

On the Homographic Dependence of Activation Energy and Viscosity Arrhenius' Temperature for Some Pure Fluids

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Viscosity is the most important hydraulic fluid characteristic and it is one of the significant quantities which are affected by pressure and temperature. Using of statistical methods for regression and correlation analysis, any eventual causal association between the melting and boiling points and the Arrhenius-type equation parameters of some classical Newtonian fluids is attempted. Empirical validations using 101 data set of viscosity of pure Newtonian fluids studied at atmospheric pressure and at different temperature domains give reliable statistical result. In fact, we found a significant strong causal correlation between the Arrhenius activation energy (E_a), the boiling point (T_b) and the Arrhenius temperature (T_A). As a result, an original empirical model modeling this relationship is suggested. The proposed model allows the prediction of the normal boiling temperature through information on viscosity Arrhenius parameters. Moreover, the proposed model is very beneficial for fluid engineering data especially for the study of systems efficiency and hydraulic components. Because of that, the need for more accurately and specific mathematical modeling of the fluid behaviour is required.

Keywords: Viscosity, Arrhenius temperature, Boiling point, Pure fluid.

INTRODUCTION

Recently we have initiated a collaborating research axis on application of thermodynamic/transport properties of some engineering fluid systems in suggested correlation equations [1-11]. Considering the experimental values in the literature, here it is extended to investigate the viscosity Arrhenius behaviour along with a new parameter applied as Arrhenius temperature (T_A), in some classical fluids at different temperatures.

In addition, we add that correlation between Arrhenius parameters reveals interesting Arrhenius temperature which is closely related to the vaporization temperature in the liquid vapour equilibrium and the limiting corresponding partial molar properties can permit us to estimate the boiling points of the pure components. In conclusion, we can ascertain that with more mathematical handlings, we will be able to reveal some physical significances of the viscosity Arrhenius parameters and it definitely develops as well as improves the

thermodynamic theories and also to predict some information on liquid-vapour diagram through the study of the viscosity *versus* temperature and molar fraction only in the liquid phase of binary mixture.

The additional study on the eventual relationship between the Arrhenius temperature and the properties of great number binary mixtures can prove how the method predicts the properties of other non-treated fluid mixtures. This will be very fruitful for hydraulic fluid quantities simulation in the optimization and design of various industrial processes, such as in chemical industry, food industry, hydraulic-mechanics cosmetics and pharmaceuticals, *etc.*

Temperature dependence of fluid viscosity

Numerous empirical expressions have been suggested in the literature for representing the fluid viscosity (η) *versus* temperature (T) through available experimental data for interpolation purpose. Generally, most of these proposed simplified mathematical and practical models use at least two parameters.

The simplest form of representation of Newtonian fluid viscosity *versus* temperature is a relationship with two parameters proposed [12-15] and known as the Andrade equation:

$$\eta = Ae^{\frac{B}{T}} \quad (1)$$

where A and B are positive constants and are characteristics of each pure liquid. Based on the Andrade equation, many researchers have suggested (quite similar) expressions. However, the most popular is the so called Arrhenius type-equation which may be linearly expressed in the natural logarithm form as follows:

$$\ln \eta = \ln A_s + \frac{E_a}{R} \left(\frac{1}{T} \right) \quad (2)$$

where R, E_a and A_s are the ideal gas constant, the Arrhenius activation energy and the pre-exponential factor of the Arrhenius equation for the pure liquid system respectively.

In this work, we focus on the Arrhenius type-equation for studying any eventual relationship between its parameters and

the melting and boiling points of some classical Newtonian liquids. If this relationship exists, it may allow to suggest new equation allowing the prediction of the boiling temperature through information on viscosity Arrhenius parameters.

Correlation analysis

Arrhenius and temperature parameters: A sample of 101 experimental data provided from the literature [15-52] on viscosity of pure Newtonian fluids (Table-1) studied at different temperature ranges and at atmospheric pressure is used in this work in order to analyze the existence of any eventual causal correlation between the Arrhenius type-equation, as knowing the activation energy (E_a) and the entropic factor (ln A_s) and eventually the melting temperature (T_m) and boiling temperature (T_b). Also, the present investigation includes additional parameters such as the Arrhenius activation temperature (T^{*} = E_a/R) and the Arrhenius temperature (T_A = -E_a/R ln A_s) as defined in previous works [1-11].

