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Method of Estimating Absolute Entropy of Municipal Solid Waste

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***Abstract*—**Entropy, as an outcome of the second law of thermodynamics, measures the level of irreversibility associated with any process. The identification and reduction of irreversibility in the energy conversion process helps to improve the efficiency of the system. The entropy of pure substances known as absolute entropy is determined at an absolute reference point and is useful in the thermodynamic analysis of chemical reactions; however, municipal solid waste (MSW) is a structurally complicated material with unknown absolute entropy. In this work, an empirical model to calculate the absolute entropy of MSW based on the content of carbon, hydrogen, oxygen, nitrogen, sulphur, and chlorine on a dry ash free basis (daf) is presented. The proposed model was derived from 117 relevant organic substances which represent the main constituents in MSW with known standard entropies using statistical analysis. The substances were divided into different waste fractions; namely, food, wood/paper, textiles/rubber and plastics waste and the standard entropies of each waste fraction and for the complete mixture were calculated. The correlation of the standard entropy of the complete waste mixture derived was found to be $\mathbf{s^o_{msw}}$= 0.0101C + 0.0630H + 0.0106O + 0.0108N + 0.0155S + 0.0084Cl (kJ.K^{-1}.kg) and the present correlation can be used for estimating the absolute entropy of MSW by using the elemental compositions of the fuel within the range of 10.3% ≤ C ≤ 95.1%, 0.0% ≤ H ≤ 14.3%, 0.0% ≤ O ≤ 71.1%, 0.0% ≤ N ≤ 66.7%, 0.0% ≤ S ≤ 42.1%, 0.0% ≤ Cl ≤ 89.7%. The model is also applicable for the efficient modelling of a combustion system in a waste-to-energy plant.

***Keywords*—**Absolute entropy, irreversibility, municipal solid waste, waste-to-energy.

I. Introduction

DISPOSAL of municipal solid waste (MSW) has become a major environmental challenge affecting people throughout the world following the growth in population and urbanization [1]. An appropriate MSW treatments offer practical solutions to control the environmental contaminations and climate change as well as sources of fuel for future energy needs [2]. The energy recovery from municipal waste can be operated in a cost-effective way with provision of useful energy with low carbon footprint to the society [3]. In order to maximize the waste utilization by converting it to useful energy in the energy conversion process, absolute entropy of waste is required. Entropy being an outcome of second law of thermodynamics combines with the first law for efficient improvement in the thermodynamics thermal systems.

Many solid fuels like coal, biomass and municipal solid waste have unknown structures and chemical compositions, their entropy values cannot be calculated directly because of the lack of standard absolute entropy values [4]. Only few methods for the prediction and estimation of the absolute entropy of carbon-based fuels with complex bond interactions and unknown thermodynamics properties have been proposed based on the characteristics of the known homogeneous organic substances in the fuel. Eisermann et al. [5] on estimating the thermodynamic properties of coal, char, tar and ash, approximated the standard entropy of coal by evaluating the behaviour of the standard entropies of a number of aliphatic and aromatic hydrocarbons as a function of several elemental ratios: H/(C+N), O/(C+N), N/(C+N), and S/(C+N). Shieh and Fan [6] developed a method for estimating the absolute entropy of a structurally complicated material by adopting the concepts of the dead (or reference) state and the properties of the constituents in the material based on the first and second laws of thermodynamics. They assumed that the entropy of a fuel is equal to the entropies of the elements forming it. Ikumi et al. [7] estimated the entropies of coals as well as that of coal liquids by using empirical expressions on the basis of statistical studies of corresponding mole ratios of hydrogen, oxygen, nitrogen and sulphur elements to the carbon element. To estimate the specific entropy of the organic matter in biomass, Song et al. [8] developed a model based on the Shieh and Fan [6] method. Also, Song et al. [9] extended the Shieh and Fan [6] model by using the major organic constituents of the solid fuel to estimate the entropy. However, the applications of these models are limited, as they are applicable to coal, biomass, or mixed solid fuels, and some are derived from the solid fuels containing elements C, H, O, N, and S only.

