanol	Butanol
1	Density at 20 °C, кg/m^3	740	792	789	809
2	Heating value, MJ/кg	46	19,5	26	38
3	Vaporization heat , кJ/кg	330	1104	850	591,2
4	Boiling point, $T_{b.}$, °C	33-156	64,7	78	117
5	Flash point, T_f°C	< 0	6	13	34
6	Ignition point ,$T_{i.}$, °C	257	440	363	345
7	Vapor pressure at 20 °C	60	37	15,8	6,1
8	The amount of air needed for burning of 1 kg of the fuel	14,9	6,52	9	12,7
9	Octane number: research octane number (RON) motor octane number (MON)	80-98 82-84	106-135 87-95	108 94	118 106

Table 1: Physical, chemical and performance characteristics of gasoline and some alcohols.

Despite the high octane (ON) of methanol, its use as an oxygen-containing and oxygen-generating additive for accelerating oxidation is not widespread due to its high toxicity, corrosion ability and high production complexity. It should also be noted that alcohol fuels are more environmentally friendly, because as a result of their combustion less toxic substances are produced in the exhaust gases [4].

Recently, leading global companies such as British Petroleum and Dupont have been testing gasoline and biobutanol based fuels. However, mixtures of butyl alcohol and methyl tert-butyl ether (MTBE) look promising enough to reduce the use of MTBE, which is abandoned by manufacturers due to its toxicity. Diesel fuel mixtures are also being investigated. The main problem with the use of such fuel is the poor miscibility of the components due to the water content (4-6%) in industrial ethanol brands and its instability at low temperatures [5-7].

The authors of [8,9] propose mixtures containing emulsifiers to improve the stability of diesel-alcohol emulsions. Various surfactants, in particular potassium stearate, a mixture of diethanol amine and oleic acid are used as emulsifiers [10]. Bench and operational studies have shown that the operation of a diesel engine on ethanol-containing fuels can reduce diesel consumption by up to 17.5 - 19% [11]. In the exhaust gas, the soot content is reduced by 11 - 72%, and the nitrogen oxides by 15 - 75% compared to the engine work on pure petroleum diesel [12].

The problem of using gasoline-alcohol mixtures can be solved in two ways: by adding stabilizers to the mixture that prevent the stratification of the mixture into components [13] or by the use of absolute water-free ethanol [14]. The use of absolute alcohol is quite promising as it improves the the fuel environmental performance. Since gasoline dissolves in alcohol, reducing the amount of alcohol in the mixture adversely affects its stability. Some authors [15,16] investigate the dependence of the ethanol-gasoline mixtures stability on the degree of ethanol absolutization and its content. Consumption of ethanol-based blended fuels is increasing. Some experts estimate that by 2020, annual ethanol production will reach 120 billion liters per year [15-17]. To reduce the cost of producing alcohol fuels, a method that combines the distillation processes of brags and the evaporation of alcohol and hydrocarbon mixtures has been developed. In the alcohol column, steam of distillate and gasoline are recycled together, which minimizes the water content in the alcohol fuel. Such gasoline is called benzanol or gasohol [18-20].

Blended fuels based on vegetable oils and their transesterified forms

The use of vegetable oils as fuel for a diesel engine has been studied by many researchers [21,22], despite their difference in certain characteristics from diesel fuel, in particular, lower calorific value, higher density and viscosity, which can cause engine knock, delay ignition formed deposits in the combustion chamber [23,24]. The authors of this article [25] stated that an increase in the amount of carbon in the ratio C:H causes an increase in the duration of ignition delay, which affects the process of oil combustion in the engine. However, thermal modification of oils by heating improves their properties and eliminates the effect on combustion characteristics by reducing the density, viscosity and surface tension of oils [26,27]. An important advantage of using oils is the reduction of nitrogen and sulfur oxide emissions [28,29]. Due to the negative effects on the engine when working on pure oils, much research is devoted to the study of the properties of mixtures of oils and diesel fuels. For such blendsdifferent plants of oil as crude and waste are used: palm, sunflower, corn, soybean, rapeseed, etc [30,31]. The use of mixtures containing from 20% to 70% soybean oil causes uneven engine operation and filter clogging. As a result of the heat treatment, the soybean oil blend gains physical andchemical properties similar to diesel fuels. But the environmental performance of such mixtures is much better than pure diesel fuels [32,33].

Sunflower oil mixtures containing 50% of the oil cause significant buildup of deposits that damage the engine. However, if the mixture contains 20% oil and 80% diesel, then the deposits are negligible and does not cause engine power loss. Such mixtures can be used for short-term engine operation [25]. The use of mixtures containing 5% oil does not cause significant changes in engine performance [34,35]. Recent studies of mixtures containing flaxseed oil have shown good results in the emission of harmful substances in the exhaust gases [36]. Operation of engines on mixtures containing 20 - 25% linseed oil does not cause engine damage. Physico-chemical properties and performance characteristics of 10% of mixtures are similar to diesel fuel [37-38].

Mixtures of diesel fuel and 5 - 10% corn oil also reduce the emission of nitrogens, sulfur and carbon (II) oxides and can be used in the operation of internal combustion engines, since they are almost indistinguishable from diesel in properties [39,40]. Good performance during short-term engine operation revealed mixtures of diesel fuel and palm oil, but long-term engine operation causes deposits and damages the fuel injection system [41].

