

Application of non-linear free energy relationships to parachors (P) of some aromatic and aliphatic compounds

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The present study involves the effect of substituents on parachors (P) of five aromatic and five aliphatic series. Non-linear Hammett and Taft correlations for both aromatic and aliphatic series have been observed. Both the plots give negative slopes (ρ and ρ^*) for electron donating groups and positive slopes (ρ and ρ^*) for electron withdrawing groups. The plots of aromatic series have intersecting point at H (Hammett $\sigma = 0$) and the plots of aliphatic series have their intersecting point at CH_3 (Taft $\sigma^* = 0$). We are the first ones to observe the conformity of LFER to the physical property parachor. Plausible interpretations for the conformity of LFER to parachors are presented.

Keywords: Parachor (P), Hammett equation, Taft equation, Non-linear free energy relationships (LFER)

Sugden¹ was the first to formulate an equation relating parachor (P) and surface tension (γ). There was an extensive review by Quayle² summarizing the parachors of several hundred molecules. Conception and significance of the parachor was briefly described by Exner³ describing it more lucidly is an additive property consisting of the sum of the parachors of individual atoms in a molecule. A lucid historical note by Saraswathi Balasubrahmanyam⁴ appeared in Current Science recently. Parachor of atomic silicon⁵ and its π -bond character in some silicon compounds were characterized with use of parachors⁶. Application of Hammett and Taft equations to parachor data did not find a place in literature except a small mention about application of Hammett equation to parachor data and thyromimetic activity without detailed explanation⁷. The present work is a detailed study of the observation of non-linear Hammett and Taft correlations to some aromatic and aliphatic compounds.

Methods

All correlations were done using KaleidaGraph software, Reading, PA, USA. All the chemical structures were drawn using ChemDraw. Parachor data is from refence². The parachors of some of the

compounds which are not available were calculated using atomic parachor data². All Hammett σ and Taft σ^* are values are from references⁸ and⁹.

Results and Discussion

Hammett and Taft equations are known as versatile tools for predicting organic reaction mechanisms¹⁰. Apart from the usual linear Hammett plots for several chemical reactions and for several physical properties, there are many reports on non-linear Hammett plots¹¹. The first ever report of the effect of substituents on dipole moments appeared as one paragraph note in Nature was in 1937 (Ref. 12) which was contemporary to the emergence of Hammett equation¹³. In this note a logical assumption is made about the correlation between dissociation constants of organic acids with their dipole moments. No detailed report is available in this publication¹¹. We have reported a V-shaped non-linear Hammett plot with suitable explanations from the study of effect of substituents on dipole moments of benzoic acids¹⁴. We have also reported a non-linear Taft relation applied to surface tensions of aliphatic acids and discussed on the inter-molecular hydrogen bonding *versus* intra-molecular hydrogen bonding¹⁵ and on the phase transition temperatures of several aliphatic

compounds¹⁶. In spite of the extensive study of application of both Hammett and Taft equations to physical properties the perceptiveness of chemists did not fall on parachor. We have taken up the present study in depth choosing five aromatic series comprising of toluenes, chlorobenzenes, nitrobenzenes phenols and methyl benzoates, and five aliphatic series comprising of acids, amines, alcohols, hydrocarbons, and methyl acetates.

It was clearly explained based on atomic and molecular dimensions like radii that the parachor is a function of molecular volume¹⁷ supporting Sugden's original interpretation. Therefore, one can assume that there will be a change in molecular size or molecular volume with addition of extra substituents to a molecule. The change in molecular volume of a molecule will be a function of the volume of the substituent and its electronic properties like intramolecular attractions or repulsions. And also, as pointed out by Saraswathi Balasubrahmanyam⁴ in her history and suggestion about parachor and molecular volume, a molecule is an aggregate of particles like electrons and nuclei that are held together by electrostatic and magnetic forces which are responsible for total volume of the molecule or atom. Therefore, at this juncture it is noteworthy to think about the change in molecular volume by addition of substituents. As shown in Fig. 1, the family of aromatic and aliphatic compounds with various substituents X and with a common functional group Y is supposed for discussion. As shown in Fig. 1A, if the substituent X is electron donating, it pushes the electron cloud toward the functional group Y *via* the bulk of the molecule resulting an increase in size of the molecules. And if