The aim of this work is to present a correlation for estimating the standard entropy of municipal solid waste that contains elements C, H, O, N, S, and Cl from its elemental compositions on a dry ash free basis.

II. Derivation Method

Municipal solid waste (MSW) contains mainly organic polymers like plastics, wood, paper, textile, rubber, and food waste. The entropies of these polymers in the organic waste are estimated or evaluated by the entropies of their organic monomers structures as there is no significant difference between the entropies of the solid organic monomers and their polymers [10].

The method for calculation of estimated standard entropy of

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municipal solid waste was done by the computation of homogenous or pure organic substances with known standard entropies in the waste material. Based on the absolute entropies and elemental compositions of the selected organic substances, a first-order polynomial correlation was derived statistically for the standard entropy estimate of the waste fractions and the mixture.

A. Collection, Selection and Classification of Relevant Data

In the present work, a large number of 117 organic compounds with their standard entropies were collected from published literature and presented in Table I. The data points were selected based on the molecular structures of the organic substances that are associated or linked with the formation of larger molecular structure network of municipal solid waste. The organic compounds were classified into the six categories of waste fractions; namely, food, plastic, textile and rubber, wood and paper. This was accomplished by considering the molecular structure of the organic compounds that can be found in each of the molecular structures in the waste fractions. For wood, it contains three major chemical components: cellulose, hemicellulose, and lignin [11]. Each of the chemical structure of the wood constituents [12], [13] was studied and organic compounds (monomers) that can be made or found from these structures were selected. In the food, the main structural elements identified are proteins, carbohydrate, and lipids [14]. The molecular structures of these food components [15], [16] were also investigated and organic monomers that were linked with the structure were selected. The same method was carried on the chemical structures of plastic [17], textile [18], [19], and rubber [20] materials with identifications of biologically important molecules which form the building structure of their polymers.

TABLE I
STANDARD ENTROPIES AT 298.15K OF ORGANIC COMPOUNDS RELEVANT TO MSW [5], [6], [8]-[10], [23]

