

Prediction of Freezing Point of Jet Fuel Compounds using Group Contribution Method

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Freezing point (T_f) is the temperature at which a liquid substance turns into a solid when cooled and this can be determined experimentally or by modelling. For aviation fuels, the freezing point test is relevant as impeding fuel flow may have disastrous results on planes, such as interference with fuel atomization. The freezing point of jet fuel can negatively be affected due to certain components found in the fuel as blend. In this study, a modelling method called Group Contribution method (GC) is used to predict the freezing point of jet fuel. The GC approach was tested and verified using different chemical data set. This research consisted of predicting the freezing point of 232 compounds using the first order-groups, second order-groups and third order-groups identified from the molecular structure and proving the validity of the model using 20 test data set of compounds. The regression using linear regression curve fitting using MATLAB was performed to obtain the contribution values for each group. The statistical analysis giving a standard deviation (SD) of 22.22 K, an average absolute error (AAE) of 15.62 K, an average relative error (ARE) of 7.23 % and a correlation coefficient, R^2 of 0.8829. The model was tested and verified to be an appropriate method to predict the freezing point of jet fuel liquid compounds.

1. Introduction

Jet fuel, derived from hydrocarbon sources and organic chemicals, is commonly used in aviation engines, and consists of formulated refined products from crude oil and organic compounds. Its properties, such as boiling point, freezing point, melting point, viscosity, density, and heat of combustion, must be considered to avoid hazardous effects on the engine (Chevron Corporation, 2006). High-density jet fuel with lower temperatures is more suitable for higher altitude applications. Isomerization of tetrahydrodicyclopentadiene and tetrahydrotricyclopentadiene, can reduce the freezing point and high viscosity of high-density fuels (Nie et al., 2021). Duong et al. (2016) measured the freezing point of mixtures (soap-derived biokerosene (SBK)/ butylcyclohexane, dodecane/ butylcyclohexane, and dodecane/ butylbenzene) using the JIS K2276 test method. The finding from their study showed that the freezing point of butylcyclohexane+SBK and butylcyclohexane+ dedocane are having a freezing points in a range of $-5\text{ }^{\circ}\text{C}$ to $-20\text{ }^{\circ}\text{C}$, which is out of the target range of the jet fuel standard. According to the ASTM D1655 standard, the commercial kerosine type jet fuel (Jet A1) must has the maximum freezing point of $-47\text{ }^{\circ}\text{C}$. Experimentally determining the properties of fluid substances is time-consuming, expensive, or not feasible due to thermal decomposition issues and in product design aspect, the potential chemicals might not be commercially available. Freezing point of organic compounds are also challenging to measure experimentally due to their thermal lability and difficulty in measuring critical properties. The physico-chemical property of a compound can be predicted if a reliable model is available and suitable to be applied for the system or selected compounds.

Yunus et al. (2022) performed a study on green jet fuel blend using waste cooking oil to identify a reliable model for freezing point and flash point of jet fuel blend. Their study utilised the derived models for jet fuel blend freezing point property prediction obtained from literature (AlMulla and Albahri, 2017). Their results on the green

