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# Gibbs Energy Additivity Approaches for Isentropic Compressibility of Biodiesels (Ethyl Ester)

**N Sirimongkolgal**
Faculty of Engineering and Industrial Technology, Bansomdejchaopraya Rajabhat University, Bangkok, Thailand;

**S Phankosol**
Faculty of Engineering and Industrial Technology, Bansomdejchaopraya Rajabhat University, Bangkok, Thailand

**T Chum-in**
Key Laboratory of Thermal Fluid Science and Engineering of MOE, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, China.;

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***Abstract*** Isentropic compressibility is important for injection timing of liquid fuel. The isentropic compressibility of biodiesels are correlated to number of carbon atoms, number of double bond(s) and temperature. In this work, an empirical approach for isentropic compressibility of biodiesels can be estimated by using number of carbon atoms (of fatty acid, z) and number of double bonds (nd). The proposed equations are easy to use and the estimated isentropic compressibility values of biodiesels at different temperatures agree well with the literature values. The bias and average absolute deviation of isentropic compressibility values of biodiesels at 293.15 to 343.15 K are 0.00 and 0.41 %, respectively. The isentropic compressibility outside temperature between 293.15 to 343.15 K may be possibly estimated by this model but accuracy may be lower.

*Index Terms* - Biodiesel, Gibbs Energy, Isentropic Compressibility.

## 1. INTRODUCTION

Biodiesel is renewable energy for diesel engine obtained from the transesterification of alkyl monoesters of fatty acids prepared from vegetable oils, animal fats, waste greases and waste cooking oil. Besides being produced from green renewable/sustainable sources, contributes less to greenhouse gases emissions and it can replace petrodiesel totally or partially in conventional diesel engines without modification. [1,2] The chemical component of biodiesel may vary not only from country to country, but they may also vary from batch to batch. [3] Alkyl esters are important component of biodiesel, consist of chains of about 16–18 carbon atoms with varying degrees of saturation.[4] The fatty acid profile of biodiesel corresponds to that of parent oil or fat and is a major factor influencing its fuel properties. Isentropic compressibility ($K_s$) is important injection timing of liquid fuel, as well as its indirect effect on engine noise and pollutant formation. [5] Douhéret et al. [6] were approach to determining $K_S$ of a liquid was from the density and speed of sound data via the Newton - Laplace Equation,

$$K_s = \frac{1}{\rho U^2} \tag{1}$$

where $\rho$ is density (kg/m$^3$)
$U^2$ is speed of sound (m/s)

Experimental determination of speed of sound and density of biodiesels from different sources and processes is time consuming, but they are necessary for mathematical models. A good mathematical model should provide not only the accurate surface tension value but it should also correlate the chemical structure of the estimated substance for further development or refinement of the model. Krisanangkura et al. [7] proposed correlation of isentropic compressibility and free energy additivity, shown general equation in equation (2):

$$\ln K_s = \ln C \frac{\Delta G_{k_s}}{RT} \tag{2}$$

where $G_{ks}$ is the change in Gibb's energy associated with isentropic compressibility; C is constant; R is universal gas constant; $T$ is absolute temperature.

Equation (2) had been applied for estimation of isentropic compressibility of different fatty acid methyl esters (FAMEs) and biodiesels by Krisanangkura et al. [7], shown average absolute percent difference of 16 different biodiesels (neat and blended) was 0.92%.

This work presents and evaluates a simple model, based on that presented by Krisanangkura et al. [7], applied to describe the isentropic compressibility of fatty acid ethyl esters (FAEEs) using the number of carbon atoms and double bonds present in fatty acid ethyl ester molecule.

## 2. THEORY

*Isentropic compressibility of fatty acid ethyl ester*

For a fatty acid ethyl ester compound having the molecular structure of $CH_3–(CH_2)_{z-1}–COOHC_2H_5$, Applied divided the molecule into different groups from Martin's equation [8] modified by Phankosol et al. [9]; $COOHC_2H_5$, $CH_2$, $CH_3$. The free energy of transfer from solution to gas of the molecule was derived from the sum of the free energies of all the contribution groups.

$$\Delta G = \Delta G_f + z\delta G + n_d \delta G_{db} \tag{3}$$

where $\Delta G_f$, $\delta G$ and $\delta G_{db}$ are assigned for the free energy of interfacial interaction of functional group, change in free energy/carbon atom and change in free energy/double bond.