The first statistical investigation (Table-2) of the mean of the above-mentioned temperature parameters gives the following classification:

TABLE-1
ARRHENIUS PARAMETERS OF SOME PURE LIQUIDS STUDIED LITERATURE, ARRHENIUS ACTIVATION ENERGY (E_a) (kJ mol⁻¹),
THE LOGARITHM OF THE ENTROPIC FACTOR OF ARRHENIUS (ln A_s) (Pa·s), THE ARRHENIUS TEMPERATURE (T_A) (K),
THE BOILING POINT (T_b) (K) AND CALCULATED VALUES FROM THE eqn. 4

#	Pure component	T _b (K)	E _a (kJ mol ⁻¹)	ln A _s	T _A (K)	E _{a calc.} (kJ mol ⁻¹)	Ref.
1	n-Heptane	371.15	8.6196	-11.302	91.723	8.5536	[15]
2	n-Pentane	309.25	6.0998	-10.886	67.393	6.0669	[15]
3	Butyl alcohol	390.85	19.114	-13.689	167.94	20.632	[15]
4	Aniline	457.28	19.997	-13.564	177.32	20.293	[15]
6	Ethyl acetate	350.15	9.9183	-11.728	101.72	10.057	[15]
7	Benzene	353.15	10.940	-11.812	111.39	11.408	[15]
8	Diethyl ether	307.75	7.5203	-11.446	79.021	7.4716	[15]
9	Acetic acid	391.15	11.213	-11.308	119.26	12.027	[15]
10	Acetone	329.20	7.4406	-11.097	80.643	7.5055	[15]
11	Toluene	383.75	9.0229	-11.135	97.461	9.1682	[15]
12	m-Xylene	412.25	8.7485	-10.975	95.872	8.7694	[15]
13	Carbone tetrachloride	349.87	10.329	-11.152	111.39	11.457	[15]
14	Chlorobenzene	405.15	8.7094	-10.695	97.939	9.0651	[15]
15	ethylbenzene	409.15	9.1110	-11.027	99.375	9.2111	[15]
16	<i>o</i> -Xylene	417.15	9.8360	-11.145	106.14	9.9866	[15]
17	Water	373.15	15.835	-13.414	141.98	16.057	[16]
18	Benzene	353.15	5.5473	-9.7233	68.618	5.9968	[17]
19	Chlorobenzene	405.15	8.2327	-10.452	94.732	8.6799	[17]
20	Water	373.15	12.043	-11.900	121.71	12.660	[18]
21	Benzyl alcohol	478.45	26.777	-16.486	195.34	23.135	[19]
22	Ethylamine	289.75	20.025	-15.048	160.05	25.063	[19]
23	<i>n</i> -Octanol	468.15	24.955	-14.945	200.83	24.652	[20]
24	Propylene glycol	461.35	47.765	-22.128	259.62	41.731	[21]
25	Butane-1,4-diol	503.15	33.359	-16.210	247.51	34.193	[22]
26	Butane-1,2-diol	465.15	43.910	-20.681	255.36	39.780	[22]
27	TEGMBE*	545.15	22.963	-14.119	195.60	21.375	[23]
28	1,4-Butanediol	503.15	33.904	-16.438	248.07	34.346	[24]
29	1,4-Butanediol	503.15	34.033	-16.485	248.31	34.412	[24]
30	1,2-Butanediol	465.15	45.933	-21.510	256.84	40.299	[24]
31	1,2-Butanediol	465.15	46.763	-21.857	257.32	40.469	[24]
32	Propylene glycol	461.35	37.551	-18.266	247.25	37.417	[24]
33	Tetrahydrofuran	339.15	6.9072	-10.474	79.310	7.2761	[25]
34	1-Butanol	390.85	19.742	-13.925	170.52	21.195	[25]
35	2-Butanol	372.15	24.852	-15.860	188.46	26.769	[25]
36	1-Chlorobutane	351.65	7.2626	-10.699	81.639	7.4718	[25]
37	N,N-Dimethylformamide	425.00	9.0530	-10.780	101.00	9.2970	[26]
38	Formamide	483.15	16.410	-12.442	158.63	16.546	[26]