jet fuel blended with the commercial aviation fuel, Jet A1 fuel that met with the ASTM standard. Yarveicy et al. (2014) provided a least square support vector machine (LSSVM) prediction model to calculate the freezing point depression of different electrolyte solutions. The model was compared with the CPA (cubic-plus-association) Equation of State (EoS) combined with the Debye-Hückel electrostatic term. The model was accurate with R^2 0.9992 while CPA-EoS has a slightly higher absolute deviation (0.02 – 0.75) compared to LSSVM model (0.01 – 0.50). The model however requires a computational extensive for the machine learning task and applicable for range of trained electrolyte solutions. Alonso et al. (2011) utilised excess Gibbs energy model for predicting the freezing point of multicomponent mixture. Their model focussing on the electrolyte and non-electrolytes for food applications. Gharagheizi et al. (2014) has done the prediction for freezing point of 17,000 organic compound using group contribution model. Their model utilised simple molecule structures and substructures to predict the freezing point which resulting a squared correlation coefficient of 0.735. Another study for estimation of freezing point using group contribution method was conducted by Lazzús (2016) which focusing on ionic liquid. The models involved cation and anion groups with a correlation coefficient R^2 0.9138. Marrero and Gani (2001) introduced a group contribution (GC) method to estimate the properties of pure compounds using three level of groups, from simple groups (1st order) to the higher order groups involved polyfunctional and structural groups (2nd and 3rd order). They successfully demonstrate the methods for prediction of normal boiling point, critical temperature, critical volume, critical pressure, standard enthalpy of vaporization, standard enthalpy of formation, standard Gibbs energy, normal melting point and standard enthalpy of fusion. Later, Hukkerikar et al. (2012) introduced the GC+ method by combining the GC and atom connectivity index (CI) for 18 pure component properties including boiling point, melting point, flash point, etc. Frutiger et al. (2016) demonstrate the group contribution approach for estimation of flammability-related properties such as Lower and Upper Flammability Limits (LFL and UFL), Flash Point (FP) and Auto Ignition Temperature (AIT). The method proves that it can be used for property prediction with R^2 values within a range of 0.72 – 0.99. Yunus and Zahari (2017) applied the same approach to estimate the heat of combustion for fuel, and Kashinath et al. (2020) extend the method for cetane number prediction. Therefore, a reliable estimation model is needed to determine the freezing point property of fuel components or any substances. GC method can provide quick estimation with less computational effort. Due to that reason, the GC approach introduced by Marrero and Gani (2001) were utilized to develop the freezing point model. The aim of this study is to develop a model for the prediction of freezing point of jet fuel by using the group contribution approach.

2. Methodology

2.1 Data collection

A good quality data used for the property model estimation is very vital to ensure the accuracy and reliability of the model. It is therefore very important to select a reliable database for model development. This database contains compounds from common families that are suitable as jet fuel such as hydrocarbon, alcohol, acid, ether, aromatic and also polyfunctional compounds. About 252 molecules with freezing point values were collected from literature (Yaws, 1999), which divided into 232 training data set and 20 test set. These molecules were broken down into three level of groups (1st, 2nd and 3rd order groups) based on the listed group in Marrero and Gani (2001). The frequency of each group appear in the molecule was counted. For the first order, the group -CH₂ frequency is 278 times, -CH₃, 233 times, aCH 105 times, -OH 85 times, -CH_{2cyc} 71 times, -CH 44 times, CH₂O 26 times, -COOH 25 times, CH₃O, -Br, -Cl are 17 times, CH₃COO 16 times, aC-CH₃ 14 times, ac-OH, CH₂COO are 10 times, and other groups are less than 10. For the functional groups that are appeared frequently such as aromatic aCH, alcohol -OH, ether CH₂O, acid COOH, and ester -CH₃COO has high tendency to provide more accurate result. For group that appear once might have high relative error in the prediction.

2.2 Property function selection

The selection of a suitable function for the freezing point prediction is important to ensure it best fit the experimental data and postulate a reliable prediction capability. The selection was performed based on the trend of the freezing point data and occurrences of CH₂ groups that shows a linear relationship, hence the freezing point model is represented as Eq(1).

$$T_F - T_{Fo} = \sum N_i T_{F1i} + w \sum M_j T_{F2j} + z \sum O_k T_{F3k} \quad (1)$$

where, T_F is the predicted freezing point, T_{Fo} is the universal constant, N_i , M_j and O_k are the occurrences, T_{F1i} , T_{F2j} and T_{F3k} are the freezing point contributions for the first, second and third order groups. Similar function was observed for the critical volume, standard Gibbs energy, standard enthalpy of formation and standard enthalpy of formation from study by Marrero and Gani (2001).

2.4 Parameter regression

The parameter regression was done using the least-square regression method with the goal of minimizing the sum of squares representing the disparities between the experimental and estimated values of the freezing point. The parameter regression for freezing point contributions T_{F1i} , T_{F2j} and T_{F3k} was carried out in three stages. The first stage was to determine the T_{F1i} and T_{F0} values where w and z were set as zero. The first stage results were used for the second stage to determine the second order contribution, T_{F2j} . The last stage was to determine the contributions for the third order group, T_{F3j} by utilising the results from the first and the second stage.