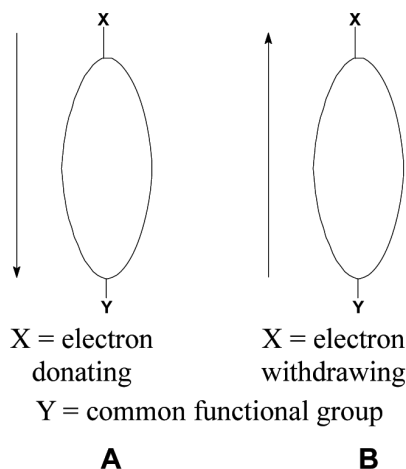


Fig. 1 — Family of aromatic and aliphatic compounds with various substituents

the substituent X is electron withdrawing as shown in Fig. 1B, it pulls the electron cloud *via* bulk of the molecule from the functional group Y resulting again an increase in size of the molecules. Therefore, whether the substituent X is either electron donating or electron withdrawing the size of the molecule ought to be more than the unsubstituted compound and it will be minimum. The only difference in the two situations is that the volume increase takes place due to electronic repulsions and attractions caused by the substituent X with opposite vectors. Therefore, one would expect an increase in size of the molecule with increasing electron donating or electron withdrawing capacities of the substituent with a minimum with the substituent H (Hammett $\sigma = 0.0$) in aromatic series and with CH_3 (Taft $\sigma^* = 0.0$) substituent in aliphatic series.

If we look at Fig. 1, one can clearly say that when we add the substituent at para-position, the dipole moment changes. An appreciable number of articles^{14,19-25} have been published about conformity of dipole moments to LFER. But from previous reports^{19,21,22,24,25} it appears that dipole moment arises because of polarity and since σ and σ^* is the index of polarity, the conformity to LFER is tested. Further Rao *et al.*,²¹ observed that Resonance seems to account for the influence of substituents on dipole moments just as on the reactivity of organic compounds. The effect of substituents on the reactivity of organic compounds is given by LFERs. Therefore, one would predict a relation of dipole moments of benzenes with reactivities. Rao *et al.*,²¹ conveyed that such correlations would only be crude approximation and the substituents for which the observed dipoles are rough measures of charge distribution approximately conformed to the LFER. This was the rationale they had considered to test the applicability of LFER to the physical property like dipole moment.

If one goes by the discussion in foregoing paragraph and observe Fig. 1, with the addition of substituent, there is change in parachor value and dipole moment. Dipole moments in some instances conform to LFER, hence parachor can also be tested for conformity to LFER. In fact, Fig. 2 to Fig. 11 clearly depict the conformity of LFER to parachor.

The basic equation for parachor is

$$P = \gamma^{1/4} M / (\rho_L - \rho_V) \quad \dots(1)$$

Where γ is the surface tension, M molar mass, ρ_L is the liquid density and ρ_V vapor density in equilibrium with liquid.