39	N,N-Dimethylacetamide	438.55	9.7590	-10.914	107.54	9.9946	[26]
40	2-Methoxyethanol	397.65	15.185	-12.602	144.93	15.977	[27]
41	Water	373.15	15.510	-13.284	140.42	15.774	[27]
42	N,N-Dimethylacetamide	438.55	9.4260	-10.896	104.05	9.5714	[27]
43	2-Ethoxyethanol	408.15	15.803	-12.682	149.87	16.592	[28]
44	N,N-Dimethylacetamide	438.55	9.7973	-10.934	107.77	10.023	[28]
45	1,4-Dioxane	374.15	12.660	-11.853	128.47	13.710	[29]
46	Water	373.15	15.920	-13.443	142.43	16.139	[29]
47	Isobutyric acid	426.65	11.126	-11.200	119.48	11.634	[30]
48	Water	373.15	15.749	-13.383	141.54	15.976	[30]
49	Ethanol	351.15	13.204	-12.166	130.50	14.552	[31]
50	Water	373.15	15.433	-13.232	140.28	15.749	[31]
51	Methanol	337.75	9.9340	-11.528	103.64	10.486	[32]
52	Water	373.15	15.640	-13.334	141.07	15.891	[32]
53	Methyl benzoate	475.75	15.142	-12.404	146.82	14.879	[33]
54	Cyclohexane	353.89	14.461	-12.831	135.56	15.395	[33]
55	<i>n</i> -Hexane	341.88	9.1501	-11.798	93.281	9.0037	[33]
56	Heptane	371.15	11.292	-12.367	98.023	9.3471	[33]
57	Octane	398.75	13.275	-12.886	123.90	12.599	[33]
58	<i>p</i> -Xylene	411.15	8.3920	-10.761	93.800	8.5322	[34]
59	Dimethyl sulfoxide	462.15	14.058	-11.872	142.42	14.424	[34]
60	<i>o</i> -Xylene	417.15	9.5730	-11.044	104.26	9.7519	[35]
61	Dimethyl sulfoxide	462.15	14.333	-12.002	143.63	14.602	[35]
62	Ethylene glycol	470.15	29.941	-16.146	223.03	29.762	[36]
63	1,4-Dioxane	374.15	11.669	-11.430	122.78	12.808	[36]
64	Water	373.15	16.684	-13.742	146.02	16.807	[36]
65	Triethyl amine	361.95	8.2120	-11.248	87.810	8.1423	[37]
66	Water	373.15	15.786	-13.389	141.80	16.024	[38]
67	Glycerol	455.15	59.608	-24.143	296.95	60.270	[39]
68	TEGMME**	395.15	21.245	-13.638	187.36	24.975	[40]
69	<i>n</i> -Heptane	371.15	14.344	-12.613	136.78	15.176	[41]
70	Propargyl alcohol	387.65	15.105	-12.607	144.10	16.069	[42]
71	Allyl alcohol	370.15	15.305	-12.866	143.07	16.339	[42]
72	<i>t</i> -Butanol	355.55	32.029	-18.476	208.50	35.392	[42]
73	2-Propanol	355.15	21.950	-15.032	175.63	24.353	[42]
74	1-Propanol	370.15	17.792	-13.415	159.51	19.639	[42]
75	Methanol	337.75	10.298	-11.654	106.27	10.873	[43]
76	Ethanol	351.15	12.309	-11.830	125.14	13.625	[43]
77	<i>n</i> -Propanol	370.15	17.786	-13.407	159.56	19.649	[43]
78	Pyridine	388.55	14.925	-12.635	142.08	15.693	[43]
79	Bromobenzene	429.15	16.771	-13.450	149.98	16.153	[44]
80	Chlorobenzene	405.15	7.9331	-10.406	91.687	8.3215	[44]
81	Ethylbenzene	409.15	8.5016	-10.807	94.617	8.6406	[44]
82	Benzene	353.15	14.955	-13.254	135.71	15.443	[44]
83	Dimethylsulfoxide	462.15	11.721	-10.975	128.44	12.467	[44]
84	3-Amino-1-propanol	458.65	36.059	-18.036	240.46	35.486	[45]
85	Isoamyl alcohol	403.15	21.640	-14.322	181.72	23.187	[45]
86	2-Propanol	355.15	25.430	-16.403	186.46	27.526	[45]
87	Ethanol	351.15	15.680	-12.997	145.10	17.324	[45]
88	1,4-Dioxane	374.15	12.074	-11.607	125.11	13.173	[45]
89	<i>n</i> -C ₁₀ H ₂₂	447.25	10.633	-11.355	112.63	10.558	[46]
90	<i>n</i> -C ₂₀ H ₄₂	616.25	18.630	-12.675	176.78	17.367	[46]
91	<i>n</i> -C ₂₂ H ₄₆	642.15	18.750	-12.469	180.86	17.639	[46]
92	<i>n</i> -C ₂₄ H ₅₀	664.55	18.962	-12.253	186.13	18.113	[46]
93	N-Methylacetamide	478.15	19.128	-13.155	174.88	19.318	[47]
94	2-Methoxyethanol	397.65	14.504	-12.662	137.77	14.770	[47]
95	Water	373.15	15.568	-13.307	140.70	15.824	[47]
96	Propylene carbonate	513.15	14.192	-11.729	145.53	14.234	[48]
97	1,2-Diethoxyethane	394.15	7.5690	-10.819	84.142	7.5176	[49]
98	Acetonitrile	354.65	6.9895	-10.793	77.885	7.0171	[50]
99	2-Methoxyethanol	397.65	15.704	-12.822	147.31	16.393	[51]
100	Tetrahydrofuran	339.15	6.7382	-10.393	77.977	7.1186	[52]
101	Methanol	337.75	10.198	-11.629	105.47	10.755	[52]