2.4 Statistical analysis

The statistical analysis was done by evaluating the standard deviation (SD), the average absolute error (AAE), and the average relative error (ARE) using Eq(2) to Eq(4). These parameters are important where AAE measures the average of the errors' magnitude between the predicted and actual values. Meanwhile, ARE is the ratio of the absolute error of the measurement to the actual measurement. ARE can determine the magnitude of the absolute error in terms of the actual size of the measurement.

$$SD = \sqrt{\frac{\sum(X_{pred} - X_{exp})^2}{N}} \quad (2)$$

$$AAE = \frac{1}{N} \sum_j |(X_j^{exp} - X_j^{pred})| \quad (3)$$

$$ARE = \frac{1}{N} \sum_j \left| \frac{(X_j^{exp} - X_j^{pred})}{X_j^{exp}} \right| \times 100 \quad (4)$$

where X_{pred} represents the predicted freezing point, X_{exp} is the experimental value of freezing point and N is the number of compounds.

2.5 Model validation

The freezing point prediction model is verified using test data set of 20 chemicals. These chemicals were selected from the collected data and separated from the training data set used in the regression. The obtained contributions were used to predict the freezing point and compared with the experimental T_F value. It is to validate the accuracy of the model in estimating the freezing point of a chemical in jet fuel.

3. Results and discussion

The result of regression is tabulated in Table 1 (first order group), Table 2 (second order group) and Table 3 (third order group). Based on the collected data set, Table 1 shows only 81 out of 220 groups from the first order having the estimated contribution, while 40 out of 130 groups from the second order (Table 2) and five (5) out of 74 groups from the third order groups. The comparison on the predicted and experimental freezing point from the regression of training data set is given in Figure 1a, considering all three levels of group that gave R^2 value of 0.8829. The testing dataset is shown in Figure 1b with R^2 value of 0.8384.

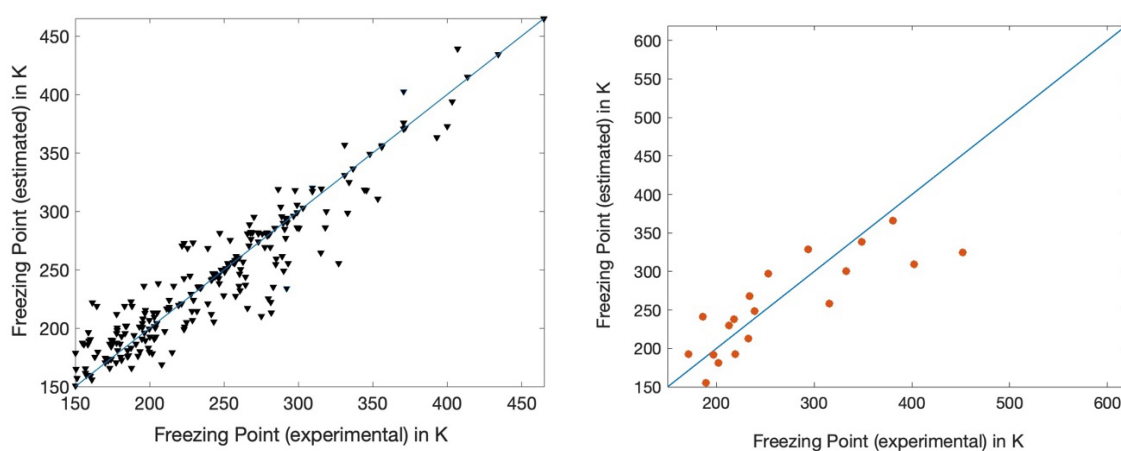


Figure 1: Predicted vs experimental freezing point a) using training data and b) testing data set

