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High pressure physicochemical properties of 2-methylfuran and 2,5-dimethylfuran – second generation biofuels



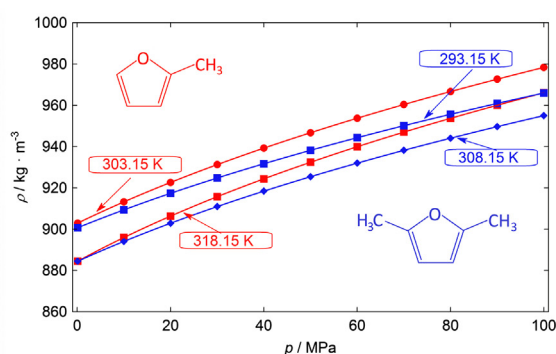
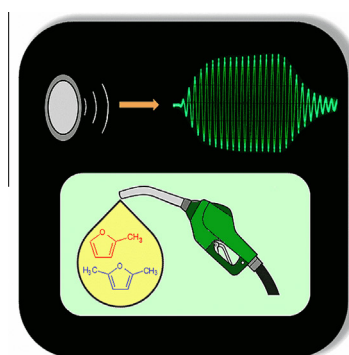
Sylwia Jęzak, Marzena Dzida*, Michał Zorębski

Institute of Chemistry, University of Silesia, Szkolna 9, 40-006 Katowice, Poland

HIGHLIGHTS

- 2-Methylfuran (MF) and 2,5-dimethylfuran (DMF) were investigated.
- Speed of sound was measured at high pressure.
- T and/or p can compensate differences in density and compressibility of MF and DMF.
- Isentropic compressibilities of MF and DMF are between gasoline and diesel fuels.

GRAPHICAL ABSTRACT



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ABSTRACT

The two compounds of the furan family such as 2-methylfuran (MF) and 2,5-dimethylfuran (DMF) are studied. These furan heterocycles can be obtained from cellulosic biomass and are indicated as a potential bioethanol or petroleum-based fuels substitute. Optimization of the injection process requires knowledge of densities and compressibilities of fuel as well as effect of temperature and pressure on this properties. For this reason the speeds of sound in MF and DMF were measured at temperatures from 293 K to 318 K and pressures from 0.1 MPa to 101 MPa. The densities were measured for MF and DMF under atmospheric pressure in the temperature range from 273 K to 333 K and 273 K to 363 K, respectively. The isobaric heat capacities were measured at atmospheric pressure for temperatures from 293 K to 323 K. The densities, isobaric heat capacities, isentropic compressibilities, isothermal compressibilities, and isobaric thermal expansions for temperatures from 293.15 K to 318.15 K and for pressures up to 100 MPa were calculated using acoustic method. In this work effect of temperature and pressure on density, isentropic compressibility, and isobaric thermal expansion of MF and DMF have been discussed. This paper also examined density, isentropic compressibility, and isobaric thermal expansion of MF and DMF compared to isooctane, ethanol, ekodiesel ultra, and biodiesel. The results obtained show that differences between densities and isentropic compressibilities of MF and DMF can be compensated by temperature and/or pressure. Comparison of thermophysical properties of MF and DMF with fuels and biofuels shows that MF and DMF can be considered as a fuel additive.

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1. Introduction

Escobar et al. [1] pointed out that the transportation sector in the world consumes about 47% of primary energy deriving from fossil fuels. The energy demand is increasing all the time due to

* Corresponding author.

E-mail address: mhd@ich.us.edu.pl (M. Dzida).

the rapid development of motorization and industrialization, while petroleum reserves are diminishing. Moreover, the combustion of the fossil fuels are the major source of greenhouse gas emission [2,3]. This situation forces to search other, alternative and renewable sources of energy such as biomass [4,5].

The renewable biomass-derived fuels have reached worldwide interest [6]. So far three generations of biofuels have been distinguished [5,7]. First generation biofuels are derived from edible feedstocks [5,7]. The second generation biofuels are produced mainly from lignocellulosic biomass, non-food feedstocks [5,7]. The third one is obtained mainly from algae biomass [5,7–9]. Since 2007, when Román-Leshkov et al. [10] proposed new method of production of DMF from biomass with low energy consumption and high yield [11], promising candidates as second-generation biofuels, derived from lignocellulosic biomass are furans such as MF and DMF. Román-Leshkov et al. [10] synthesized DMF in two-step catalytic conversion process of D-fructose by means of an intermediate 5-hydroxymethylfurfural. Binder and Raines [12] also synthesized DMF from lignocellulosic biomass in two chemical reactions. Thananattachon and Rauchfluss [13] converted fructose derived from lignocellulose into DMF using formic acid. Furan derivatives can be obtained through the pyrolysis of D-glucose [14] as well as fast pyrolysis of lignocellulosic-biomass [15].