*TEGMBE = triethylene glycol monobutyl ether; ** TEGMME = triethylene glycol monomethyl ether

$$\bar{T}_A < \bar{T}_m < \bar{T}_b < \bar{T}^*$$

where there is no clear intersection between any consecutive (CI), Fig. 1 confirms this observation. Moreover, according to the coefficient of variation (CV), the Arrhenius activation temperature (T^*) is the most dispersed variable, inversely to the boiling temperature (T_b) which is the most homogenous.

Correlation analysis: Firstly, we studied direct mutual correlation between the Arrhenius parameters ($\ln A_s E_a$ and T_A). Fig. 2 shows that the pair-wise scatter plots are a suitable

TABLE-2
DESCRIPTIVE STATISTICS OF TEMPERATURES
PARAMETERS: ARITHMETIC MEAN (\bar{T}_i),
CONFIDENCE INTERVAL (CI), STANDARD ERROR
(SE) AND COEFFICIENT OF VARIATION (CV)

Parameters	\bar{T}_i (K)	CI (mean)	SE	CV (%)
T_A	143.80	131.17 – 156.44	5.029	36
T_m	227.16	217.32 – 240.69	4.065	23
T_b	406.68	390.31 – 423.05	6.514	16
T^*	2012.88	1707.90 – 2317.90	122.000	61

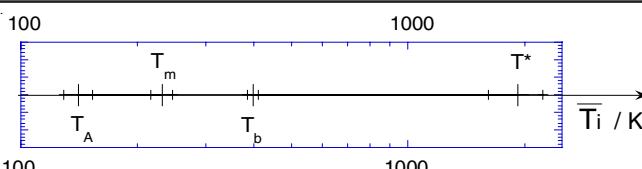


Fig. 1. Classification means of different temperatures (\bar{T}_i) used in the present statistical investigation. Great vertical bar (l): the average; small vertical bar (l): delimitation of (CI)

graphical method to deduct any eventual correlation. Based on the graphs, we deduced that there is clear relationship between some of the pair-wise analysis. In fact, comparison between the scatter plots (a,c) and (b,d) of Fig. 2, we observe that the T_A -dependence of (E_a) is “stronger” than of ($\ln A_s$). In addition, comparison between the scatter plots (b) and (d) of Fig. 2, it is concluded that the ($1/T_A$)-dependence of (E_a) is more “net” than the T_A -dependence of (E_a). So, in the light of these observations and especially the probable existence of vertical and horizontal asymptotes in Fig. 2d, we tested a suggested homographic dependence linking $Y = E_a$ with $X = (1/T_A)$ expressed as follows:

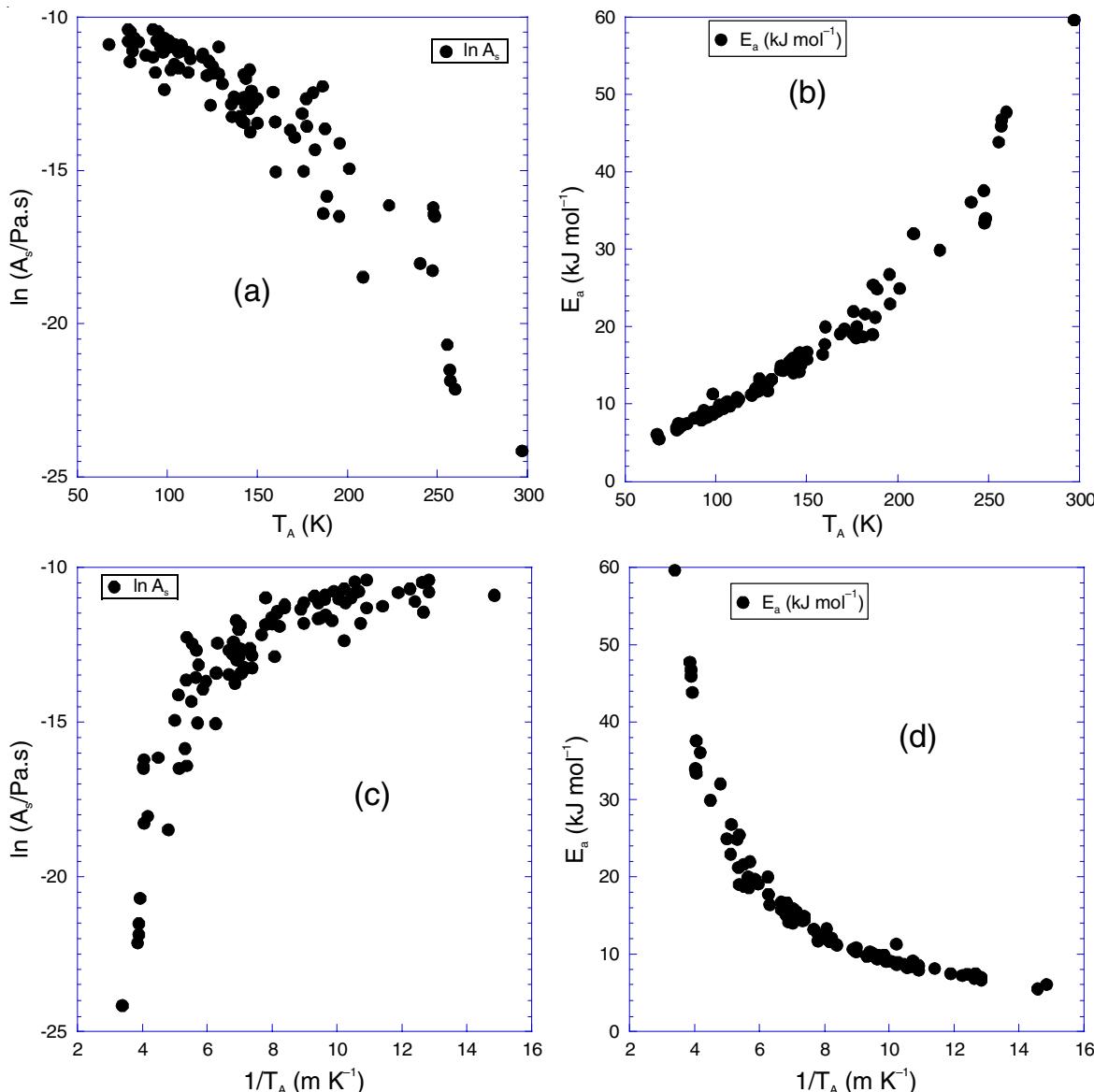


Fig. 2. Scatter plots of the direct mutual correlation between the Arrhenius parameters ($\ln A_s E_a$ and T_A) of some pure solvents

$$Y = \frac{AX + B}{CD + D} \quad (3)$$

To consider about the dependence of the viscosity with some physico-chemical properties, we have tested the boiling temperature (T_b) which is already predicted through the temperature-dependence presented in previous works [1-11]. For that, in order to measure up and assess eventual relationship between the target parameters, we have carried out the Spearman's rank correlations tests [53], where the null hypothesis assumes the independence of the variables. Table-3 presents the result of the test for three pairs.