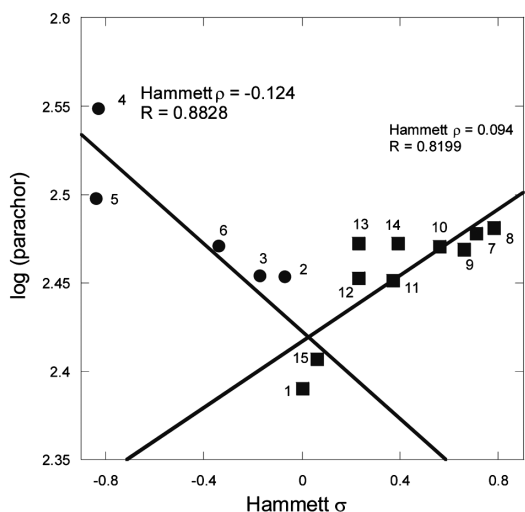


Fig. 2 — Plot of log (parachor) vs. Hammett σ of Toluenes

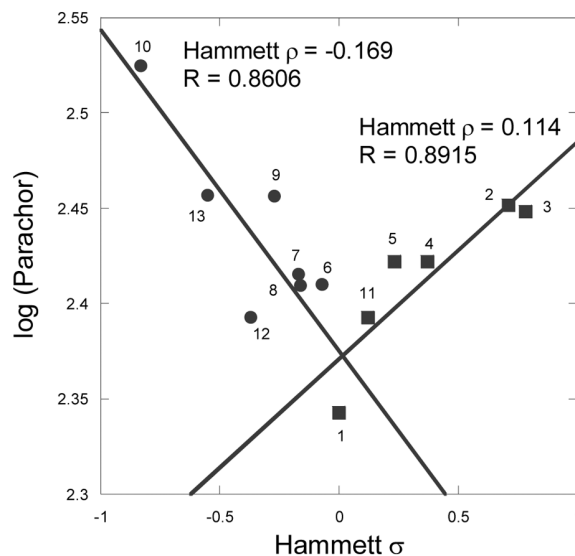


Fig. 5 — Plot of log (parachor) vs. Hammett σ of phenols

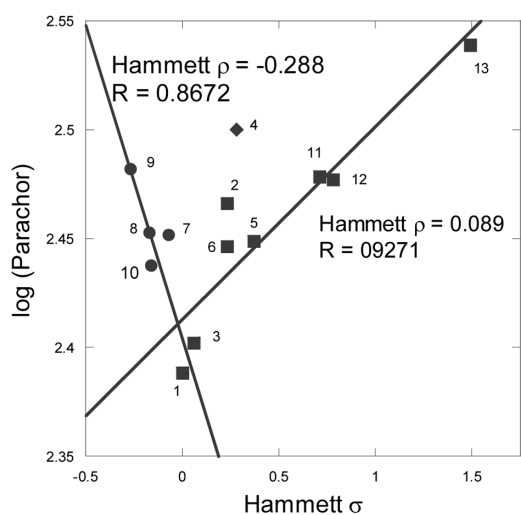


Fig. 3 — Plot of log (parachor) vs. Hammett σ of chlorobenzenes

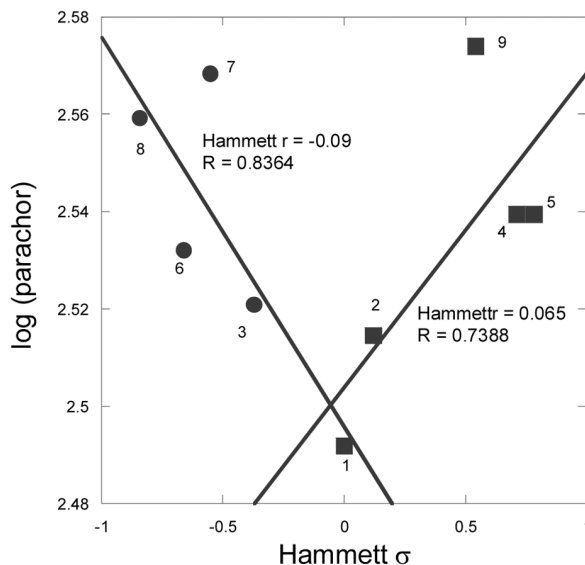


Fig. 6 — Plot of log (parachor) vs. Hammett σ of benzoates

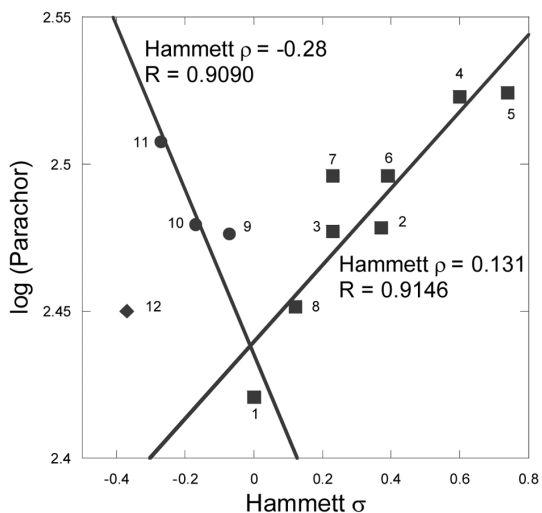


Fig. 4 — Plot of log (parachor) vs. Hammett σ of nitrobenzenes

At the temperature when surface tension is unity, parachor becomes equal to the molar volume. In other words, the parachor gives a measure of the molar volume at temperatures at which different liquids have the same surface tension. According to Kopp's law¹⁸, the molar volumes are additive, so parachor should also be additive.

When one mole of water is added to pure water, its volume contribution to water is different from when one mole of water is added to pure ethanol. This is a good example to convey the concept of partial molar volume concept.