Name	Formula	S^0(kJ/kg.K)
FOOD		
1. Allantoin	$C_4H_6N_4O_3$	1.233
2. Alloxan	$C_4H_2N_2O_4$	1.314
3. Arginine	$C_6H_{14}N_4O_2$	1.439
4. Asparagine	$C_4H_8N_2O_3$	1.322
5. Aspartic acid	$C_4H_7NO_4$	1.279
6. Citric acid	$C_6H_8O_7$	1.312
7. Creatine	$C_4H_9N_3O_2$	1.445
8. Cystine	$C_6H_{12}N_2O_4S_2$	1.347
9. D-Glutamic acid	$C_5H_9NO_4$	1.230
10. l-Lactic acid	$C_3H_6O_3$	1.579
11. L-Phenylalanine	$C_9H_{11}NO_2$	1.293
12. L-Proline	$C_5H_9NO_2$	1.425
13. Maleic acid	$C_4H_4O_4$	1.373
14. Malic acid	$C_4H_6O_5$	1.199
15. Methionine	$C_5H_{11}NO_2S$	1.552
16. Phenanthrene	$C_{14}H_{14}$	1.207
17. Trytophan	$C_{11}H_{12}N_2O_2$	1.229
18. Tyrosine	$C_9H_{11}NO_3$	1.181
19. Uric acid	$C_5H_4N_4O_3$	1.030
20. Valine	$C_5H_{11}NO_2$	1.527
21. Xanthine	$C_5H_4N_4O_2$	1.059
22. Stearic acid	$C_{18}H_{36}O_2$	1.531
23. Taurine	$C_2H_7NO_3S$	1.231
24. Urea	CH_4N_2O	1.742
25. Hexadecanoic acid	$C_{16}H_{32}O_2$	1.764
26. Adenine	$C_5H_5N_5$	1.118
27. Creatinine	$C_4H_7ON_3$	1.483
28. L-Serine	$C_3H_7O_3N$	1.419
29. L-Glutamine	$C_5H_{10}O_3N_2$	1.335
30. DL-Alanyl glycine	$C_5H_{10}O_3N_2$	1.460
31. Glycylglycine	$C_4H_8N_2O_3$	1.438
32. Alanine	$C_3H_7NO_2$	1.450
33. Cysteine	$C_3H_7NO_2S$	1.402
34. Dimethyl sulfone	$C_2H_6O_2S$	1.509
35. D-Lactic acid	$C_3H_6O_3$	1.593
36. Fumaric acid	$C_4H_4O_4$	1.447
37. Guanine	$C_5H_5N_5O$	1.061
38. Gycine	$C_2H_5NO_2$	1.379
39. Isoleucine	$C_6H_{13}NO_2$	1.586
40. Leucine	$C_6H_{13}NO_2$	1.586
41. L-Glutamic acid	$C_5H_9NO_4$	1.279
42. 1-Hexadecanol	$C_{16}H_{34}O$	1.864
43. Hypoxanthine	$C_5H_4ON_4$	1.070
44. Glycolide	$C_4H_4O_4$	1.354
PLASTIC		
45. 1,3,5-Trioxane	$C_3H_6O_3$	1.476
46. Benzophenone	$C_{13}H_{10}O$	1.346
47. Biphenyl	$C_{12}H_{10}$	1.358
48. Hexachloroethane	C_2Cl_6	1.002
49. Diphenyl carbonate	$C_{13}H_{10}O_3$	1.300
50. Diphenyl ether	$C_{12}H_{10}O$	1.372
51. Diphenylcarbinol	$C_{13}H_{12}O$	1.330
52. Polypropylene,isotatic	$(C_3H_6)n$	1.662
53. Polypropylene,syndiotic	$(C_3H_6)n$	1.798
54. Pyromellitic dianhydride	$C_{10}H_2O_6$	1.087
55. Naphthalene	$C_{10}H_8$	1.306
56. Succinic acidic	$C_4H_6O_4$	1.417
57. Cyanuric acid	$C_3H_3N_3O_3$	1.947
58. Acetamide	C_2H_5NO	1.840
59. Durene	$C_{10}H_{14}$	1.166
60. Hexamethlenetetramine	$C_6H_{12}N_4$	1.116
61. Triphenylene	$C_{18}H_{12}$	1.273
62. Hydroquinone	$C_6H_60_2$	1.182
63. Melamine	$C_3H_6N_6$	1.251
64. Phthalic acid	$C_8H_6O_4$	1.215
65. Phathalic anhydide	$C_8H_4O_3$	1.405
66. Triethylenediamine	$C_6H_{12}N_2$	1.947
67.4,4'-diphenylmethane diisocyanate	$C_{15}H_{10}N_2O_2$	1.329
68. Polyisocyanurate	$(C_{15}H_{10}N_2O_2)n$	1.175
69. Tridecanolactone	$C_{13}H_{24}O_2$	1.893
70. Polytridecanolactone	$(C_{13}H_{24}O_2)n$	1.656
71. Polyvinylidene chloride	$(C_2H_2Cl_2)n$	0.894
72. Polyvinyl chloride	$(C_2H_3Cl)n$	1.042
73. poly(1-butene), isotactic	$(C_4H_8)n$	1.836
74. Polystyrene	$(C_8H_8)n$	1.294
TEXTILE		
75. 3-Nitrobenzoic acid	$C_7H_5NO_4$	1.227
76. 1,2-Diphenylethene	$C_{14}H_{12}$	1.390
77. Adipic acid	$C_6H_{10}O_4$	1.504

