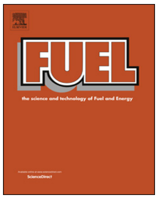
Fuel 234 (2018) 135–141

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel



Full Length Article

Evaluation of polyoxymethylene dimethyl ethers as a new type of diesel additives

Jianguang Qi, Yufeng Hu*, Jiangge Niu, Weiting Ma, Siqi Jiang, Yichuang Wang,
Xianming Zhang, Yuehui Jiang

State Key Laboratory of Heavy Oil Processing and High Pressure Fluid Phase Behavior & Property Research Laboratory, China University of Petroleum, Beijing 102249, China

ARTICLE INFO

Keywords:
Physical properties
Diesel
$PODE_n$
Measurements
Theoretical modelling

ABSTRACT

Polyoxymethylene dimethyl ethers ($PODE_n$) is an emerging biofuel that has a great potential to become one of the competitive alternative fuels for diesel engine. In the present study, systematic studies were made to evaluate the influences of $PODE_n$ on the physical fuel properties that are routinely used to define the quality of diesel fuels. Such fuel properties were measured for the three (diesel + $PODE_n$) mixtures, diesels, and $PODE_n$. The measured properties included cetane number, lubricity, density, viscosity, solidifying point, cold filter plugging point, flash point, and heat capacity. Comparisons of measured properties of the mixtures and their individual components were made to achieve the following purposes: First, to reveal the changes in the properties of diesels upon the addition of $PODE_n$ and in the properties of the (diesel + $PODE_n$) mixtures as the weight percent of $PODE_n$ is increased; Second, to see whether or not the addition of $PODE_n$ worsens the properties of diesels, in particular, whether or not the measured values of the (diesel + $PODE_n$) mixtures exceed the standard values set by the Beijing-VI. Novel theoretical equations were developed to quantitatively predict the density, viscosity, and heat capacity of the (diesel + $PODE_n$) mixtures in terms of the corresponding properties of the diesel and $PODE_n$. Comparisons were made between measured and predicted properties and the agreements are excellent.

1. Introduction

Diesel engines have been widely used because of their high thermal efficiency and high compression ratio [1]. However, the formation of soot in diesel engines during combustion has long been a problem [2]. The addition of oxygenated compounds to diesels can reduce soot formation during combustion. In particular, polyoxymethylene dimethyl ethers ($PODE_n$, *i.e.*, $CH_3O(CH_2O)_nCH_3$ with $2 \le n \le 8$) has high oxygen content, moderate boiling point, desired miscibility with diesel fuels, and average cetane number (CN) of more than 76 [3,4]. Furthermore, because the boiling point and vapor pressure of $PODE_n$ are very close to those of diesels, $PODE_n$ can be directly blended into diesels without modification to infrastructure of the diesel engines [5–7]. Therefore, $PODE_{3-8}$ has potential to serve as an environmental friendly additive to drastically reduce the soot formation inside the diesel engines [8].

The combustion and emission characteristics of the (diesel + $PODE_n$) mixtures have been extensively investigated. The amounts of fine particulate matter (PM) and NO_x released during combustion have ever been decreased by 80–90% upon the addition of 20% $PODE_{3-8}$ in the given diesels [3–5]. The combustion and emission characteristics of the (diesel + $PODE_{3-6}$) mixtures with volume blending ratios 0, 15% and 25% have been investigated in a multi-cylinder heavy duty diesel engine [9]. The blend of 15% and 25% $PODE_{3-6}$ decreased soot emissions by 88% and 95%, respectively [9]. The combustion behavior, the mechanisms of soot formation, and the emission performance of $PODE_{3-5}$ and the (commercial diesel fuels + $PODE_{3-5}$) blends with 10–30% $PODE_{3-5}$ have been investigated using a Euro 4 production diesel engine [10,11]. The tests with the multi-cylinder engine showed that the use of $PODE_{3-5}$ and [commercial diesel fuels + $PODE_{3-5}$ (50%)] simultaneously reduced NO_x and PM emissions. For the 10–12% blend that could be used in non-dedicated engines, the PM emission was decreased by about 40% [10]. The emission performance of neat $PODE_{3-5}$ and [commercial diesel fuel (90%) + $PODE_{3-5}$ (10%)] in an old Euro-2 diesel car over the NEDC driving cycle has been measured and evaluated [11]. The PM emissions thus reduced were about 18%, with a 5.9% increase in NO_x emission [11]. Diesel and the (diesel + $PODE_{3-4}$) blends with 10–30% $PODE_{3-4}$ by volume have been tested in a light-duty direct injection diesel engine without any modifications on the engine fuel supply system [6]. [Diesel + $PODE_{3-4}$ (20%)] produced lower soot emissions than diesel

