A RE-EXAMINATION OF THE EÖTVÖS EQUATION

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The Eötvös equation (*1*) was re-examined from a purely empirical standpoint on a restricted but carefully selected set of compounds; the main intention was to test how an empirical equation of several variables can be verified by experiments. At a fixed temperature Eq. (*1*) is able to predict one of the three experimental quantities — surface tension, density, critical temperature — from the remaining two with a fair precision, but the range of validity as to the structure is rather restricted. At variable temperature and for a given compound, Eq. (*1*) describes the temperature dependence of surface tension provided the temperature dependence of density is known; however, the precision is low and a simpler and a more effective equation can be derived. If both temperature and structure are varied, Eq. (*1*) infers relationships of very low precision or even clearly invalid.

The Eötvös equation[1-3] (*1*) relates the surface tension γ and the relative density d of a given liquid compound with its relative molecular weight M and critical temperature T_c:

$$(\forall X)(\forall T < T_c) \quad \gamma d^{-2/3} M^{2/3} = k(T_c - T - \vartheta) . \qquad (1)$$

The left-hand side is called the molar surface energy. The symbols $\forall$ were added to show explicitly that the equation should hold for variable temperature T (lower than T_c) and for variable structure X as well. More exactly, it is supposed to hold[3-5] for non-associated liquids with common constants $k = 2{\cdot}12 \,.\, 10^{-3}\ \text{kg s}^{-2} \,.\, \text{K}^{-1}$ and $\vartheta = 6$ K. For more polar, associated liquids k is lower and no more independent of temperature; many such observations even for less polar liquids lead finally to the conclusion[2] that the equation is actually invalid. In practice Eq. (*1*) could be recommended for estimating any of the quantities involved (even the molecular weight[4]) from the remaining ones; alternatively and more frequently it may be viewed as a dependence of γ on temperature[5] provided the temperature dependence of d is known. However, the experimental verification[2-6] has been carried out only by plotting the left-hand side against T for a particular compound and estimating k. This test is of low statistical weight and rather incomplete. In particular it does not allow to optimize all the constants involved, not does it reveal the relative importance of each variable quantity. (The example of the parachor demonstrated[7] that a quantity involved in an empirical relationship can be almost without significance for its validity, the cause being a particular mathematical form and/or the stochastic dependence of certain quantities). The precision of Eq. (*1*) has never been evaluated statistically and the range of validity is not known with more precision.

The present paper is concerned with the re-examination of the Eötvös equation from a purely empirical point of view, it means that the agreement with experimental data is considered as the only criterion of validity. It is, however, not a trivial task to evaluate this agreement statistically if several variables are present. The following points deserve special attention: *1*) The range of validity with respect both to the structure of the liquid compound and temperature must be delimited (even when approximately). *2*) Optimum values must be found for all the constants involved. It means that not only k and ϑ in Eq. (*1*) but also the exponents at M, d, and γ are to be treated as empirical constants. *3*) The "validity" of an empirical equation means in fact its predicting power[8], as expressed *e.g.* by the statistic ψ. With a relationship of several variables this predicting power need not be equal for each of them and must be examined separately. *4*) The above-mentioned examination can be affected by stochastic dependence of some variables. *5*) The five variable equantities involved in Eq. (*1*) are very different in character: two of them (γ and d) depend on structure and temperature as well, two (M and T_c) on structure only, and one on temperature only (T itself). Hence it seems necessary to investigate separately the dependence on structure at a fixed temperature and the dependence on temperature for a particular compounds.

All these tasks could not be fulfilled exactly due to lack of necessary experimental data. However, we see the main reason of the present investigation in testing on a practical example, how an empirical relationship of several variables should be examined before it is said to be experimentally verified. Another reason why the Eötvös equation deserves a reinvestigation is its relation to the equation of Macleod[9], formulated more frequently using the concept of parachor[10,11]:

$$(\forall T) \quad \gamma^{1/4} d^{-1} M = P \qquad (2)$$

Since the parachor P depends on structure, Eq. (*2*) can be valid only for variable temperature. In this application it has never been compared to Eq. (*1*), although the two equations are treated in many textbooks almost on the same page[3,4]. In principle, Eq. (*1*) and (*2*) can either be at variance so that one has to decide which is better, or they can be valid simultaneously and further relationships can be inferred from them. The parachor itself has been disproved as an additive quantity, but the temperature dependence according to Eq. (*2*) is approximately fulfilled in a restricted range of compounds[7].

EXPERIMENTAL

The values of the surface tension, relative density, and critical temperature were selected mostly from Timmermans' tables[12], only exceptionally from another source[13]. Whenever possible, the values for one compound were taken from one author. In general we preferred always to have a restricted number of actually reliable data. If a series of very similar compounds was at disposal (alkanes), only several representatives were selected in order to obtain a set with as diverse structures as possible.