Name	Formula	S^0(kJ/kg.K)
78. 2-Methlnaphthalene	$C_{11}H_{10}$	1.547
79. Acenaphthene	$C_{12}H_{10}$	1.225
80. Anthracene	$C_{14}H_{10}$	1.164
81. 1,4-Benzoquinone	$C_6H_4O_2$	1.506
82. Diphenylamine	$C_{12}H_{11}N$	1.666
83. Pyrene	$C_{16}H_{10}$	1.112
84. Thiourea	CH_4N_2S	1.523
85. Ammonium thiocyanate	CH_4N_2S	1.842
86. 3-Nitroaniline	$C_6H_6N_2O_2$	1.276
87. Resorcinol	$C_6H_6O_2$	1.341
88. Triphenylmethane	$C_{19}H_{16}$	1.277
89. Triphenylmethanol	$C_{19}H_{16}O$	1.265
90. Isoquinoline	C_9H_7N	1.324
91. Acridine	$C_{13}H_9N$	1.161
92. 2-Nitrobenzoic acid	$C_7H_5O_4N$	1.247
93. 1,3-Phenylenediamine	$C_6H_8N_2$	1.429
94. Dicyanodiamide	$C_2H_4N_4$	1.538
95. ε-Caprolactam	$C_6H_{11}NO$	1.531
96. Poly-ε-Caprolactam	$(C_6H_{11}NO)n$	1.529
97. Polyglycolide	$(C_4H_4O_4)n$	1.304
WOOD		
98. L-Sorbose	$C_6H_{12}O_6$	1.226
99. o-cresol	C_7H_8O	1.530
100. Oxalic acid	$C_2H_2O_4$	1.220
101. P-Cresol	C_7H_8O	1.547
102. Sucrose	$C_{12}H_{22}O_{11}$	1.052
103. D-Mannitol	$C_6H_{14}O_6$	1.309
104. Pentachlorophenol	C_6HCl_5O	0.946
105. Galactose	$C_6H_{12}O_6$	1.140
106. Phenol	C_6H_6O	1.530
107. 2-Hydroxybenzoic acid	$C_7H_6O_3$	1.290
108. Glucose	$C_6H_{12}O_6$	1.161
109. Xylose	$C_5H_{10}O_5$	0.956
110. 3-Hydroxybenzoic acid	$C_7H_6O_3$	1.281
111. 4-Hydroxybenzoic acid	$C_7H_6O_3$	1.272
112. Benzoic acid	$C_7H_6O_2$	1.372
113. Catechol	$C_6H_6O_2$	1.364
114. Lactose	$C_{12}H_{22}O_{11}$	1.128
115. O-Hydroxybenzoic acid	$C_7H_6O_3$	1.290
116. O-Hydroxybenzoic acid	$C_7H_6O_3$	1.281
117. O-Hydroxybenzoic acid	$C_7H_6O_3$	1.272

B. Selection of Suitable Methods of Correlations

Three assumed algebraic expressions for estimating entropies of solid fuels from the previous models and the present work based on their correlation with elemental compositions of the fuels were used, as shown in Table II. The constants terms of these algebraic expressions were calculated with statistical method by using regression analysis on the 117 data points of organic compound. The correlation that has the least error values and highest coefficient of determination, as described in Section II *C*, were selected.

C. Selection of the Best Correlation

For the selection of the best correlation in Table II, three statistical parameters were used as evaluating parameters for each of the three correlations, which were computed as follows:

$$AAE = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{S_{est}^{o}-S^{o}}{S^{o}}\right| \times 100\% \quad (1)$$

$$ABE = \frac{1}{n}\sum_{i=1}^{n}\frac{S_{est}^{o}-S^{o}}{S^{o}} \times 100\% \quad (2)$$

$$R^2 = 1 - \sum_{i=1}^{n}\frac{(S_{est}^{o}-S^{o})^2}{(S^{o}-\bar{S}^{o})^2} \quad (3)$$

where S_{est} and S^o denote the estimated and absolute entropy values, respectively. $\bar{S}^o$ is the average absolute entropy value. AAE is the average absolute error of a correlation. A smaller error of correlation will occur when AAE is low, which indicates higher accuracy. ABE denotes the average bias error of correlation. A positive value of ABE indicates an overall overestimation, while a negative value implies an overall underestimation. The smaller the absolute value of ABE, the smaller the bias of correlation. R^2 denotes coefficient of determination. R^2 is used as a comprehensive parameter to measure the accuracy of the model. A higher R^2 value means a better estimation and fitting [21]. The above three parameters are the important statistical criteria and are primarily employed to assess correlations [8], [9], [22]. Hence, they are used here as evaluating parameters. Table II showed the values of the three parameters for the three correlations.