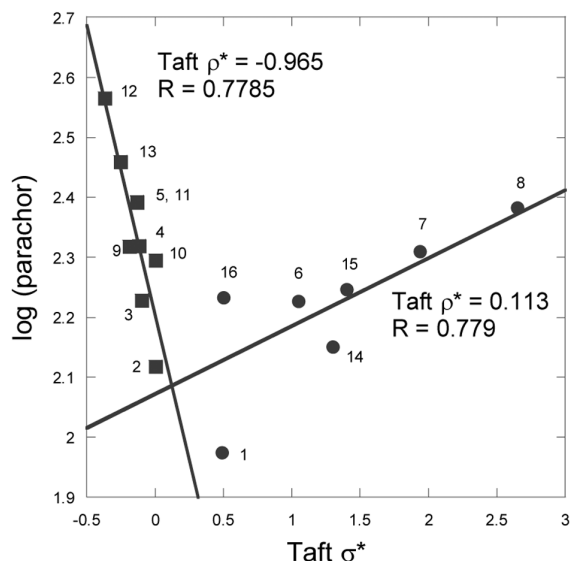


Fig. 7 — Plot of log (parachor) vs. Taft σ^* (aliphatic carboxylic acids, RCOOH)

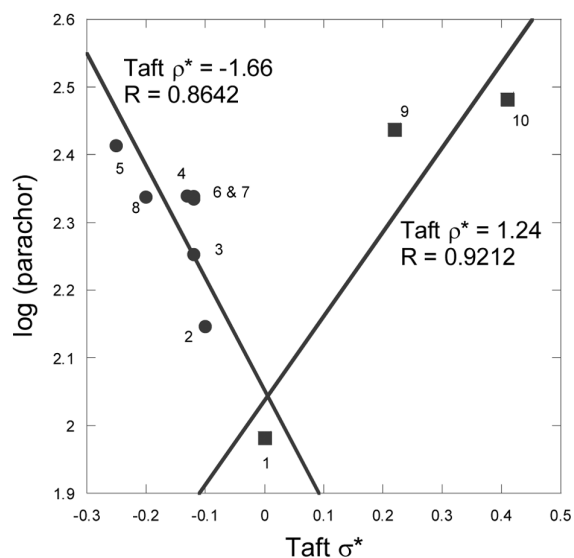


Fig. 8 — Plot of log (parachor) vs. Taft σ^* of aliphatic amines

Now, let us consider the parachor property. When considering parachor, the system in question is pure liquid and the vapor in equilibrium with the liquid. When we talk about density of liquid part of the system, it is mass per unit volume of the liquid. When we talk about vapor density part of the system, it is the mass per unit volume in vapor phase. The difference gives density of molecules which have not vaporized (those molecules which have not vaporized). Now $M \times V/M = V$ (right hand side part of equation 1, other than $\gamma^{1/4}$) which is volume of molecules which have not vaporized. The volume