A number of works are devoted to blends of diesel fuel with rapeseed oil, including the authors of this article [42]. The authors of [43-45] argue that the most effective is the use of mixtures containing less than 20% rapeseed oil. The use of these mixtures reduces the emissions of the most significant toxic components of the exhaust gas, and engine efficiency and fuel consumption vary within 2%, which is acceptable. [44] It is advisable to use mixtures containing up to 10% of vegetable oils. As a result of temperature treatment, the mixture with the waste oils shows better results. Such mixtures are indistinguishable from diesel in density, viscosity, combustion temperature, filtering capacity and do not significantly affect engine power and fuel consumption. The main advantage of using oil-diesel mixtures is the low toxicity of exhaust gases [46,47], which was confirmed by the authors of this article as a result of the analysis of air quality in the Trans-Carpathian region of Ukraine [36].

The transesterified form of vegetable oils, biodiesel, is devoid of these disadvantages and has properties close to those of diesel, which was confirmed by the authors of this article [48]. As is known [49], the production of biodiesel involves the use of catalysts and includes the following stages: transesterification, separation of the ester and glycerol phase, filtration and alcohol distillation. Transesterification of vegetable oils with lower aliphatic alcohols is a catalytic process. Alkaline, acidic or enzymatic catalysts [50] are used, whereby alkaline catalysts are more reactive and transesterification can occur by a mixed ion-radical mechanism [51]. Today, technologies based on homogeneous alkaline and acid catalysis are common. Solutions of alkali metal hydroxides [52-55], strong mineral acids (sulfate and hydrochloric) or alkylbenzosulfonic acids [56] are commonly used as homogeneous catalysts. However, the use of mineral acids as catalysts requires higher reaction temperatures and makes the process longer. The residues of such catalysts are corrosive to the engine parts [57].

The heterogeneous catalysis use eliminates the stage of neutralization of alkali or acid residues in solutions, but the process takes place under harsher conditions (200 - 300 ° C and pressures up to 20 atm), and some catalysts lose their activity rather quickly and require regeneration [58]. An example of solid heterogeneous catalysts is pre-dehydrated zeolite type A in an alkaline form (Li, Na, K). According to the authors [59] at the amount of catalyst 1 - 1.5%, temperature 350 - 400 ° C and molar ratio of oil: alcohol 4: 1 the reaction lasts 24 h, and the oil conversion is 96%. Obviously, such a large reaction time is a significant drawback. The use of a mixed WO_3/ZrO_2 catalyst in the transesterification with methanol at 250 ° C, at a molar ratio of methanol to 40: 1 oil, still requires 20 h of reaction to achieve an oil conversion of 90% [57-62]. Major heterogeneous catalysts such as CaO [63], MgO [64] and nano-MgO [65] have been used in supercritical transesterification to reduce the initial parameters. The results of the application of these catalysts show a significant effect of temperature on the yield and reaction time.

Biodiesel certainly has a number of advantages over traditional petroleum diesel: safety, non-toxicity, less harmful substances in exhaust gases, etc. Other benefits include potential use of renewable fuel, higher cetane numbers and better lubricating properties. The disadvantages of biodiesel include its low energy consumption, which causes a loss of engine power of 5-

10% and a change in density and viscosity during long-term storage, which can clog fuel filters [66]. Therefore, biodiesel blends with diesel have become widely used. The authors of [67] tested mixtures B10, B20 and B0 (pure biodiesel). The results of studies have shown that these mixtures meet ASTM D 6751. There was a clear correlation between the emission reductions and the biodiesel content of the mixture: emissions after using of B10 were reduced by 9.21% and after using B20 by 23.68%.

According to the authors [68], increasing the content of biodiesel in mixtures changes the density, viscosity, flash point, turbidity and filterability. The optimum content of methyl esters based on methyl esters should be less than 20% in order for the mixture to meet ASTM D 6751 and EN 14214. The composition of fatty acid residues affects the low temperature properties. However, at low content (10-20%) of biodiesel in the mixture, this effect is not observed [69]. In general, biodiesel blends with diesel have better properties than pure biodiesel or petroleum diesel. This applies to environmental performance, since it significantly reduces the exhaust fumes. An important disadvantage of using such mixtures is an increase in the amount of nitrogen oxide emissions, which, according to the authors [70-71], can be reduced by adjusting the fuel injection angle.

Glycerol waste is a technological problem for biodiesel producers. Recently, studies have been conducted towards the etherification of waste glycerol and the study of the glycerol esters characteristics as oxygenating additives to fuels [72]. Glycerol ethers, when added to diesel oil mixtures, reduce the viscosity and improve the low-temperature properties of these mixtures [73]. These compounds are also considered to be high-quality additives that can be used as fuel alone or in blends with biodiesel [74,75]. Promising as additives to motor fuels are both ethers of glycerol and its acetals. They improve the properties of motor fuels, for example, solketal has high octane mixing. The addition of only 10% solketal raises the octane number of gasoline by 9.4 units [76]. The authors of this article investigated the complex improvement of octane number of gasoline and cetane number of diesel, by dewaxing of gasoline with thiourea and diesel with urea, which is also a promising way of improving motor fuels [77].

Conclusion

- Oxygen-containing and oxygen-generating additives improve the oxidation of various composite materials, including motor fuels. Alcohol-containing fuels have many advantages, including higher octane and cetane numbers and fewer harmful exhausts. The main drawback of these fuels is their instability. However, the use of emulsifiers and absolute (99.9%) ethanol eliminates this drawback.
- Vegetable oils can be used as diesel additives only in quantities not exceeding 5-10% and at low loads, otherwise they cause engine damage.
- Vegetable oil-based biodiesel has significantly better operational and environmental performance, but its use results in reduced engine power. Therefore, the use of mixtures of diesel with biodiesel in a ratio of 1: 1 is the most effective way to reduce the amount of toxic gases into the atmosphere.
- Waste glycerol can be esterified under certain conditions and used as a fuel additive. Mixtures of diesel fuel with glycerol ethers have excellent characteristics as oxygen-generating additives to diesel fuels.

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