Table 1: First order group and their contribution

| No. | Group | T _{f1i} | No. | Group | T _{f1i} | No. | Group | T _{f1i} |
|-----|----------------------|------------------|-----|---------------------|------------------|-----|-------------------------------------|------------------|
| 1 | CH ₃ | -25.1779 | 28 | CH ₂ COO | 37.4507 | 55 | -Br | 13.0571 |
| 2 | CH ₂ | 6.4107 | 29 | CHCOO | 64.3918 | 56 | -F | -32.9191 |
| 3 | CH | 19.7042 | 30 | CCOO | 62.0679 | 57 | -Cl | -15.4623 |
| 4 | C | 111.7097 | 31 | HCOO | 9.4082 | 58 | OCH ₂ CH ₂ OH | 7.3985 |
| 5 | CH ₂ =CH | -37.1819 | 32 | aC-COO | 77.3272 | 59 | OCHCH ₂ OH | 49.5052 |
| 6 | CH=CH | 46.5267 | 33 | COO | 41.5902 | 60 | -O-OH | 29.4213 |
| 7 | CH ₂ =C | 10.0609 | 34 | CH ₃ O | -11.5888 | 61 | CH ₂ SH | 89.2957 |
| 8 | CH=C | 4.2740 | 35 | CH ₂ O | 16.1671 | 62 | -SH | -55.0482 |
| 9 | C=C | 0.0000 | 36 | CH-O | 65.4424 | 63 | SO | 152.4181 |
| 10 | CH ₂ =C=C | 20.6231 | 37 | C-O | 44.3680 | 64 | SO ₂ | 138.1115 |
| 11 | CH#C | 62.0679 | 38 | aC-O | -0.5076 | 65 | SO ₄ | 102.2294 |
| 12 | C#C | 92.0236 | 39 | aC-NH ₂ | 4.5858 | 66 | C ₂ H ₃ O | -24.9888 |
| 13 | aCH | 14.8347 | 40 | NH ₂ | 72.1454 | 67 | CH ₂ cyc | 2.5637 |
| 14 | aC* | -4.4161 | 41 | CH ₂ CN | 13.0242 | 68 | CHcyc | 44.3950 |
| 15 | aC* | 3.0983 | 42 | aC-CN | -2.2164 | 69 | Ccyc | 102.9660 |
| 16 | aC-CH ₃ | 8.1605 | 43 | aC-NO ₂ | 47.1468 | 70 | CH=CHcyc | 0.8458 |
| 17 | aC-CH ₂ | -7.0135 | 44 | NO ₂ | 80.7040 | 71 | CH=Ccyc | 16.2655 |
| 18 | aC-C | 129.9025 | 45 | ONO | 92.2338 | 72 | C=Ccyc | 33.5817 |
| 19 | OH | 18.3884 | 46 | ONO ₂ | 22.0485 | 73 | CH ₂ =Ccyc | -22.7888 |
| 20 | aC-OH | 67.8606 | 47 | CH ₂ Cl | 24.2134 | 74 | N | 114.7331 |
| 21 | COOH | 84.8248 | 48 | CHCl ₂ | 39.4513 | 75 | O | 27.3682 |
| 22 | aC-COOH | 10.5701 | 49 | CCl ₃ | 32.8257 | 76 | CO | 70.7432 |
| 23 | CH ₃ CO | 16.5265 | 50 | CHF ₂ | -2.6939 | 77 | S | 39.7681 |
| 24 | CH ₂ CO | 92.7778 | 51 | CF ₃ | -16.1656 | 78 | -O- | 57.1131 |
| 25 | aC-CO | 54.6865 | 52 | aC-Cl | -34.5119 | 79 | >CO | 13.7810 |
| 26 | CHO | 14.4579 | 53 | aC-Br | -20.1533 | 80 | SiO | 82.9783 |
| 27 | CH ₃ COO | 23.2407 | 54 | -I | 77.0727 | 81 | Si | 82.9783 |