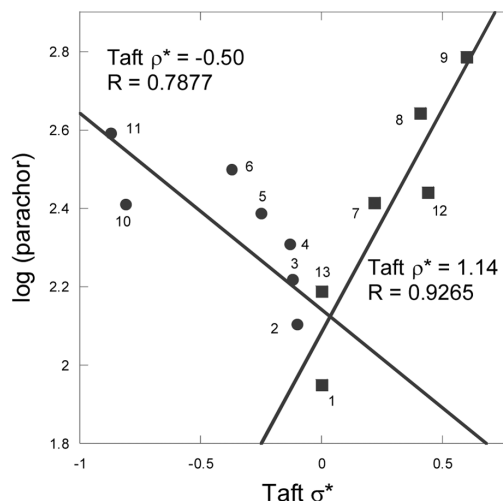


Fig. 9 — Plot of log (parachor) vs. Taft σ^* of aliphatic alcohols

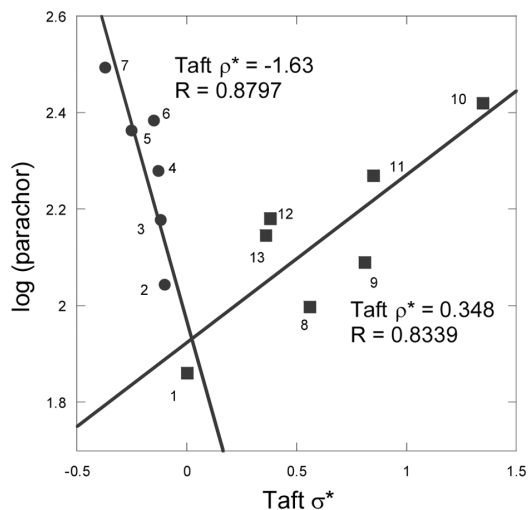


Fig. 10 — Plot of log (parachor) vs. Taft σ^* of aliphatic hydrocarbons

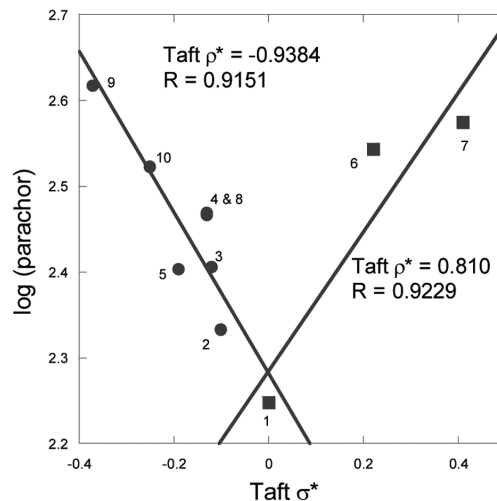


Fig. 11 — Plot of log (parachor) vs. Taft σ^* of aliphatic methyl esters

contribution of molecules in liquid phase would be different from volume contribution in vapor phase. (analogous to the volume contribution of one mole of water added to pure water is different from its volume contribution of one mole of water added to pure ethanol). Books and literature refer to this quantity as molar volume, but this can also be perceived as partial molar volume. To be more comprehensible, when methyl group is added at para position to Toluene, its volume contribution in liquid phase is different from its volume contribution in vapor phase. Thus, books and literature which refer to this quantity Parachor as molar volume, can also be perceived as partial molar volume. Since partial molar volume is related to chemical potential which obeys LFER, so should parachor. Parachor is the product surface tension and molar volume which is sought of partial molar volume. Parachor is directly proportional to surface tension and molar volume (sought of partial molar volume). Since both surface tension¹⁶ and molar volume (perceived as partial molar volume) conform to LFER, there is no reason why parachor should not conform to LFER (Fig. 2 to Fig. 11). Thus, the rationale for parachor conforming to LFER.

Further, according to Charles Law, at constant pressure volume is a function of temperature. Parachor is a molar volume and hence temperature dependent. Therefore, Parachor is considered to be an energy associated property; and energy associated property is likely to obey LFER.

In the backdrop of above plausible reasonings of parachor conforming to Linear Free energy relationships, one can write Hammett and Taft equations as:

$$\log\left(\frac{P_X}{P_H}\right) = \rho\sigma_X \quad \dots(2) \text{ Hammett equation}$$

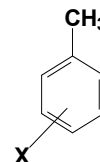
$$\log\left(\frac{P_X}{P_H}\right) = \rho^*\sigma_X^* \quad \dots(3) \text{ Taft equation}$$

where P_X is the parachor of substituted compounds and P_H that of unsubstituted compounds, ρ and ρ^* are the Hammett and Taft reaction constants respectively. And σ_X and σ_X^* are the Hammett and Taft substituent constants respectively.