From basic thermodynamics,

$$\Delta G = \Delta H + T\Delta S \tag{4}$$

Eq. (5) is obtained by combining Eqs. (2)-(4).

$$\ln K_s = \ln C - \frac{\Delta H_f}{RT} + \frac{\Delta S_f}{R} - \frac{z\delta H_f}{RT} + \frac{z\delta S_f}{R} - \frac{n_d \delta H_{db}}{RT} + \frac{n_d \delta S_{db}}{R} \tag{5}$$

Grouping

$$\ln K_s = a + bz + \frac{c}{T} + \frac{dz}{T} + n_d e + \frac{n_d f}{T} \tag{6}$$

where

$$a = \ln C + \frac{\Delta S_f}{R} \quad (6.1)$$

$$b = \frac{\delta S_f}{R} \quad (6.2)$$

$$c = -\frac{\Delta H_f}{R} \quad (6.3)$$

$$d = \frac{\delta H_f}{R} \quad (6.4)$$

$$e = \frac{\delta S_{db}}{R} \quad (6.5)$$

$$f = -\frac{\delta H_{db}}{R} \quad (6.6)$$

*Isentropic compressibility of biodiesel*

Allen et al. [10] used the Dalton- type mass-average equation (Eq. (7)) for calculation of Isentropic compressibility of biodiesels.

$$K_{s_{Biodiesel}} = \sum_{i=1}^{n} x_i K_{s_i} \quad (7)$$

where $x_i$ and $Ks_i$ are mole or mass fraction of component (FAEE) $i$ and isentropic compressibility of $FAEE_i$. Eq. (8) is obtained by combining Eqs. (6) and (7),

$$\ln K_s = a + \frac{c}{T} + z_{ave}(b + \frac{d}{T}) + n_{d(ave)}(e + \frac{f}{T}) \quad (8)$$

where

$$z_{ave} = \frac{\sum_{i=1}^{n} x_i z_i}{\sum_{i=1}^{n} x_i} \quad (9)$$

$$n_{d,ave} = \frac{\sum_{i=1}^{n} x_i n_{d,i}}{\sum_{i=1}^{n} x_i} \quad (10)$$

## 3. METHODOLOGY

*Experimental data*

The experimental data density and speed of sound for calculating isentropic compressibility of fatty acid ethyl esters (FAEEs) and biodiesels were obtained from obtained from Freitas *et al*. [11]

*Numeric constants of equation (6)*

The six numeric values of equation (6) (*a*, *b*, *c*, *d*, *e* and *f*) for FAEEs were solved by multiple liner regression according to Phankosol et al.[12]. Multiple linear regressions method was easily carried out on Microsoft Excel spreadsheet and 2016 version was chosen in this study.

*Statistical analysis*

The average absolute deviations (AAD), bias, standard error ($\sigma_{\bar{x}}$) and coefficient of determination ($R^2$) were calculated by equations (11), (12), (13) and (14), respectively:

$$AAD(\%) = \frac{100}{N} 1\sum_{i=1}^{N}\left[\left|\frac{P_{\exp} - P_{cal}}{P_{cal}}\right|\right] \quad (11)$$

$$Bias(\%) = \frac{100}{N} 1\sum_{i=1}^{N}\left[\frac{P_{\exp} - P_{cal}}{P_{cal}}\right] \quad (12)$$

$$\sigma_{\bar{x}} = \frac{\sigma}{N} \quad (13)$$

$$R^2 = \left[\frac{N\sum_{i=1}^{N} P_{\exp}P_{cal} - \left(\sum_{i=1}^{N} P_{\exp}\right)\left(\sum_{i=1}^{N} P_{cal}\right)}{\sqrt{\left[\sum_{i=1}^{N} P^2{}_{\exp} - \left(\sum_{i=1}^{N} P_{\exp}\right)^2\right]\left[\sum_{i=1}^{N} P^2{}_{cal} - \left(\sum_{i=1}^{N} P_{cal}\right)^2\right]}}\right]^2 \quad (14)$$

where $P_{exp}$ stands for experimental value reported elsewhere, $P_{cal}$ is the calculated value and N is the number of data points.