* fused with aromatic ring; * fused with non-aromatic subring

Table 2: Second Order group and their contribution

| No. | Group | T _{f2i} | No. | Group | T _{f2i} |
|-----|---|------------------|-----|--|------------------|
| 1 | (CH ₃) ₂ CH | -3.9378 | 21 | CHm=CHn-Cl (m,n in 0..2) | -1.8693 |
| 2 | (CH ₃) ₃ C | 4.9434 | 22 | CHn=CHm-COO-CHp (m,n,p in 0..3) | 3.1759 |
| 3 | CH(CH ₃)CH(CH ₃) | 1.6353 | 23 | CHm=CHn-CHO (m,n in 0..2) | -23.5336 |
| 4 | CHn=CHm-CHp=CHK (k,m,n,p in 0..2) | -20.7850 | 24 | CHm=CHn-COOH (m,n in 0..2) | -5.7567 |
| 5 | CH ₃ -CHm=CHn (m,n in 0..2) | 11.0241 | 25 | aC-CHn-OH (n in 1..2) | 40.8054 |
| 6 | CH ₂ -CHm=CHn (m,n in 0..2) | 3.4341 | 26 | aC-C(CH ₃) ₃ | -2.7346 |
| 7 | CHCHO or CCHO | -0.6462 | 27 | (CHn=C)cyc-CH ₂ (n in 0..2) | -0.0995 |
| 8 | CH ₃ COCH ₂ | -0.5950 | 28 | CHcyc-CH ₃ | 16.7442 |
| 9 | CH ₃ COOH or CH ₃ COC | 48.2536 | 29 | CHcyc-CH ₂ | -22.4256 |
| 10 | CHCOOH or CCOOH | 4.9168 | 30 | CHcyc-CH | 3.1336 |
| 11 | CH ₃ COOCH or CH ₃ COOC | 1.3490 | 31 | CHcyc-OH | 3.5546 |
| 12 | CHOH | 2.6454 | 32 | CHcyc-O- | -22.8164 |
| 13 | COH | -18.2048 | 33 | Ccyc-CH ₃ | 27.5261 |
| 14 | CHm(OH)CHn(OH) (m,n in 0..2) | -3.0807 | 34 | Ccyc-OH | 27.5261 |
| 15 | CH ₃ -O-CHn-COOH (n in 1..2) | 65.5527 | 35 | AROMRINGs1s2 | 14.3212 |
| 16 | COO-CHn-CHm-OOC (n, m in 1..2) | -6.4909 | 36 | AROMRINGs1s3 | -14.9894 |
| 17 | NC-CHn-COO (n in 1..2) | -30.9616 | 37 | AROMRINGs1s4 | 3.3874 |
| 18 | COCHnCOO (n in 1..2) | 10.1911 | 38 | AROMRINGs1s2s3 | 1.7249 |
| 19 | CHm-O-CHn=CHp (m,n,p in 0..3) | 16.6820 | 39 | AROMRINGs1s2s4 | 30.8010 |
| 20 | CHm=CHn-F (m,n in 0..2) | -5.8217 | 40 | AROMRINGs1s3s5 | 18.5555 |

Table 3: Third Order group and their contribution

| No. | Group | T_{F3i} | No. | Group | T_{F3i} |
|-----|--|-----------|-----|-----------------|-----------|
| 1 | HOOC-(CH _n) _m -COOH (<i>m</i> >2, <i>n</i> in 0..2) | -31.4978 | 3 | AROM.FUSED[2] | 29.1004 |
| 2 | OH-(CH _n) _m -OH (<i>m</i> >2, <i>n</i> in 0..2) | -2.6847 | 4 | AROM.FUSED[2]s1 | -29.0480 |
| | | | 5 | N multiring | 0.0418 |