TABLE-3
SPEARMAN RANK CORRELATION TESTS

Parameters	Spearman rho	Prob > t
$\ln A_s$	T_b	-0.24
E_a	T_b	0.40
T_A	T_b	0.47

Nevertheless, we think that an implicit association probably exists. For that purpose, we have tried various possible relationships between two or more transformed parameters. In first step, we have found interesting relations for which Fig. 3 illustrates the two best correlations found in this primary investigation. Indeed, a strong nonlinear correlation exists between the viscosity activation energy E_a and the boiling temperature (T_b).

Thus, using non-linear regression method to determinate the expression which best fit this correlation, we propose the homographic dependence linking $Y = E_a$ and $X = (1/T_A - 1/T_b)$ by the eqn. 3.

As a numerical application for the studied set of 101 solvents we suggest the eqn. 4 as a practical expression with correlation coefficient $R = 0.9935$, standard deviation $SD = 1.26$ and standard error (SE) = 0.16.

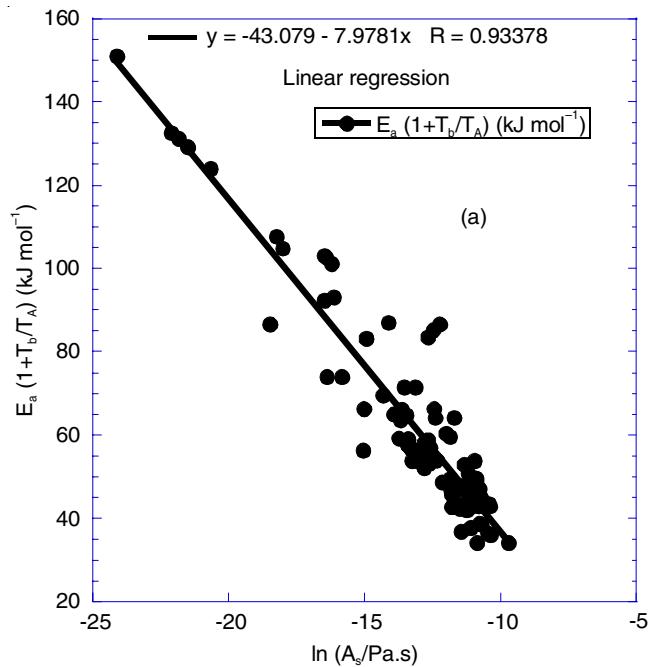


Fig. 3. Correlations between arrhenius parameters (E_a) or ($\ln A_s$) and some transformed variables for some pure classical solvents [Ref. 15-52]

$$E_a = \frac{0.07 \times \left(\frac{1}{T_A} - \frac{1}{T_b} \right) + 69.5}{\left(\frac{1}{T_A} - \frac{1}{T_b} \right) - 0.016} \quad (4)$$

where E_a is in kJ mol^{-1} and T_A and T_b in mK.

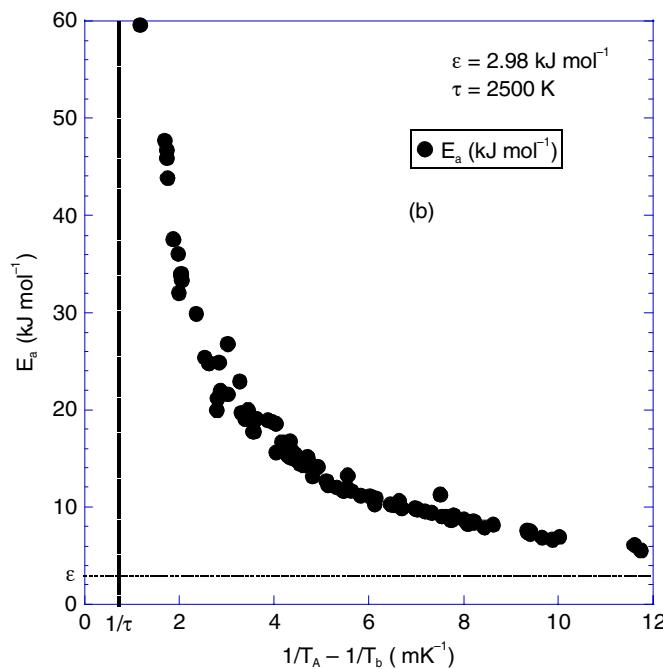
The calculated values of activation energies ($E_{a,\text{cal}}$) are presented in Table-2 and comparison between the experimental values ($E_{a,\text{exp}}$) are depicted by Fig. 4a,b. Nevertheless, the increase of absolute deviation of activation energy with the decrease of $(1/T_A - 1/T_b)$ is not an inconvenient that because of the increase of the activation energy in the corresponding region (Fig. 3b).