D. Comparative Study with Published Models

The validation of the proposed correlation is carried out by comparing it with other published correlations of solid fuels with their standard entropies. In each model, absolute average error (AAE), average bias error (ABE) and coefficient of determination (R^2) are calculated to evaluate the correlations. The detail results are presented in Table III.

TABLE II
CORRELATIONS FOR THE SELECTION OF THE PROPOSED MODEL FOR STANDARD ENTROPY, S^o

No.	empirical expression	Criteria for selection	AAE (%)	R^2	ABE (%)	Reference
1	$S^o = a_1C + a_2H + a_3O + a_4N + a_5S$	Assuming S^0 to be a linear function of it constituents.	9.970	0.44	-0.385	[8], [9]
2	$S^o = a_1C + a_2H + a_3O + a_4N + a_5S + a_6Cl$	Assuming S^0 to be a linear function of it constituents including Cl	8.293	0.54	1.118	Current model
3	$S^o = a_1 + a_2(H/C) + a_3(O/C) + a_4(N/C) + a_5(S/C)$	Assuming S^0 to be function of several elemental ratios: (H/C), (O/C), (N/C), (S/C)	9.557	0.40	1.507	[7]

where C,H,O,N,S and Cl represents carbon, hydrogen, oxygen, nitrogen, sulphur and chlorine, respectively, in % by mass on a dry ash free basis. a_1, a_2, a_3, a_4, a_5 and a_6 are constants of correlation.

TABLE III
PROPOSED CORRELATION COMPARED WITH PREVIOUS MODELS

No.	Correlation (MJ/kg)	Unit	Application	AAE (%)	ABE (%)	R^2	Ref.
1	S^o = 0.0101C + 0.0630H + 0.0106O + 0.0108N + 0.0155S + 0.0084Cl	kJ/kgK	MSW	8.293	1.118	0.54	Current model
2	S^o= 0.0086C + 0.0780H + 0.0106O + 0.0103N + 0.0118S	kJ/kgK	Mixed waste	10.412	-1.437	0.46	[9]
3	S^o = 0.0055C + 0.095H + 0.0096O + 0.0098N + 0.0138S	kJ/kgK	Biomass	13.481	-8.527	0.49	[8]
4	S^o = 5.69 + 13.12(H/C) + 14.19(O/C) + 21.45(N/C) + 0.0138S	J/mol.carbon.K	Coal	35.433	-31.077	0.001	[7]
5	$S^o = a_1 + a_2 \exp(-a_3 (H/C+N)) + a_4 (O/C+N) + a_5 (N/C+N) + a_6 S/(C+N)$	kJ/kmol.carbon.K	Coal	31.034	-15.030	6* 10^-4	[6]

where a_1 = 37.1653 , a_2 = -31.4767, a_3 = 0.564682, a_4 = 20.1145, a_5 = 54.311, a_6 = 44.6712

III. RESULTS AND DISCUSSION

In Table II, the algebraic expression of the current model was selected as a suitable form of correlation for estimating absolute entropy of municipal solid waste. This was the correlation that had the least error i.e. absolute average error (AAE) and highest coefficient of determination (R^2) with higher accuracy among the three assumed expressions. By using this expression with regression analysis for correlation of standard entropy of waste fractions and the mixture as well as their elemental components, the five correlations for estimating the standard entropy of waste fractions and the mixture of waste were derived and expressed as follows:

- For Plastic waste;

$$s^o_{pl} = 0.0087C + 0.0753H + 0.0134O + 0.0077N + 0.084Cl \quad (kJ.K^{-1}.kg) \quad (4)$$

$10.3\% \leq C \leq 94.8\%$, $0.00\% \leq H \leq 14.3\%$,

$0.0\% \leq O \leq 54.2\%$, $0.00\% \leq N \leq 66.7\%$.

with ABE, AAE and R^2 of 0.722, 7.314 and 0.7674, respectively.