As a fixed temperature we chose $T = 293$ K. In this case the number of compounds is restricted mainly by the availability of T_c. After the delimitation of the range of validity, the basic set contained 18 compounds: n-pentane, 2,2-dimethylbutane, 2,3-dimethylbutane, cyclopentane, methylcyclopentane, cyclohexane, methylcyclohexane, benzene, toluene, tetrachloromethane, bromoethane, 1-chloropropane, fluorobenzene, chlorobenzene, bromobenzene, diethyl ether, dimethyl sulfide, dimethyl disulfide.

The temperature coefficients $\partial\gamma/\partial T$ and $\partial d/\partial T$ were determined from measurements[12,13] at 293 and 303 K, exceptionally at 288 and 303 K. They were assumed as valid for $T = 293$ K. In this case it was most difficult to obtain the values of $\partial\gamma/\partial T$, on the other hand T_c was no more needed for the equations examined. Hence the basic set was extended by including some compounds originally eliminated (n-hexane, ethylbenzene, propylbenzene, butylbenzene, *cis*-decalin, *trans*-decalin) and the following additional ones: dichloromethane, trichloromethane, iodomethane, 1,2-dibromoethane, iodoethane, perfluoroheptane, 1-chlorooctane, 3-chloropropene, α-chlorotoluene, 1-bromonaphthalene, methoxybenzene, diethyl sulfide, carbon disulfide. As the value of $\partial\gamma/\partial T$ for 2,2-dimethylbutane and 2,3-dimethyl butane were not available, this extended set consisted of 35 compounds.

RESULTS AND DISCUSSION

Relationship at a Fixed Temperature

For a given temperature, say $T = 293$ K, Eq. (*1*) is a relationship between four properties of a given liquid compound (γ, d, M, T_c). When the left-hand side is plotted against T_c (Fig. 1), the range of validity can be roughly delimited and the constants K and ϑ estimated. As anticipated, compounds with strongly polar groups violate Eq. (*1*) clearly; even the presence of a carbonyl group is sufficient. Fig. 1 reveals also another deviation, not yet noted, *viz.* that of higher alkanes. A similar trend, although less pronounced, is observed with alkyl benzenes. These compounds, alkanes beginning with C_6H_{14} and alkyl benzenes beginning with ethylbenzene, must be excluded from the validity range. Similar deviations of higher esters were noticed[3], but hydrocarbons have been assumed to follow Eq. (*1*) exactly. In qualitative terms one could say that the Eötvös equation is best fulfilled for rigid, approximately spherical molecules. It is not clear whether it holds for higher branched paraffins, since *e.g.* 2,2,4-trimethylpentane deviates distinctly and was excluded, too, together with *cis*- and *trans*-decalins. For the remaining set of 18 compounds (the basic set – see Experimental) the correlation is excellent according to the common evaluation of the correlation coefficient (Table I, line *1*), but is only good according to the more rigorous statistic ψ.

The statistic ψ is a generally applicable measure of agreement between theory and experiment. It has been originally advanced for large sets and its statistical distribution was not dealt with[8]. If used for smaller sets, its value must be viewed as an estimate with unknown confidence interval.

In our case it is physically more meaningful than some common statistics, *e.g.* the "goodness of fit"[14] or R-factor[15], since the average values of γ, d, M, or T_c are far from zero, and their variances are not very large.

In any case the correlation is not sufficiently precise for determining the two constants, k and ϑ, simultaneously with a reliability. The least-squares values (Table I) are $k = 2{\cdot}16$ with a standard deviation of 0·04, and $\vartheta = 3{\cdot}4 \pm 4{\cdot}6$. These values correspond to the full straight line in Fig. 1, but the original value $k = 2{\cdot}12$ together with $\vartheta = 0$ (dashed line) give practically the same fit; only the combination $k = 2{\cdot}12$ and $\vartheta = 6$ is evidently not good (dot-and-dashed line). Due to the large uncertainty in ϑ we considered the choice $\vartheta = 0$ most reasonable. Further calculations are carried out with this value but the results are not sensitive to it. The choice[3,4] $\vartheta = 6$, hitherto usual, was probably conditioned by inclusion of more polar compounds into the validity range.