## 4. RESULTS AND DISCUSSION

*Isentropic compressibility of short chain saturated FAEEs*

Generally, the linear free energy relationship in Eq. (3) is limited to a narrow range of carbon numbers for many physical properties of FAEEs] Knotts et al. [13] also used two parachor values for methylene groups. One was for methylene of short chain length (C1–C11) and another for long chain length (C12 and longer). Thus, in this study, FAMEs were divided into two groups, C5:0–C12:0 and C12:0–C22:0 or higher.

$$\ln K_s = 17.5454 - \frac{973.923}{T} + z(0.118605 + \frac{27.1938}{T}) \quad (15)$$

where *Ks*, *z* and *T* are isentropic compressibility ($Pa^{-1}$), the number of carbon atoms on the fatty acid chain and absolute temperature (K), respectively.

$$n_{d,ave} = \frac{\sum_{i=1}^{n} x_i n_{d,i}}{\sum_{i=1}^{n} x_i}$$

The isentropic compressibility of short chain saturated FAEEs (C4–C12) generated from equation (15) at temperature between 293.15 and 343.15 K shown percent differences (in parentheses) between the literature [11] and the calculated values. The AAD is 1.62. The highest and lowest relative deviation (D (%)) are ±2.78 and ±0.03, respectively and shown D (%) in figure 1.

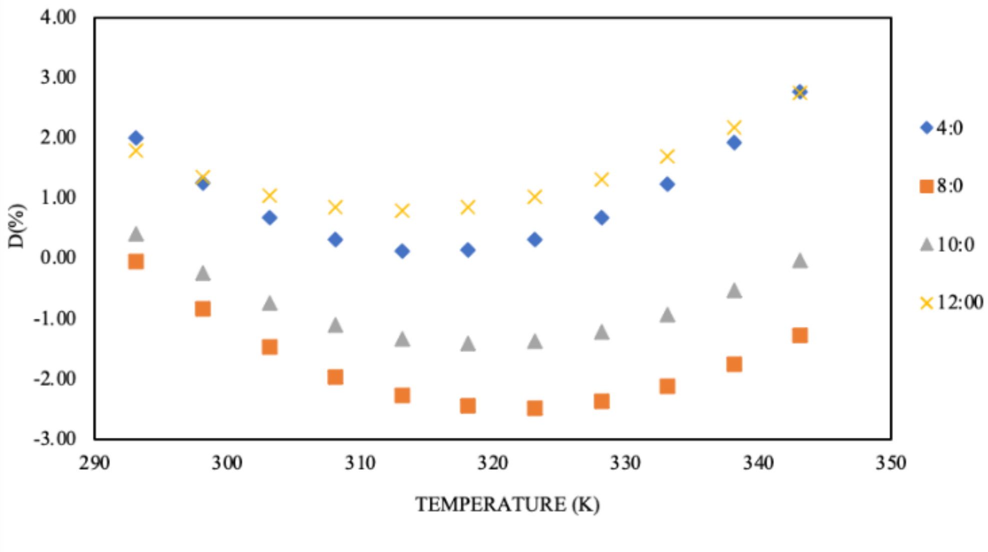


Figure 1
Relative deviations (D(%)) between *Ks* of short chain saturated FAEEs (C4–C12) by equation (15) and the experimental values reported by Freitas *et al*. [11]

The plot between the calculated ($Ks_{cal}$) and literature ($Ks_{(lit)}$) values is linear with the intercept, slope, $R^2$ and standard error of 0.1889, 0.9776, 0.9906 and 0.016, respectively.

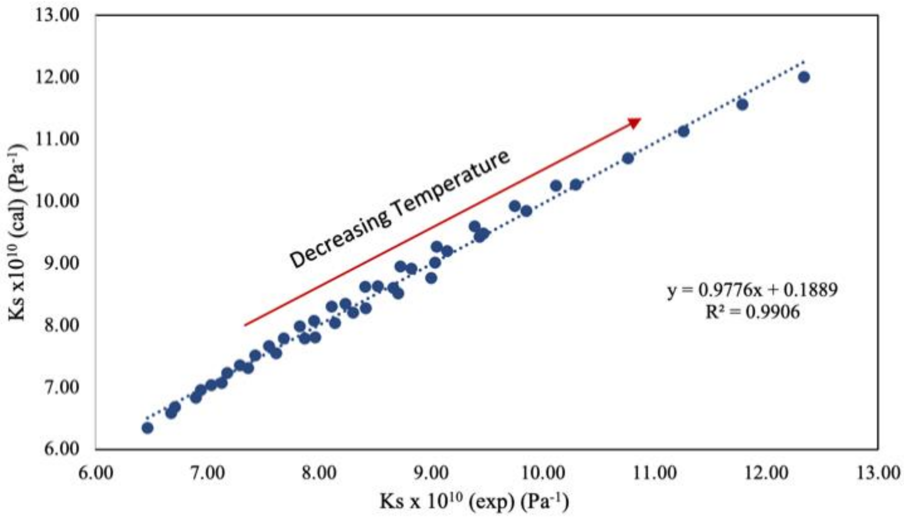


Figure 2
Correlation of the estimated Ks to the experiment values short chain saturated FAEEs (C4–C12) [11] at 283.15 to 363.15 K