- For Textile/Rubber waste;

$$s^o_{tr} = 0.0097C + 0.0635H + 0.0128O + 0.0136N + 0.0165S \quad (kJ.K^{-1}.kg) \quad (5)$$

$15.8\% \leq C \leq 95.1\%$, $3.0\% \leq H \leq 9.7\%$,

$0.0\% \leq O \leq 55.2\%$, $0.0\% \leq N \leq 66.7\%$,

$0.0\% \leq S \leq 42.1\%$

with ABE, AAE and R^2 of 0.714, 6.476 and 0.5457, respectively.

- For Wood/Paper waste;

$$s^o_{wp} = 0.0162C + 0.0116H + 0.0081O + 0.00691Cl \ (kJ.K^{-1}.kg) \quad (6)$$

$26.7\% \leq C \leq 77.8\%$, $0.4\% \leq H \leq 7.7\%$,

$5.1\% \leq O \leq 71.1\%$, $0.0\% \leq Cl \leq 66.3\%$

with ABE, AAE and R^2 of 0.329, 5.215 and 0.728, respectively.

- For Food waste;

$$s^o_{fo} = 0.0065C + 0.0808H + 0.0127O + 0.0101N + 0.0100S \quad (kJ.K^{-1}.kg) \quad (7)$$

$19.2\% \leq C \leq 92.3\%$, $1.4\% \leq H \leq 14.1\%$,

$0.0\% \leq O \leq 59.7\%$, $0.0\% \leq N \leq 51.9\%$,

$0.0\% \leq S \leq 34.0\%$

with ABE, AAE and R^2 of 0.414, 5.886 and 0.6922, respectively.

- For mixed waste;

$$s^o_{msw} = 0.0101C + 0.0630H + 0.0106O + 0.0108N + 0.0155S + 0.0084Cl \ (kJ.K^{-1}.kg) \quad (8)$$

$10.3\% \leq C \leq 95.1\%$, $0.0\% \leq H \leq 14.3\%$,

$0.0\% \leq O \leq 71.1\%$, $0.0\% \leq N \leq 66.7\%$,

$0.0\% \leq S \leq 42.1\%$, $0.0\% \leq Cl \leq 89.7\%$,

with ABE, AAE and R^2 of 1.118, 8.293, and 0.5414, respectively.

Comparing the five equations obtained, the results show that the standard entropy correlations for each waste fraction in MSW are more accurate than the standard entropy correlation for the waste mixture. Indicating that municipal solid waste being a heterogeneous mixture with complicated structures has lesser amount of disorder in their molecular structure than each of the waste fraction. It also indicates that the boundary systems of waste mixture are stronger and have a lower irreversibility within the system than the waste fractions. Though, the estimated standard entropy of waste mixture did not show best prediction with the experimental data (absolute entropies); however, with correlation coefficient (R) of 0.736 it can be acceptable when compared with past models.

The validity of the current model is compared with widely used correlations as presented in Table III and represented in Figs. 1-5. The results obtained demonstrate that the proposed correlation shows significantly better estimations and accuracy

when considering the errors (AAE and ABE) and coefficient of determination (R^2) compared to the other correlations. It is found that other correlations [6]-[9] underestimated the standard entropy values with higher errors and least coefficient of determination (R^2), indicating that their models are less accurate when applied in municipal solid waste.

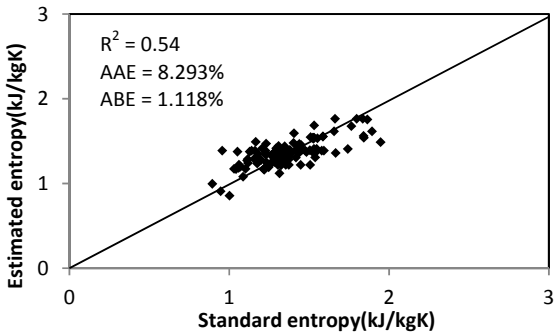

Fig. 1 Comparison between the standard entropy and estimated entropy proposed in this work