The goodness of fit for the linear model was calculated based on the coefficient of determination (R^2) and standard deviation (SD). The accuracy of the model can be evaluated from the average absolute error (AAE), the average relative error (ARE) as presented in Table 4. The first order groups regression resulting an R^2 value of 0.8538. The R^2 value was improved to 0.8782 when considering the second order, and slightly better at 0.8829 when considering the third orders. The model improved when considering the second order groups especially for aromatic and alicyclic compounds with only one ring and polyfunctional acyclic compounds. Large and complex compounds were considered in the third order groups, however, only five (5) third order groups are available from the training set, the prediction considering third order group give less significance on the predicted value. The AAE values indicate the deviation of the predicted value from the experimental value about 15.62 K with ARE 7.23 %. The result is compared with the study by Hukkerikar et al. (2012) for the prediction of normal melting point. This study gave a slightly better prediction compared to the proposed model using CI method which gave prediction with $R^2 = 0.7135$, AAE = 38.68 K, and ARE=12.32 %. However, their study utilised more data and improved their prediction using stepwise regression method. The accuracy of the model in this study can be further improved by incorporate more data for training of the model.

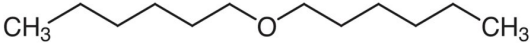
Table 4: The statistical analysis of the regression step

| Parameter | First Order | Second Order | Third Order |
|-----------|-------------|--------------|-------------|
| SD (K) | 24.7807 | 22.65 | 22.22 |
| AAE (K) | 17.78 | 16.00 | 15.62 |
| ARE (%) | 8.11 | 7.35 | 7.23 |
| R^2 | 0.8538 | 0.8782 | 0.8829 |

3.4 Model validation

The freezing point model was verified to prove the validity and accuracy of the model. This verification was done for 20 test data set using the contributions obtained from the first, second and third order contributions shown in Table 1 – 3. The result of prediction for 20 set data having values of AAE =33.88 K, ARE =12.22 % and R^2 is 0.8384. The test data prediction having higher AAE and ARE due to some compounds were having groups with less than 5 occurrences in the training set. To illustrate the prediction of freezing point, a sample of freezing point estimation for hexyl ether is illustrated in Table 5. Since hexyl ether is a simple and straight chain molecule, it can be described with only three first order groups. By using the model, the prediction given a value of 212.85 K compared to the experimental value of 230.15 K. The prediction result for hexyl ether has absolute error 17.3 K and a relative error of 7.51 % that indicates the percentage deviation from the actual value.

Table 5: Prediction of freezing point of hexyl ether

| Property | Value |
|------------------------------|--|
| Formula Molecule | C ₁₂ H ₂₆ O |
| T_F (experiment) | 230.15 K |
| Molecule Structure |  |
| Universal constant, T_{Fo} | 189.34 K |
| First-Order Group | Occurrences (N_i) Contribution (T_{Fi}) |
| CH ₃ | 2 -25.1779 |
| CH ₂ | 9 6.4107 |
| CH ₂ O | 1 16.1671 |
| Using Eq(1) | $T_F = T_{Fo} + \sum N_i T_{Fi} + w \sum M_j T_{F2j} + z \sum O_k T_{F3k}$ $T_F = 189.34 + 2(-25.1779) + 9(6.4107) + 16.1671$ $T_F = 212.85 \text{ K}$ $\text{Relative error} = 7.51 \%$ |

4. Conclusions

This study proposed a model that capable of determining the freezing point of jet fuel components. The outcome of this study was a model that can predict the freezing point of jet fuel component using group contribution method where different chemical structures described as first, second and third order groups. It is observed from the results an improvement in the correlation coefficient value from 0.8538 to 0.8829 of the first order group regression and third order group regression. The freezing point prediction model was verified and proven to be a good prediction model to estimate the freezing point of jet fuel and other liquid compounds with acceptable accuracy of the results and the correlation coefficient of 0.8384, AAE 33.88 K and ARE 12.22 %. The proposed group contribution model of freezing point is useful for determination of the freezing point of compounds within the listed groups as presented in Table 1 - 3. Future work can be conducted to include more experimental data of freezing point to increase the model accuracy, improve further AAE and ARE values, and also expand the model by including more groups. Therefore, the model can be applied for a wide range of compounds.

Acknowledgments

The author would like to thank Universiti Teknologi Malaysia (UTM) and Ministry of Higher Education Malaysia for the financial supports and fund this research through FRGS Grant (FRGS/1/2021/TK0/UTM/02/38).

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