



## Full Length Article

# Experimental investigations on the thermophysical properties of methyl myristate in alcoholic solutions



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## ABSTRACT

Due to the limitations of using biodiesel directly or blended with petroleum diesel, alcohols are always employed as the additives to improve the certain properties of biodiesel. In this work, experimental investigations are conducted on the thermophysical properties, especially the density and viscosity, of binary mixtures containing the component of the biodiesel, namely methyl myristate, with higher alcohols, including 1-propanol, 1-butanol and 1-pentanol at atmospheric pressure from 303.15 K to 333.15 K. The Vogel-Fulcher-Tammann equation is introduced to correlate the viscosity. The derived property of thermal expansion coefficient is calculated based on the experimental density. The excess properties are obtained to further understand the interactions between the ester and the alcohols. The positive value of the excess molar volume and negative value of viscosity deviation are observed, indicating that the increasing contribution of the repulsive forces between methyl ester and the alcohols.

## 1. Introduction

Due to the depletion of the conventional non-renewable diesel and the rising anthropogenic emissions of green-house gases, biodiesel is recognized as the promising alternative source and has been widely explored in the recent years [1]. Biodiesel is composed of fatty acid methyl/ethyl esters (e.g. methyl myristate, ethyl myristate), and typically derived from the common oils like animal fats or vegetable oils by transesterification with alcohols [2–4]. Biodiesel is non-toxic, more environmentally friendly and importantly, renewable and bio-degradable compared to fossil diesel [2].

Biodiesel can be used in current engines without further more changes and, has considerable combustion performances. However, the limitations to use biodiesel directly or blended with petroleum diesel are the cold flow properties when temperature goes below 273.15 K and the increasing emissions of  $\text{NO}_x$  [5,6]. To reduce the negative influences, investigations have focused on the alcohols as the additives to improve the certain properties (e.g. viscosity, density, and surface tension et al.) of biodiesel. For instance, short-chain alcohols, such as methanol and ethanol, are introduced and studied; but the blend of biodiesel or diesel with ethanol would exhibit phase separation when the temperature is under 283.15 K [6,7]. It is shown that with the increase of the carbon in the alcohols, the properties of the blends will be improved because the alcohols possess high calorific values and have good solubility with biodiesel [8]. Therefore, more studies have

conducted the use of higher alcohols, e.g. 1-propanol, 1-butanol, 1-pentanol, as the additives blended with biodiesel [9–12].

It is well known that the thermophysical properties of pure biodiesel or mixtures with chemicals are of great importance for the scientific investigations as well as the scale-up design and optimization. Much attention has given to the systems of biodiesel with short-chain alcohols. Barabas et al. reported the volumetric properties for the binary mixtures of rapeseed oil with ethanol at atmospheric pressure [13]. However, little attention has given to the investigations on the thermophysical properties of fatty acid methyl esters with these higher alcohols. Laza et al. researched the basic properties such as viscosity, heating value, density, and volatility characteristics for the blends of vegetable oil with higher alcohols (1-propanol, 2-propanol, isobutanol, 1-butanol, and 2-butanol) [14].

In this work, the thermophysical properties of densities and viscosities for the binary mixtures of fatty acid methyl ester (i.e. methyl myristate) with higher alcohols (i.e. 1-propanol, 1-butanol and 1-pentanol) are measured at atmospheric pressure (0.0967 MPa) from 303.15 K to 333.15 K to provide the experimental data for the further studies or industrial processes in which these binary mixtures are involved. The excess properties and the derived properties are calculated to understand the relationship between the ester and the alcohols.

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**Table 1**  
Specifications of the ester and alcohols studied in this work.

Substance	CAS No.	Chemical formula	Initial mass fraction purity
Methyl myristate	124-10-7	CH <sub>3</sub> (CH <sub>2</sub> ) <sub>12</sub> COOCH <sub>3</sub>	> 0.98
1-Propanol	71-23-8	CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> OH	≥ 0.995
1-Butanol	71-36-3	CH <sub>3</sub> (CH <sub>2</sub> ) <sub>3</sub> OH	> 0.995
1-Pentanol	71-41-0	CH <sub>3</sub> (CH <sub>2</sub> ) <sub>4</sub> OH	> 0.99

## 2. Experimental section

### 2.1. Materials

The details of the ester and alcohols conducted in this work are given in Table 1. The substances were purchased from Shanghai Aladdin biochemical technology Co., Ltd (Aladdin), and the mass fraction was better than 0.98. The chemicals were used in the measurements without further purification.

The samples of binary mixtures composed of ester with alcohols were prepared by gravity using the analytical balances (AB204-N and ME204, Mettler-Toledo) with an accuracy of 0.0001 g.

### 2.2. Density measurement

The densities of the pure chemicals and the binary samples were conducted by an Anton Paar digital vibrating U-tube densimeter (DMA 5000 M) at atmospheric pressure (0.0967 MPa) with the temperatures ranging from 303.15 K to 333.15 K. The repeatability of the vibrating densimeter supplied by the manufacturer was  $1.0 \cdot 10^{-6} \text{ g}\cdot\text{cm}^{-3}$ . The relative standard uncertainty of the density was specified as 0.001. The densimeter was well-cleaned and dried before and after the experimental measurements using the chemicals of acetone and methanol. Also, the densimeter was calibrated using the dry air and the bidistilled water provided by the Anton Paar. During the experimental measurement, the density was measured in triplicate.