There are quite a few physical properties which conform to LFER, such as quadrupole moment, dipole moment, stretching spectra, surface tension, melting points and boiling points. In fact, surface tension, melting point and boiling points conforming to LFER were reported by our group. This is the first detailed study of parachor conforming to LFER and plausible explanation presented.

Table 1 to Table 10 depict the data of the parachors and Fig. 2 to Fig. 6, and Fig. 7 to Fig. 11 are the Hammett and Taft plots for the five aromatic and five aliphatic series respectively. The numbers representing the points in the figures are their numbers in the tables.

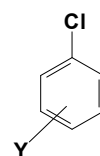
Table 1 — Parachor values of Toluenes



S. No.	X	Hammett σ	Parachors from Ref. 2
1	H	0.00	245.7
2	3-CH ₃	-0.07	284.2
3	4-CH ₃	-0.17	284.5
4	4-N(CH ₃) ₂	-0.83	353.7*
5	4-NH(CH ₃)	-0.84	314.7*
6	4-NHOH	-0.34	295.7*
7	3-NO ₂	0.71	300.6
8	4-NO ₂	0.78	302.8
9	4-CN	0.66	294.4
10	3-CN	0.56	295.6
11	3-Cl	0.37	282.8
12	4-Cl	0.23	283.6
13	4-Br	0.23	296.8
14	3-Br	0.39	296.8
15	4-F	0.06	255.3

*Calculated from the atomic parachors

Table 2 — Parachor values of chlorobenzenes

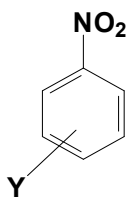


S. No.	X	Hammett σ	Parachor From Ref. 2
1	H	0.00	244.5
2	4-Br	0.23	292.5
3	4-F	0.06	252.4
4	4-I	0.28	316.4#
5	3-Cl	0.37	281.0
6	4-Cl	0.23	279.5
7	3-CH ₃	-0.07	283.6*
8	4-CH ₃	-0.17	283.6
9	4-MeO	-0.27	303.3**
10	3-NH ₂	-0.16	273.9**
11	3-NO ₂	0.71	300.9
12	4-NO ₂	0.78	300.0
13	3,4-di-NO ₂	1.49	346.0

#Assumed as it is a position isomer of 4-methyl-1-chlorobenzene.

**calculated using atomic parachors, #not included in the correlation

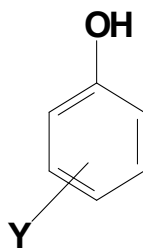
Table 3 — Parachor values of nitrobenzenes



Sl. No.	X	Hammett σ	Parachor from Ref. 2
1	H	0.00	263.6
2	3-Cl	0.37	300.9
3	4-Cl	0.23	300.0
4	3,4-diCl	0.60	333.4
5	3,5-diCl ₂	0.74	334.5
6	3-Br	0.39	313.3
7	4-Br	0.23	313.3
8	3-OH	0.12	282.9
9	3-CH ₃	-0.07	299.4
10	4-CH ₃	-0.17	301.6
11	4-OMe	-0.27	321.8
12	4-OH	-0.37	280.8*

*Not included in the correlation

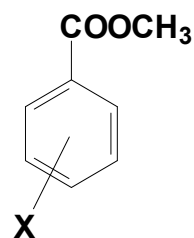
Table 4 — Parachor values of phenols



S. No.	X	Hammett σ	Parachor from Ref. 2
1	H	0.00	220.2
2	3-NO ₂	0.71	282.9
3	4-NO ₂	0.78	280.8
4	3-Cl	0.37	264.3*
5	4-Cl	0.23	264.3*
6	3-CH ₃	-0.07	257.1
7	4-CH ₃	-0.17	260.2
8	3-NH ₂	-0.16	256.7*
9	4-OMe	-0.27	286.1*
10	4-N(CH ₃) ₂	-0.83	334.7*
11	3-OH	0.12	247.1*
12	4-OH	-0.37	247.1*
13	4-NHNNH ₂	-0.55	286.3*