*Surface tension of long chain saturated and unsaturated FAEEs*

Long chain fatty acids are major components found in naturally occurring fats and oils and they are feed stock of biodiesel. However, long chain saturated fatty acids occurred in nature are both saturated and unsaturated with different number of double bonds. [9]. All six numeric constants of equation (6) were solved by multiple linear regression according to Phankosol et al. [12] The equation for estimating isentropic compressibility ($Pa^{-1}$) is shown in equation (16):

$$\ln K_s = 19.0003 - \frac{606.467}{T} + z(0.00433 + \frac{1.4817}{T}) - n_d \left( 0.07103 + \frac{13.392}{T} \right) \quad (16)$$

where $Ks$, $z$, $n_d$ and $T$ are isentropic compressibility ($Pa^{-1}$), the number of carbon atoms on the fatty acid chain, number of double bonds and absolute temperature (K), respectively.

The estimated Ks of long chain saturated and unsaturated FAEEs at 293.15-343.15 K by equation (16) are summarized in table 1. Percent differences between the calculated and literature isentropic compressibility values are listed in the parentheses, shown D (%) in figure 2. The calculated isentropic compressibility values for both saturated and unsaturated FAEEs agree well with the literature values. The overall AAD and Bias were 0.64% and 0.18%, respectively. The highest difference was 2.08% (ethyl myristate at 343 K).

Table 1
Estimated isentropic compressibility ($Pa^{-1}$) of long chain saturated and unsaturated FAEEs at 293.15-343.15 K by Eq. (22). Numbers in parentheses are D (%) between the calculated and the literature values [11].

| Temp. (K) | Isentropic compressibility ($Pa^{-1}$) x$10^{10}$ | | | | |
|---|---|---|---|---|---|
| | 14:0 | 16:0 | 18:0 | 18:1 | 18:2 |
| 293.15 | 6.2051 | 6.0897 | 5.9765 | 5.8269 | 5.6811 |
| | *1.04** | - | - | *1.25* | *1.22* |
| 298.15 | 6.4318 | 6.3133 | 6.1969 | 6.0372 | 5.8816 |
| | *0.62* | - | - | *0.73* | *0.76* |
| 303.15 | 6.6589 | 6.5373 | 6.4178 | 6.2478 | 6.0822 |
| | *0.33* | *-0.46* | - | *0.33* | *0.43* |
| 308.15 | 6.8862 | 6.7615 | 6.6391 | 6.4585 | 6.2829 |
| | *0.17* | *-0.71* | - | *0.06* | *0.20* |
| 313.15 | 7.1137 | 6.9859 | 6.8605 | 6.6693 | 6.4834 |
| | *0.13* | *-0.84* | - | *-0.11* | *0.09* |
| 318.15 | 7.3412 | 7.2104 | 7.0820 | 6.8800 | 6.6837 |
| | *0.20* | *-0.86* | *-1.39* | *-0.17* | *0.09* |
| 323.15 | 7.5686 | 7.4348 | 7.3034 | 7.0905 | 6.8838 |
| | *0.38* | *-0.77* | *-1.37* | *-0.12* | *0.20* |
| 328.15 | 7.7957 | 7.6590 | 7.5247 | 7.3007 | 7.0834 |
| | *0.65* | *-0.58* | *-1.25* | *0.03* | *0.39* |
| 333.15 | 8.0226 | 7.8830 | 7.7458 | 7.5106 | 7.2826 |
| | *1.05* | *-0.29* | *-1.04* | *0.28* | *0.68* |
| 338.15 | 8.2491 | 8.1066 | 7.9666 | 7.7201 | 7.4813 |
| | *1.52* | *0.10* | *-0.74* | *0.62* | *1.07* |
| 343.15 | 8.4751 | 8.3297 | 8.1869 | 7.9290 | 7.6793 |
| | *2.08* | *0.57* | *-0.37* | *1.03* | *1.52* |
| Bias (%) | 0.74 | -0.43 | -1.03 | 0.36 | 0.60 |
| AAD (%) | 0.74 | 0.57 | 1.03 | 0.43 | 0.60 |
| Overall Bias (%) | | 0.18 | | | |
| Overall AAD (%) | | 0.64 | | | |

*%(exp.-cal.)/exp.