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* Corresponding author.
E-mail address: huyf3581@sina.com (Y. Hu).

https://doi.org/10.1016/j.fuel.2018.07.007
Received 7 May 2018; Received in revised form 19 June 2018; Accepted 2 July 2018
0016-2361/ © 2018 Published by Elsevier Ltd.

and [diesel + $PODE_{3–4}$ (10%)] and slightly lower NO_x emissions than [diesel + $PODE_{3–4}$ (30%)] [6].

The following physical fuel properties are routinely used to define fuel quality; however, these properties of the (diesel + $PODE_n$) mixtures are seldom investigated [7]. Density, viscosity, solidifying point (SP), and cold filter plugging point (CFPP) are important properties of diesels [12–18]. They affect the start of injection, the injection pressure, and the fuel spray characteristics, thereby influencing the engine performance, combustion and exhaust emissions [13]. Because diesel injection equipment meters by volume, the change of diesel density affects the output power of diesel engine [12]. The density of the diesel also influences production, transportation, and distribution processes as well as all processes that are involved in the internal combustion engine [14]. Viscosity of fuels affects the operation of fuel injection equipment, particularly when the increase in viscosity at low temperatures affects the fuel's fluidity [17,18]. Higher viscosity causes poorer atomization of the fuel spray and less accurate operation of the fuel injectors [18]. When the temperature is near the SP of the diesel, the viscosity increases rapidly, which has a greater impact on the flow properties of the diesel. Flash point (FP) is an important property that can be used to evaluate the stability of diesels. Although the height of the FP does not directly affect the combustion performance of the diesel, the high FP is more secure in the storage of diesel fuel, the processes of fuel treatment and transportation [19,20]. Lubricity should be taken into consideration for most types of diesel fuel injection equipment, because these types of pumps depend totally on the fuel for lubrication of their moving parts [21]. CN is widely used to evaluate the ignition quality of fuels [22–25]. It measures the readiness of the fuel to auto-ignite when injected into the engine [24]. A fuel with a high CN has a short ignition delay period and starts to combust shortly after it is injected into an engine [25]. Increasing CN improves fuel combustion and tends to reduce NO_x and PM emissions [25].

Therefore, in this study the properties including CN, lubricity, density, viscosity, SP, CFPP, FP, and heat capacity of given diesels, $PODE_n$, and (diesel + $PODE_n$) mixtures were systematically measured. Systematic comparisons were made to reveal the influences of $PODE_n$ on the above-mentioned properties of the diesels. New equations for prediction of the density, viscosity, and heat capacity of the (diesel + $PODE_n$) mixtures from the corresponding properties of the diesels and $PODE_{3–7}$ were developed.

2. Materials and methods

2.1. Chemicals

Analytical grade of dimethyl sulfoxide, dimethylformamide, *N*-methylpyrrolidone, ethanol, *n*-propyl alcohol, *n*-butyl alcohol, and benzene were supplied by Aladdin Industrial Corporation (Shanghai, China), without further purification. These compounds were stored in a desiccator prior to use. Deionized water was distilled in a quartz still. Fluid catalytic cracking diesel oil (FCC) and the diesel oils that can be used above 0 °C (0# diesel) and between −10–0 °C (−10# diesel) were provided by Sinopec Beijing Yanshan Company. Their properties at 25 °C are shown in Table 1. It is clear that FCC has more unsaturated components than 0# and −10# diesels. $PODE_n$ was provided by Yuhuang Company. Its mass distribution is $PODE_3$:$PODE_4$:$PODE_5$:$PODE_6$:$PODE_7$ = 41.57%:31.20%:17.10%:8.42%:1.71%. The properties of neat $PODE_n$ are shown in Table 2. The diesel's property data of regulation of the Beijing-VI (DB11/239–2016) [26] are shown in Table 3.