*Calculated from atomic parachors

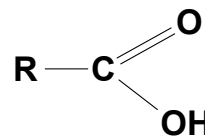
Table 5 — Parachor values of methylbenzoates



S. No.	X	Hammett σ	Parachor From Ref. 2
1	H	0.00	310.4
2	3-OH	0.12	327.0
3	4-OH	-0.37	331.8
4	3-NO ₂	0.71	346.3*
5	4-NO ₂	0.78	346.3*
6	4-NH ₂	-0.66	340.5*
7	4-NHNNH ₂	-0.55	370.1*
8	4-NHCH ₃	-0.84	362.4*
9	4-CF ₃	0.54	375.0*

*Calculated from atomic parachors

Table 6 — Parachor values of aliphatic acids



S. No.	R	Taft σ^*	Parachor From Ref. 2
1	H	0.49	94.3
2	CH ₃ -	0.00	131.2
3	CH ₃ CH ₂	-0.10	169.0
4	CH ₃ CH ₂ CH ₂ -	-0.12	208.6
5	CH ₃ CH ₂ CH ₂ CH ₂ -	-0.13	247.0
6	ClCH ₂ -	1.05	168.3
7	Cl ₂ CH-	1.94	203.8
8	Cl ₃ C-	2.65	241.3
9	(CH ₃) ₂ CH-	-0.19	207.9
10	CH ₂ =CH-CH ₂ -	0.00	197.3
11	(CH ₃) ₂ CHCH ₂ -	-0.13	246.3
12	CH ₃ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ -	-0.37	367.4
13	CH ₃ CH ₂ CH ₂ CH ₂ CH ₂ -	-0.25	288.0
14	CNCH ₂ -	1.30	118.2*
15	NO ₂ CH ₂ -	1.40	153.4*
16	NH ₂ CH ₂ -	0.50	170.8*

*Calculated using atomic parachors

Table 7 — Parachors of aliphatic amines

RNH ₂			
S. No.	R	Taft σ^*	Parachor from Ref. 2
1	CH ₃	0.00	95.9
2	Ethyl	-0.10	139.9
3	n-propyl	-0.12	178.9
4	n-butyl	-0.13	218.2
5	n-pentyl (n-amyl)	-0.25	258.9
6	iso-butyl	-0.12	216.1
7	sec-butyl	-0.12	217.8
8	(CH ₃ CH ₂) ₂ N-	-0.20	217.5
9	C ₆ H ₅ CH ₂	0.22	273.7
10	C ₆ H ₅ CH=CH	0.41	303.7*

*Calculated from atomic parachors

Table 8 — Parachor values of aliphatic alcohols

ROH			
S. No.	R	Taft σ^*	Parachor from Ref. 2
1	CH ₃	0.00	88.8
2	Ethyl	-0.10	126.8
3	n-propyl	-0.12	165.0
4	n-butyl	-0.13	202.9
5	n-pentyl	-0.25	243.3
6	n-heptyl	-0.37	315.5
7	Benzyl C ₆ H ₅ CH ₂	0.22	259.6
8	Diphenylmethyl (C ₆ H ₅) ₂ CH	0.41	439.0*
9	Triphenylmethyl (C ₆ H ₅) ₃ C	0.60 [#]	611.9*
10	Trimethylsilyl (Si(CH ₃) ₃)	-0.81	256.6*
11	Phenyl-di-methyl (Si(CH ₃) ₂ C ₆ H ₅)	-0.87	390.3*
12	Naphthylmethyl (C ₁₀ H ₇ CH ₂)	0.44	275.8*
13	Allyl (CH ₂ =CH-CH ₂)	0.00	154.0

[#]Estimated based on the σ^* values of benzyl and diphenylmethyl groups, *Calculated from atomic parachors