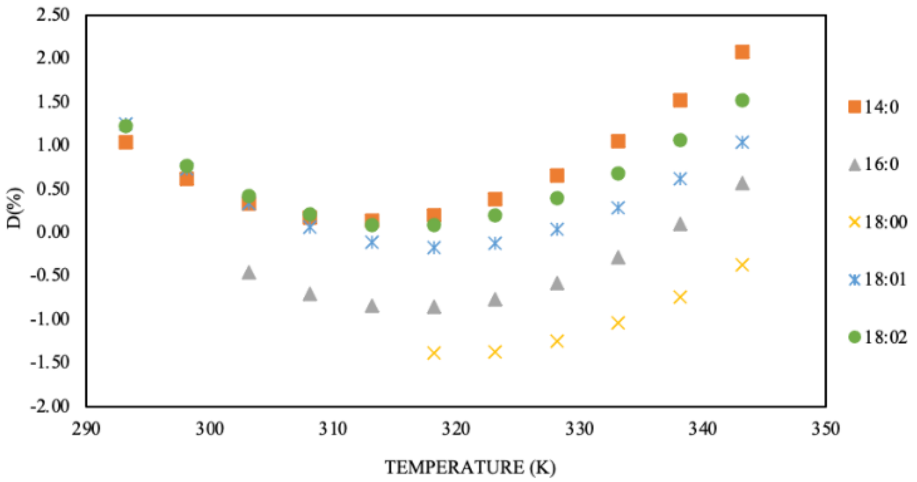


Figure 3

Relative deviations (D(%)) between *Ks* of long chain saturated and unsaturated FAEEs by equation (16) and the experimental values reported by Freitas *et al*. [11]

In addition, plot of $Ks_{cal}$ against $Ks_{lit.}$ values of both saturated and unsaturated FAEEs is linear with a slope, intercept, $R^2$ and the standard error are 0.9891, 0.0645, 0.9934 and 0.006, respectively.

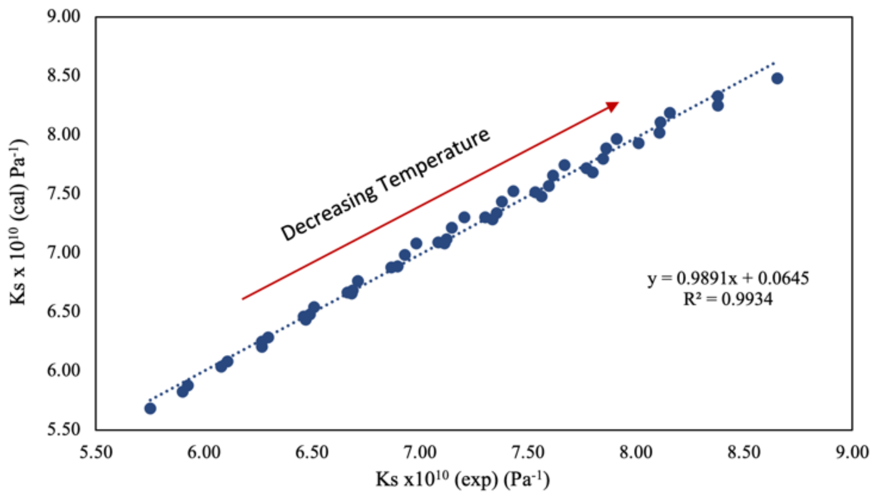


Figure 4

Correlation of the estimated *Ks* to the experiment values long chain saturated and unsaturated FAEEs [11] at 283.15 to 363.15 K

*Isentropic compressibility of biodiesel*

Commercial biodiesels are produced after transesterification of vegetable oils or animal fats using methanol or ethanol in the presence of a catalyst such as sodium hydroxide. Therefore, biodiesels are mixture of FAEEs of different compositions and vary according to the sources. As it was pointed out earlier that the isentropic compressibility of biodiesel, in this study, was simply determined by the rule of Dalton- type mass-average equation as shown in equation (7). Isentropic compressibility can be directly estimated without a prior knowledge of individual ester when the average carbon number ($z_{ave}$) and the average number of double bonds ($n_{d,ave}$) are used in place of z and nd. The $z_{ave}$ and $n_{d,ave}$ are calculated according to equations (9) and (10), respectively. Thus, only fatty acid composition and *Ks* of individual FAEE is required for the calculation. The *Ks* of individual FAEE was estimated in the previous section. FAEEs compositions of several biodiesels were reported by Freitas et al. [11]. They are summarized FAEEs compositions in Table 2. Also, the $z_{ave}$ and $n_{d,ave}$ of each biodiesel are included in the table 2. The range of $z_{ave}$ and $n_{d,(ave)}$ of soybean (S) sunflower (Sf) soybean+sunflower (S+B) palm(P) of biodiesels are 17.177 to 17.919 and 0.662 to 1.479, respectively.