Besides of attractive method of production, these compounds are very promising as separated biofuels or additives of fuel due to appropriate fundamental physicochemical properties for a fuel such as initial boiling point, research octane number (RON), energy density, heat of vaporization, and immiscibility with water [12,16]. Tian et al. [17] reported that the laminar flame speed of DMF was very similar to gasoline whereas Wu et al. [18] noticed that for blend containing 20 vol.% of DMF + 80 vol.% of isooctane + air, the laminar flame speed is higher than those for isooctane + air mixtures. Moreover, Tian et al. [17] noticed that the laminar burning velocity of DMF is closer to gasoline than ethanol. Liebergesell et al. [19] wrote that using MF reduced hydrocarbon emissions and increased efficiency in comparison with model gasolines. Eslami et al. [20] noticed that DMF amended lubricity of gasoline and ethanol, while Hu et al. [21] found that DMF in mixtures with gasoline reduced friction of engine elements. Thewes et al. [22] analyzed the impact of MF on mixture formation and combustion in a direct-injection spark-ignition (DISI) engine. These authors found that the knock resistance of MF at full load was significantly better in comparison with gasoline. Wang et al. [23] researched the DISI engine performance, combustion characteristics and emissions of MF compared to DMF, gasoline and ethanol. They found that in terms of the volumetric indicated specific fuel consumption, MF was close to DMF and gasoline and about 30% less than ethanol. Wei et al. tested MF in spark-ignition engines (SI) [24] as well as in DISI system [25], and DMF was examined by Daniel et al. [26].

DMF was also considered as a potential additive to diesel fuel such as ethanol or butanol [27]. These compounds are oxygenated biofuels, and therefore their ability to form soot precursors during their combustions is less than that of fossil fuels [27,28]. Chen et al. [27] found that DMF was better than butanol and gasoline as a diesel additive in term of reducing soot emissions. It is the effect of prolonged ignition delay which improves atomization process, among others obtaining atomic oxygen. Atomic oxygen bound in the fuel is more efficient in reducing soot emissions in comparison with molecular oxygen [27]. Zhang et al. [28] also found that DMF addition to diesel (up to 40%) caused reduction of soot emission nearly to zero. Zhang et al. [29] reported that the 2% 2-ethylhexyl nitrate (EHN) added into blend of 40 vol.% of DMF with 60 vol.% of diesel (D40) decreased the soot emission by 80% in comparison with D40.

Despite this strong interest of MF and DMF, the knowledge of their physicochemical properties is still insufficient. Densities of the MF for the temperatures 293.15 K, 298.15 K and 303.15 K were reported by Murakami et al. [30], density of MF at 293.15 K were published in two works [31,32]. Densities of MF and DMF at 293.15 K and 298.15 K were reported by Loras et al. [33]. In case of DMF density dependence on temperature was published by Mejía et al. [34] for the temperature range from 293.15 K to 358.15 K and da Silva and Aznar [35] for the temperature range from 288.15 K to 333.15 K. Verevkin and Welle [36] reported density of DMF at 293.15 K and isobaric heat capacity of DMF at 298.15 K. Isobaric heat capacity of MF at 298.15 K was reported by Carlson and Westrum [37]. Recently, Lomba et al. [38] measured speed of sound, density, refractive index as well as surface tension, dynamic viscosity and static permittivity of MF and DMF at temperatures from 278.15 K to 338.15 K under atmospheric pressure. Moreover, these authors studied $p\rho T$ behavior of MF and DMF at pressures up to 60 MPa and temperature range from 283.15 K to 338.15 K.

Taking into consideration that MF and DMF were found as a very promising gasoline additive and were also tested as a diesel additive, the knowledge of their high pressure thermophysical properties is crucial for these investigations, because the maximum pressure of fuel in the DISI engine is about 20 MPa [39] or 30 MPa [40], while in the common rail is about 250 MPa [41].

The main goal of present work is to study the effect of pressure on thermophysical properties of MF and DMF as an extension of our systematic investigation of high pressure behaviour of fuel [41], biofuels [41], and biofuel components [42–44]. The high pressure properties of MF and DMF are also examined in comparison with isooctane, ethanol, ekodiesel ultra, and biodiesel. In this paper new experimental speeds of sound in MF and DMF measured in the temperature range from 293 K to 318 K and pressures from 0.1 MPa to 101 MPa as well as the isobaric heat capacities measured for temperatures from 293 K to 323 K under atmospheric pressure are reported. Additionally, the densities measured under atmospheric pressure in the temperature range from 273 K to 333 K for MF as well as from 273 K to 363 K for DMF are presented. From experimental data, the densities, isobaric heat capacities, isentropic compressibilities, isothermal compressibilities, and isobaric thermal expansions at pressures up to 100 MPa for temperatures from 293 K to 318 K have been calculated. A method based on the suggestion of Davis and Gordon [45] with a numerical procedure proposed by Sun et al. [46] has been applied for calculations. This acoustic method, based on the experimental speed of sound, is the only one which allows direct determination of isentropic compressibility. The isentropic compressibility seems to be the most useful in estimation of the fuel injection timing because the fuel injection in engine is approximately an adiabatic process. To the best of our knowledge speed of sound in MF and DMF under high pressure as well as temperature dependence of isobaric heat capacity at atmospheric pressure have never been investigated.

2. Experimental section

2.1. Materials

2-Methylfural and 2,5-dimethylfuran were purchased from Sigma-Aldrich and used without further purification. The chemical abstract registry number (CAS number) of chemicals used in this work and their basic physicochemical properties are included in Table 1.

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