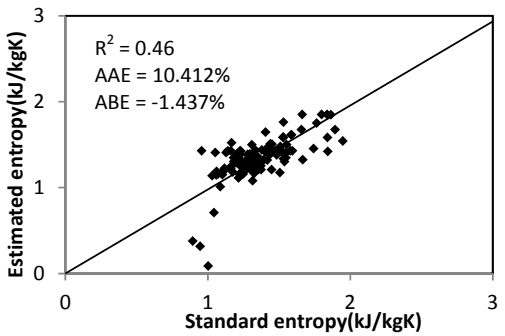

Fig. 2 Comparison between the standard entropy and estimated entropy by Song et al. [9]

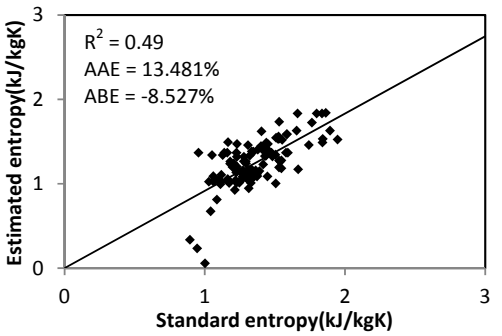

Fig. 3 Comparison between the standard entropy and estimated entropy by Song et al. [8]

IV. Conclusions

In this work, a simple method for estimating the standard entropy of municipal solid waste on (daf) from their ultimate analysis was proposed. All other methods have either used other solid fuels like coal and biomass or have ignored the inclusion of chlorine from the elemental compositions of waste in their derivations. The model developed is better than previous methods, even though there are still rooms for improvement. It is expressed as

$$s^{o}_{msw} = 0.0101C + 0.0630H + 0.0106O + 0.0108N + 0.0155S + 0.0084Cl \ (kJ.K^{-1}.kg).$$

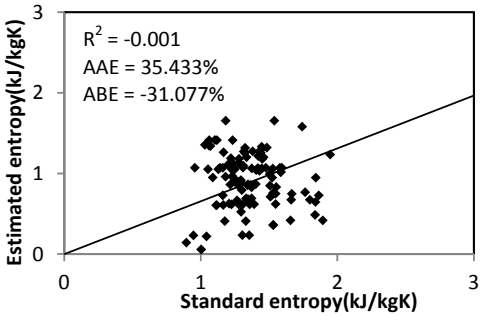

Fig. 4 Comparison between the standard entropy and estimated entropy by Ikumi et al. [7]

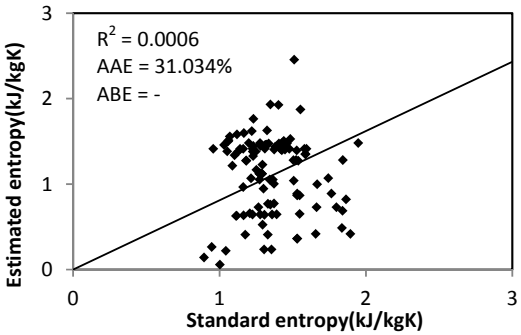

Fig. 5 Comparison between the standard entropy and estimated entropy by Eisermann et al. [5]

Due to the complicated mixture, heterogeneous molecule structure and variation of municipal solid waste chemical composition and properties, the accuracy of the new correlation developed is not high, however the current proposed model has highest accuracy when compared to its counterparts and can be used conveniently in engineering applications for estimating absolute entropy of municipal solid waste. The model is also useful for analysis and syntheses of energy resources conversion processes based on the second law of thermodynamics. The procedures are simple, robust, and reliable and can be used by both specialists and non-specialists.

Nomenclature

AAE	= average absolute error
ABE	= average bias error
MSW	= municipal solid waste
s	= specific entropy (kJ/kg.K)
S	= entropy (kJ/K)
R^2	= coefficient of determination

Subscripts

tr = textile/rubber
pl = plastic
wp = wood/paper
fo = food
est = estimate
msw = municipal solid waste
daf = dry ash free basis

Superscripts

o = reference state

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