Table 9 — Parachor values of aliphatic hydrocarbons

RH			
S. No.	R	Taft σ^*	Parachor from Ref. 2
1	Methyl (CH ₃)	0.00	72.6
2	Ethyl (CH ₃ CH ₂)	-0.10	110.5
3	n-propyl (CH ₃ CH ₂ CH ₂)	-0.12	150.8
4	n-butyl (CH ₃ CH ₂ CH ₂ CH ₂)	-0.13	190.3
5	n-pentyl ((CH ₃ (CH ₂) ₃ CH ₂)	-0.25	231.0
6	Cyclohexyl (C ₆ H ₁₁)	-0.15	242.1
7	n-heptyl ((CH ₃ (CH ₂) ₅ CH ₂)	-0.37	311.3
8	Ethylene (CH ₂ =CH)	0.56	99.5
9	Allylene (CH=C-CH ₂)	0.81	122.9
10	Phenylacetylene (C ₆ H ₅ C≡C-)	1.35	263.0*
11	Propene (CH ₃ CH=CH-)	0.36	139.9
12	Chloroethyl (ClCH ₂ CH ₂ -)	0.38	151.6
13	Iodomethyl (CH ₂ I-)	0.85	186.5

*Calculated from atomic parachors

Table 10 — Parachor values of methyl esters of monobasic aliphatic acids

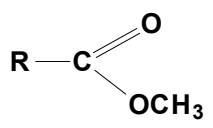
			
S. No.	R	Taft σ^*	Parachor from Ref. 2
1	CH ₃ -	0.00	177.1
2	CH ₃ CH ₂	-0.10	215.1
3	CH ₃ CH ₂ CH ₂ -	-0.12	254.5
4	CH ₃ CH ₂ CH ₂ CH ₂ -	-0.13	294.4
5	(CH ₃) ₂ CH-	-0.19	253.1
6	C ₆ H ₅ CH ₂ -	0.22	349.4
7	C ₆ H ₅ CH=CH-	0.41	375.2
8	(CH ₃) ₂ CHCH ₂ -	-0.13	292.6
9	CH ₃ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ CH ₂ -	-0.37	413.9
10	CH ₃ CH ₂ CH ₂ CH ₂ CH ₂ -	-0.25	333.2

Table 11 — Hammett and Taft reaction constants (ρ and ρ^*)

S. No.	Property/chemical series	Toluenes	Chloro benzenes	Nitro benzenes	Phenols	Methyl benzoates
1	Hammett ρ (EDS)	-0.12	-0.29	-0.28	-0.17	-0.09
2	Correlation coefficient (R)	0.8858	0.8672	0.9090	0.8606	0.8364
3	Hammett ρ (EWS)	0.09	0.09	0.13	0.11	0.07
4	Correlation coefficient (R)	0.8199	0.9271	0.9146	0.8915	0.7388
Sl. No.	Property/chemical series	Aliphatic acids	Aliphatic amines	Aliphatic alcohols	Aliphatic hydrocarbons	Aliphatic methyl esters
5	Taft ρ^* (EDS)	-0.97	-1.66	-0.50	-1.63	-0.94
6	Correlation coefficient (R)	0.7785	0.8642	0.7877	0.8797	0.9151
7	Taft ρ^* (EWS)	0.11	1.24	1.14	0.35	0.81
8	Correlation coefficient (R)	0.7790	0.9212	0.9265	0.8339	0.9229

EDS: electron substituents and EWS: electron withdrawing substituents

Conclusions

Though the correlation coefficients are poor, the trends are unmistakable. Table 11 gives the summary of all Hammett and Taft reaction constants. From the loci of Hammett and Taft plots it is understood that the higher the Hammett and Taft substituent constants, the more are the parachor of the molecules.

With the addition of substituent, there is a change in parachor value and dipole moment (which conforms to LFER). Hence parachor value was tested for conformity of LFER. The dipole moment arises because of polarity, σ and σ^* is the index of polarity. The foregoing statement is further reinforced by the higher the Hammett and Taft substituent constants, the more are the parachor of the molecules.

Conflict of interest

The authors do not have any conflict of interest.

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