Regarding the curvature shape of the scatter plots in Fig. 3b, we deduced interesting result. A strong hyperbolic correlation (eqns. 3 and 4) may exist between E_a and $(1/T_A - 1/T_b)$. For that, the following expression (eqn. 5) linking E_a with $(1/T_A - 1/T_b)$ may hold well and releases some physical significance or equation's parameters:

$$E_a = \frac{R \cdot \omega}{\left(\frac{1}{T_A} - \frac{1}{T_b} \right) - \frac{1}{\tau}} + \varepsilon_0 \quad (5)$$

where ω is dimensionless constant, R is the ideal gas constant, τ is equivalent to an absolute temperature statistically related to the minimal values that the difference between the reciprocal of Arrhenius and boiling temperatures can theoretically taken for the set of treated solvents' group and ε_0 is equivalent to the minimal energy value that the Arrhenius activation energy can theoretically taken for the set of studied of viscous fluids.

According to the Table-4, which presents the result of statistical estimation of the proposed model, the estimated parameters are statistically significant, the R -squared is near to one and the value of Fisher statistics is high [54]. The present



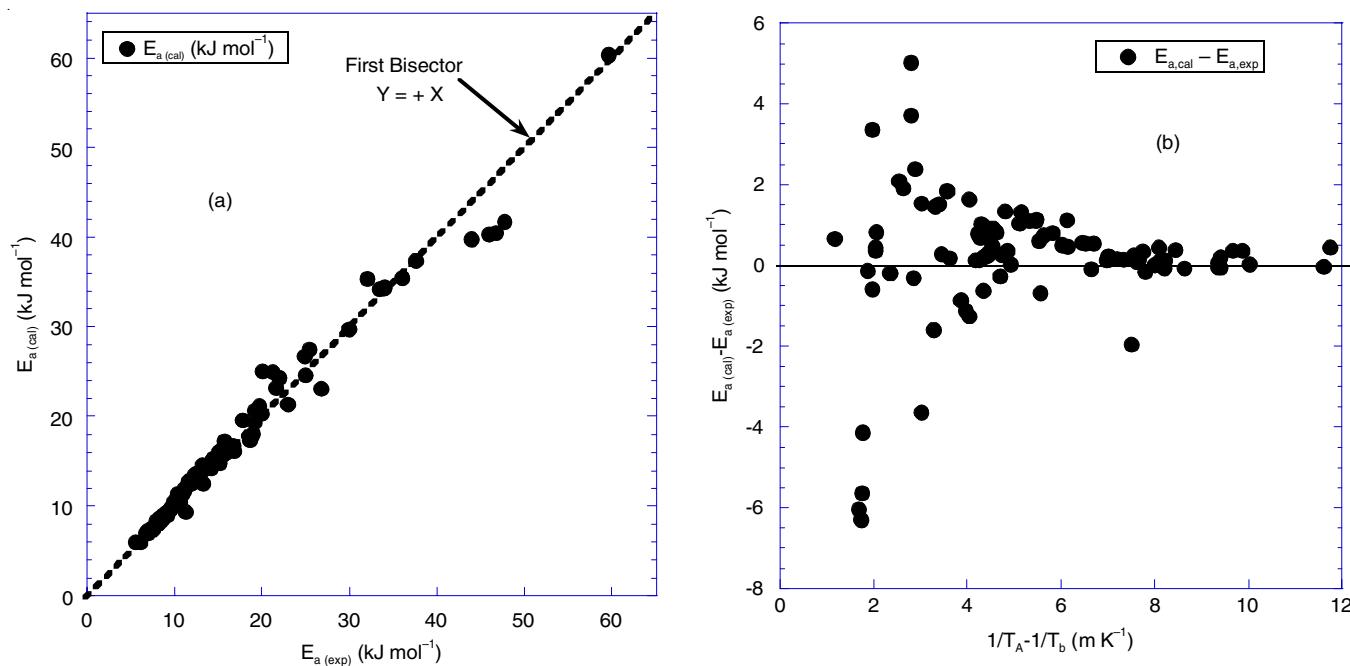


Fig. 4. Graphical comparison between the experimental ($E_{a,exp}$) and the estimated ($E_{a,cal}$) values of activation energies for some pure classical solvents [Ref. 15-52]

TABLE-4
RESULT OF STATISTICAL ESTIMATIONS OF THE eqn. 5

Eqn.	Parameter			R-squared	F-statistics
	ω	τ (K)	ε_0 (kJ mol^{-1})		
3	8.13 (6.50)	2500 (32.78)	2.98 (25.75)	0.998	7678

Note: Values between parentheses are t -statistics.

result allows as expecting a very predictive quality of approximation during their practical application.

Finally, regarding the new concept of boiling temperature prediction through the temperature-dependence of dynamic found in previous works [1-11] and the mutual correlation between the two Arrhenius parameters (E_a) and ($\ln A_s$) presented in earlier works [55,56], a 3D global view (Fig. 5) of graphical correlations between Arrhenius parameters and the