Table 3
Mass fraction (%) of different biodiesels.

| FAEEs | Mass fraction | | | |
|---|---|---|---|---|
| | S | Sf | S+B | P |
| C8:0 | 0 | 0 | 0 | 0.03 |
| C10:0 | 0 | 0 | 0 | 0.03 |
| C12:0 | 0 | 0 | 0.03 | 0.42 |
| C14:0 | 0.07 | 0.09 | 0.3 | 0.72 |
| C16:0 | 10.92 | 5.66 | 11.81 | 38.67 |
| C16:1 | 0.08 | 0.09 | 0.16 | 0.15 |
| C18:0 | 2.93 | 3.11 | 3.23 | 4.49 |
| C18:1 | 27.45 | 35.32 | 27.53 | 44.51 |
| C18:2 | 52.65 | 54.46 | 49.9 | 10.29 |
| C18:3 | 4.96 | 0.28 | 5.87 | 0.26 |
| C20:0 | 0.29 | 0.2 | 0.31 | 0.25 |
| C20:1 | 0.18 | 0.13 | 0.2 | 0.1 |
| C22:0 | 0.37 | 0.49 | 0.44 | 0.04 |
| C22:1 | 0 | 0.04 | 0.08 | 0.03 |
| C24:0 | 0.099 | 0.14 | 0.15 | 0.02 |
| $z_{ave}$ | 17.81 | 17.92 | 17.79 | 17.18 |
| $n_{d,ave}$ | 1.48 | 1.45 | 1.45 | 0.66 |
| SN | 182.68 | 181.71 | 182.88 | 187.39 |
| IV | 121.60 | 119.38 | 119.54 | 54.19 |

The isentropic compressibility of different biodiesels at temperature between 283.15 to 363.15 K are summarized in table 3 and shown D (%) in figure 5. It can be seen that the calculated isentropic compressibility ($Ks_{cal}$) are in good agreement with the literature values ($Ks_{lit}$) at all temperatures.

Table 3
Estimated isentropic compressibility ($Pa^{-1}$) of four biodiesels at 293.15-343.15 K by Eq. (16). Numbers in parentheses are D (%) between the calculated and the literature values [11].

| Temp. (K) | Isentropic compressibility ($Pa^{-1}$) x$10^{10}$ | | | |
|---|---|---|---|---|
| | S | Sf | S+B | P |
| 293.15 | 5.7671 | 5.7647 | 5.7717 | 5.9227 |
| | *0.61* | *0.72* | *0.97* | *0.79* |
| 298.15 | 5.9729 | 5.9706 | 5.9778 | 6.1376 |
| | *0.11* | *0.24* | *0.47* | *0.29* |
| 303.15 | 6.1789 | 6.1768 | 6.1842 | 6.3529 |
| | *-0.27* | *-0.06* | *0.10* | *-0.09* |
| 308.15 | 6.3850 | 6.3830 | 6.3905 | 6.5683 |
| | *-0.53* | *-0.29* | *-0.16* | *-0.36* |
| 313.15 | 6.5911 | 6.5892 | 6.5969 | 6.7838 |
| | *-0.68* | *-0.45* | *-0.29* | *-0.50* |
| 318.15 | 6.7971 | 6.7952 | 6.8031 | 6.9993 |
| | *-0.72* | *-0.51* | *-0.32* | *-0.54* |
| 323.15 | 7.0028 | 7.0010 | 7.0091 | 7.2146 |
| | *-0.65* | *-0.42* | *-0.23* | *-0.47* |
| 328.15 | 7.2081 | 7.2065 | 7.2148 | 7.4297 |

| | | | | |
|---|---|---|---|---|
| | *-0.48* | *-0.27* | *-0.06* | *-0.29* |
| 333.15 | 7.4131 | 7.4116 | 7.4200 | 7.6445 |
| | *-0.22* | *-0.01* | *0.22* | *-0.02* |
| 338.15 | 7.6176 | 7.6162 | 7.6248 | 7.8588 |
| | *0.13* | *0.33* | *0.58* | *0.35* |
| 343.15 | 7.8215 | 7.8203 | 7.8290 | 8.0727 |
| | *0.56* | *0.74* | *1.02* | *0.80* |
| Bias (%) | -0.20 | 0.00 | 0.21 | 0.00 |
| AAD (%) | 0.45 | 0.37 | 0.40 | 0.41 |
| Overall Bias (%) | | 0.00 | | |
| Overall AAD (%) | | 0.41 | | |