2.2. CN measurements

The CN values were measured by Shanghai Microspectrum Chemical Technology Service Co., Ltd and Qingdao Kebiao Testing & Research Institute Co., Ltd. The method GB/T 386-2010 was used. The uncertainty of the CN measurements is 0.9.

Table 1
Properties of the fuels considered at 25 °C.

Property	FCC	0#	−10#	Determination Method
Density[a]/($g·cm^{-3}$)	0.96882	0.83152	0.82915	DMA-4500M
Viscosity[b]/($mm^2·s^{-1}$)	5.5920	3.0221	2.6603	GB/T-265
FP[c]/°C	95	71	64	GB/T-261
SP[d]/°C	−16	−22	−20	GB/T-510
CN[e]	< 30.0	48.0	–[f]	GB/T-386
Water content	< 50 ppm	< 50 ppm	< 50 ppm	Karl Fischer
Saturation fraction[g]	0.49975	0.97397	0.97941	SH/T-0509
Aromatic fraction[g]	0.47658	0.01475	0.01388	SH/T-0509
Resins fraction[g]	0.02367	0.01128	0.00671	SH/T-0509
C_P/($J·g^{-1}·°C^{-1}$)[h]	1.32	2.04	2.13	DSC

[a] The combined expanded uncertainty (U_c) is U_c (ρ) = $5.0·10^{-5}$ $g·cm^{-3}$ (0.95 level of confidence).

[b] U_c(η) = 0.02 $mm^2·s^{-1}$ (0.95 level of confidence).

[c] U_c(FP) = 1 °C (0.95 level of confidence).

[d] U_c(SP) = 1 °C (0.95 level of confidence).

[e] U_c(CN) = 1 (0.95 level of confidence).

[f] Not detected.

[g] U_c = 0.01 (0.95 level of confidence).

[h] The experimental temperature is 20 °C, and U_c (C_P) = 0.02 $J·g^{-1}·°C^{-1}$ (0.95 level of confidence).

Table 2
Properties of components in $PODE_n$ with *n* ranging from 2 to 6 [2,9].

Formula	$CH_3O–(CH_2O)_n–CH_3$				
Value of n	2	3	4	5	6
Density/($g·cm^{-3}$)[a]	0.96	1.02	1.07	1.10	1.13
Melting point/°C	−69.7	−42.5	−9.8	18.3	48.0
Boiling point/°C	105	156	202	242	280
CN	63	78	90	100	104
Oxygen content/wt%[b]	45.2	47.0	48.1	48.9	49.5
C_P/($J·g^{-1}·°C^{-1}$)[c]	1.93				

[a] At 25 °C.

[b] Weight percent.

[c] Measured in this work at 20 °C and 0.1 MPa.

Table 3
The diesel's property data of regulation of the Beijing-VI (DB11/239-2016) [26].

Diesel	0# diesel	−10# diesel
Density[a]/$g·cm^{-3}$	0.820–0.845	0.820–0.845
Viscosity[a]/$mm^2·s^{-1}$	2.5–7.5	2.0–7.5
FP/°C	> 60	> 60
SP/°C	< 0	< −10
CFPP/°C	< 4	< −5
CN	> 51	> 51
Abrasion resistance/μm	< 460	< 460

[a] The density and viscosity values at 20 °C.

2.3. Lubricity measurements

Abrasion resistance was measured by Qingdao Kebiao Testing & Research Institute Co., Ltd. The method SH/T-0765 was used. The uncertainty of the abrasion resistance measurements is 1 μm.

2.4. Density measurements

Density (ρ) measurements were performed using the methods described in our previous studies [27–30]. The Anton Paar oscillating-tube digital densimeter (DMA-4500) was used. The temperature and instrument accuracies are ± 0.01 °C and ± $5·10^{-5}$ $g·cm^{-3}$,

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