boiling temperature of the set of studied classical solvents well confirms the investigated causal correlation presented in the present and previous works. We add that the quasi-linear dependence between the reciprocal Arrhenius temperature ($1/T_A$) and the transformed variable ($1/T_A - 1/T_b$) observed in Fig. 5, is none other than the statistical conflict between the endogenous and exogenous variables and not a real causal correlation.

Conclusion

Statistical methods are applied for the regression and correlation analysis for investigating the existence of any eventual causal correlation between the Arrhenius-type equation parameters and the boiling points of some classical Newtonian fluids. 101 data set of viscosity of pure Newtonian fluids studied at different temperature and at atmospheric pressure from the literature is utilized for empirical analysis. A significant strong nonlinear correlation between the boiling point (T_b), the Arrhenius activation energy (E_a) and the Arrhenius

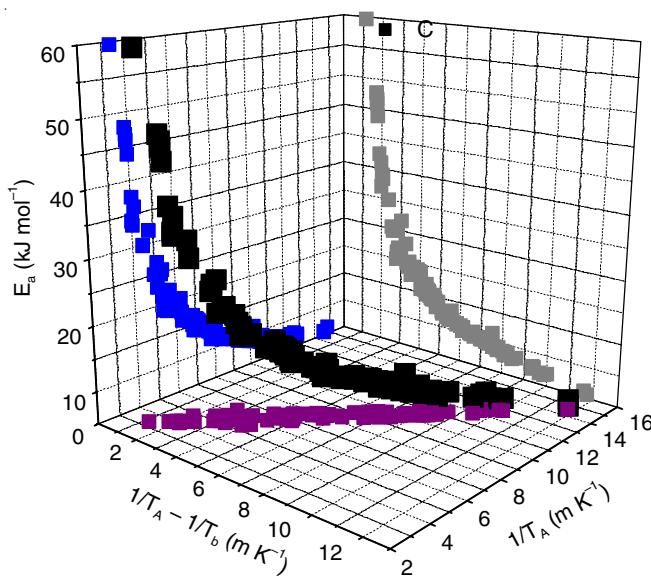


Fig. 5. 3D graphical correlations between Arrhenius activation energy (E_a), the reciprocal Arrhenius temperature ($1/T_A$) and the transformed variable ($1/T_A - 1/T_b$) for some pure classical solvents [Ref. 15-52]

temperature (T_A) is observed. In conclusion, we have proposed an empirical equation modeling of this relationship.

After all, for practical use and for giving some physical meaning to the suggested empirical model, to be fruitful for fluids engineering. We have made some variables transformations to obtain new semi-empirical models, which may be interesting for the theoreticians. This result is important given that the suggested equation allows the prediction of the boiling point only through information on viscosity Arrhenius parameters in Newtonian liquid state. Moreover, the suggested equation is very useful for engineering data especially for the study of systems efficiency and hydraulic components.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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