Biodiesel from sunflower (Sf) showed the lowest AAD of 0.37%, while biodiesel from soybean (S) gives the highest AAD of 0.45%. The overall Bias and AAD of 4 types of biodiesels were 0.00% and 0.41%, respectively. However, when the *Ks* of biodiesel was determined by Dalton-type mass-average equation, the weight factor of individual FAEEs was required for the calculation

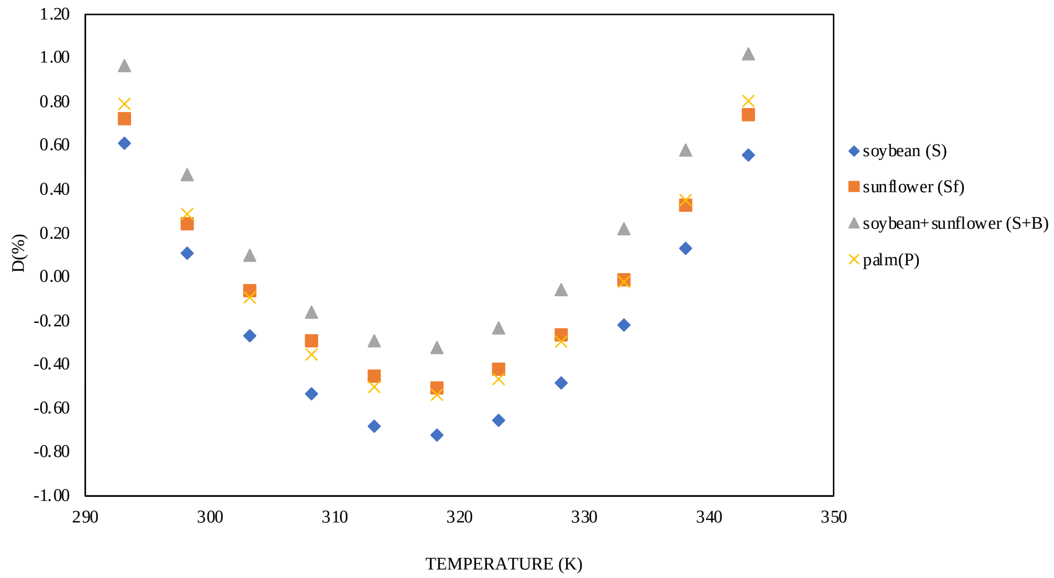


Figure 5

Relative deviations (D(%)) between *Ks* of four biodiesels by equation (16) with $z_{ave}$ and $n_{d,ave}$ and the experimental values reported by Freitas *et al*. [11]

The correlation between the estimated and literature isentropic compressibility values of biodiesels is shown in figure 6. The slope and intercept of the plot is 0.8035 and 5.2636 with the $R^2$ of 0.9276 and the standard error was 0.3977.