### 2.3. Viscosity measurement

The viscosities of the pure substances and the binary mixtures were determined using an Ubbelohde capillary viscometer (EGV 700 and EGV 702) provided by LAUDA-Brinkmann, LP. Before and after the experimental measurement, the viscometer was well-cleaned using acetone/methanol and dried. The capillary viscometer was calibrated using viscosity standards purchased from Cannon Instrument Company (State College, PA, USA). When the capillary viscometer was filled with the sample, it was then kept in a water bath conducted by a Lauda thermostat (LAUDA ECO Silver, Germany). The uncertainty for the temperature was within 0.01 K. An electronic stopwatch was applied to record the efflux time with the uncertainty of 0.01 s. The value of the

**Table 2**  
Viscosities and densities of methyl myristate studied in this work and in the literature.

T/K	This work		Literature	
	$\rho/\text{kg}\cdot\text{m}^{-3}$	$\eta/\mu\text{Pa}\cdot\text{s}$	$\rho/\text{kg}\cdot\text{m}^{-3}$	$\eta/\mu\text{Pa}\cdot\text{s}$
303.15	859.500	3510.02	859.9 [15], 859.7 [20]	3543.0 [15], 3496 [16], 3510 [18]
308.15	855.717	3132.36	856.0 [15]	3165.1 [15]
313.15	851.939	2815.27	852.2 [15], 851.7 [19], 851.6 [20]	2844.7 [15], 2841 [16], 2800 [18]
318.15	848.165	2542.82	848.4 [15]	2570.9 [15]
323.15	844.396	2309.06	844.6 [15], 843.3 [20]	2334.3 [15]
328.15	840.628	2108.54	840.8 [15]	2129.5 [15]
333.15	836.858	1930.58	837.0 [15], 835.2 [20]	1949.8 [15], 1943 [17], 1910 [18]

The standard uncertainties ( $u$ ) are  $u(T) = 0.01 \text{ K}$  and  $u_c(\rho) = 0.001$ . The combined relative expanded uncertainty ( $U_{c,\rho}$ ) is  $U_{c,\rho}(\eta) = 0.03$  with a 0.95 level of confidence.

viscosity was obtained by the multiplication of corresponding density and the efflux time with the viscometer constant. The relative expanded uncertainty of the viscosity studied in this work was specified to be less than 0.03 with the confidence level of 0.95.

## 3. Results and discussions

### 3.1. Experimental data

The measured thermophysical properties of densities and viscosities for methyl myristate as well as those in the literature are presented in Table 2 [15–20]. The values for 1-propanol are given in Table 3 [21–25], for 1-butanol in Table 4 [22,24,26–28], for 1-pentanol in Table 5 [22,29,30]. Tables 6–8 summarize the experimental data of the binary mixtures. Figs. 1 and 2 depict the relative deviations between the literature properties and calculated values using the following Eqs. (1) and (2) fitted with the experimental data measured in this work, respectively.

Generally, the density is considered as a function of temperature and can be fitted by:

$$\rho = a_1 + a_2 \cdot T + a_3 \cdot T^2 \quad (1)$$

where  $\rho$  ( $\text{kg}\cdot\text{m}^{-3}$ ) is the density;  $T$  (K) is the temperature; the fitted parameters of  $a_1$  ( $\text{kg}\cdot\text{m}^{-3}$ ),  $a_2$  ( $\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-1}$ ) and  $a_3$  ( $\text{kg}\cdot\text{m}^{-3}\cdot\text{K}^{-2}$ ) are correlated by the measured data in this work and listed in Table 9.

The Vogel-Fulcher-Tammann (VFT) equation is introduced to correlate the viscosity data of the pure chemicals and binary mixtures using the following equation:

$$\eta = \eta_0 \exp[k/(T - T_0)] \quad (2)$$

where  $\eta$  ( $\mu\text{Pa}\cdot\text{s}$ ) is the viscosity;  $T$  (K) is the temperature;  $\eta_0$  ( $\mu\text{Pa}\cdot\text{s}$ ),  $k$  (K), and  $T_0$  (K) are the fitted parameters correlated with the measured viscosities and listed in Table 10.

The VFT equation is usually used to characterize the glass-forming liquids. The parameter of  $T_0$  is considered as the “ideal glass transition temperature”. When the temperature is below the “ideal glass transition temperature”, an equilibrium glass should be existed where the mass-transferring motions are frozen [31].

The average absolute relative deviation (AARD) is always applied to evaluate the divergence and is calculated as:

$$\text{AARD}(\%) = \frac{100}{N} \sum_{i=1}^N \left| \frac{(M_{\text{cal},i} - M_{\text{lit},i})}{M_{\text{lit},i}} \right| \quad (3)$$

where  $N$  is the number of measured value points,  $M_{\text{cal}}$  and  $M_{\text{lit}}$  are the calculated data using the correlation equations and literature data, respectively.

The AARD for the densities of pure methyl myristate between the literature values and calculated data by Eq. (1) is 0.05%, for the viscosities by Eq. (2) is 0.82%. It is clear that the measured data of the

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