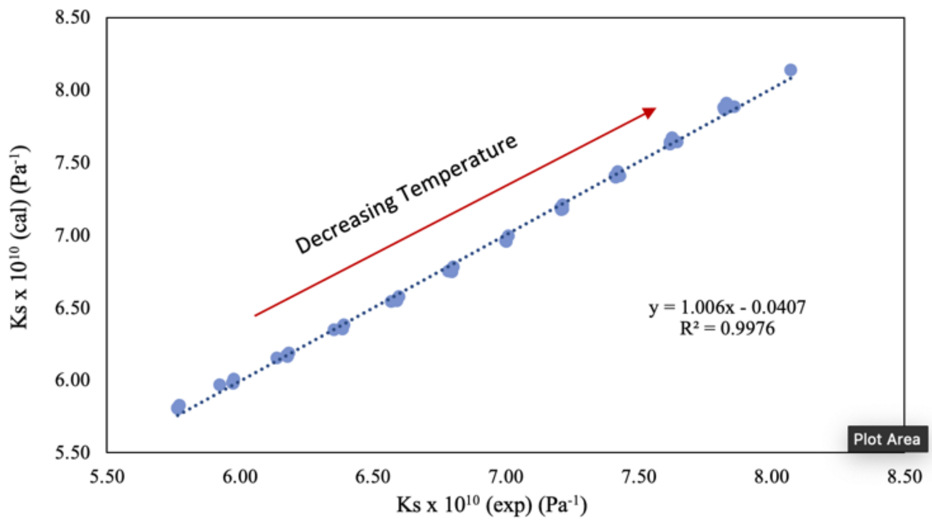


Figure 6
Correlation of the estimated *Ks* to the experiment values four biodiesels by equation (16) with $z_{ave}$ and $n_{d,ave}$ and the experimental values reported by Freitas *et al.* [11]

Although equation (16) is a good and simple model for estimating biodiesels at different temperature, determine of the z and $n_d$ requires the knowledge of FAEEs composition. The analysis must be done with gas chromatography (GC) or high-performance liquid chromatography (HPLC). The saponification number (SN) and iodine value (IV) have long been used for the characterization of fats and oils and they require no special instrumentation. z and $n_d$ can be converted to SN and IV according to equations (17), (18) and (19) [12]

$$n_d = \frac{IVxM}{25400} \tag{17}$$

$$z = \frac{M + 2n_d - 46}{14} \tag{18}$$

$$M = \frac{56000}{SN} \tag{19}$$

Equation (20) is derived from equation (16) with equations (17), (18) and (19) and can be used to estimate isentropic compressibility of biodiesels at different temperatures from the SN and IV values:

$$\ln K_s = 18.9861 - \frac{17.32}{SN} - \frac{601.65}{T} - \frac{5927.1}{TxSN} - \frac{0.1584xIV}{SN} + \frac{29.1361xIV}{TxSN} \tag{20}$$

The isentropic compressibility of pure, mixed, and total biodiesels estimated by equation (20) (at different temperatures) were very close to those calculated using the $z_{ave}$ and $n_{d,ave}$ (equation (16)). The slight differences in the calculated values between equation (16) and equation (20) are due to the conversion equation in eq (16) and equation (20), which are approximate. The correlation between the reported density and estimated values using equation (20) (44 data points) is linear with the slope, intercept, $R^2$ and standard error of 1.006, 0.021, 1.00, and 0.001, respectively.

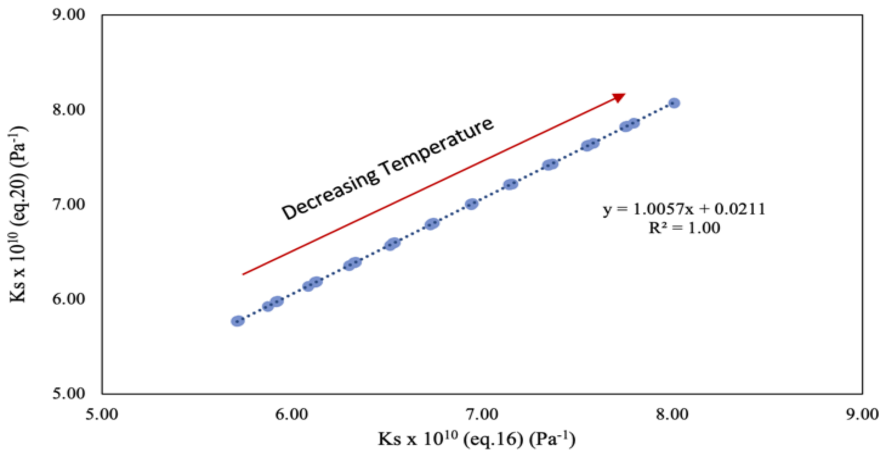


Figure 7
Correlation of the estimated *Ks* to the estimation values four biodiesels by equation (16) with $z_{ave}$ and $n_{d,ave}$ and the estimation values by equation (20).

## CONCLUSION

This work provides an empirical correlation of isentropic compressibility of a liquid to its chemical structure or chemical composition at different temperatures (293.15-343.15 K). Hence, isentropic compressibility of a biodiesel can be predicted either from the $z_{ave}$ and $n_{d,ave}$ (eq. (16)) of fatty acids or from the SN and IV (eq (20)) with approximately the same accuracy.
Besides the accuracy, both models provide two additional advantages: (1) isentropic compressibility of biodiesel can be estimated without a prior knowledge of the isentropic compressibility of individual FAEEs, and (2) all the coefficients of the equations are well-defined. This would allow further refinement of the models.

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