In the next step we transferred Eq. (*1*) into logarithms and solved it successively according to all the variables γ, d, M and T_c. The correlations were calculated by sim-

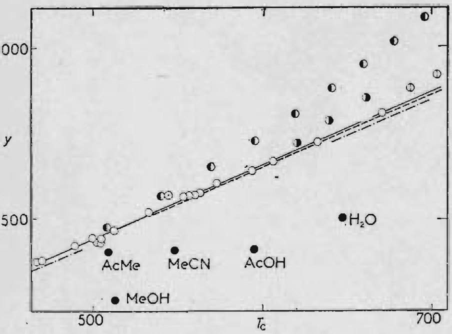

FIG. 1

Test of the Eötvös equation at a fixed temperature (293 K). The values $y = 10^{-3}\gamma \cdot d^{-2/3}M^{2/3}$ are plotted against T_c (K); ○ the basic set of nonassociated, little polar compounds, ◐ higher alkanes C_6—C_{12}, ◑ alkylbenzenes C_2—C_4, ⦶ *cis*- and *trans*-decalins, ⊙ 2,2,4-trimethylpentane, ● strongly polar compounds. Full line is the regression line with $k = 2{\cdot}16$, $\vartheta = 3{\cdot}4$; broken line corresponds to $k = 2{\cdot}12$, $\vartheta = 6$, dot-and-dashed line to $k = 2{\cdot}12$, $\vartheta = 0$

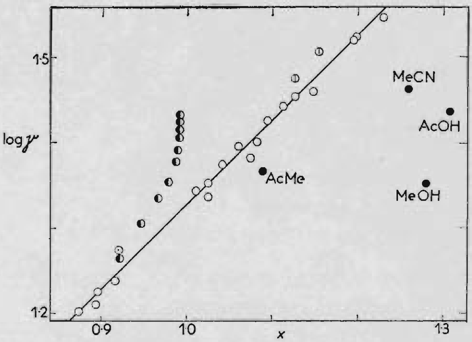

FIG. 2

Test of the Eötvös equation at a fixed temperature in the logarithmic scale. The values of $\log(10^{-3}\gamma)$ are plotted against $x = \log . [d^{2/3}M^{2/3}(T_c - 293)]$; see Fig. 1 for descriptive details ple regression with the remaining variables taken together (lines *2*, *4*, *6*, and *8* of Table I), or by multiple regression with all the variables separately (lines *3*, *5*, *7*, and *9*) The latter procedure allows to verify the values of the exponents $-2/3$ at d, $2/3$ at M,

TABLE I

Statistical test of the Eötvös equation at a fixed temperature (293 K)

No	Dependent variable	Independent variables	Correl. coeff. ψ^a	Standard deviations[b]	Constant k^c	Exponents[d] at d M	Exponents[d] at γ $T_c - 293$
1	$d^{-2/3}M^{2/3}\gamma$	T_c	0·9978 0·069	8·39[e] 122·3[e]	2·16		
2	$\log(T_c - 293)$	$\log(d^{-2/3}M^{2/3}\gamma)$	0·9952 0·11	0·010 0·095	2·06		
3	$\log(T_c - 293)$	$\log d$, $\log M^f$, $\log \gamma$	0·9987 0·061	0·005 0·095	1·32	−0·56 0·59	0·97 —
4	$\log \gamma$	$\log[d^{2/3}M^{-2/3}(T_c - 293)]$	0·9968 0·085	0·0089 0·108	2·12		
5	$\log \gamma$	$\log d$, $\log M^f$, $\log(T_c - 293)$	0·9989 0·054	0·0053 0·108	1·35	−0·58 0·60	— 1·03
6	$\log d$	$\log[M\gamma^{3/2}(T_c - 293)^{-3/2}]$	0·9960 0·099	0·012 0·125	2·17		
7	$\log d$	$\log M$, $\log \gamma$, $\log(T_c - 293)^f$	0·9977 0·082	0·0093 0·125	1·18	— 0·68	1·13 1·16
8	$\log M$	$\log[d\gamma^{-3/2}(T_c - 293)^{3/2}]$	0·9930 0·13	0·013 0·107	2·21		
9	$\log M$	$\log d$, $\log \gamma$, $\log(T_c - 293)^f$	0·9971 0·092	0·0089 0·107	1·15	−0·64 —	1·08 1·12
10	$\log\left(\frac{\gamma}{T_c - 293}\right)$	$\log(dM^{-1})$	0·9920 0·13	0·0057 0·044	1·48		
11	$\log\left(\frac{\gamma}{T_c - 293}\right)$	$\log d$, $\log M^f$	0·9922 0·13	0·0058 0·044	1·51	−0·59 0·59	— —

[a] $\psi = s_{y,x}\sqrt{(17)}/s_y\sqrt{16}$ or $s_{y,x1x2x3}\sqrt{(17)}/s_y\sqrt{14}$, respectively; [b] standard deviation from the regression $s_{y,x}$ or $s_{y,x1x2x3}$, followed by the standard deviation of the independent variable s_y are given, $n = 18$ in all correlations; [c] in 10^{-3} kg s^{-2} K^{-1}; [d] optimized values of the exponents at the respective variable in Eq. (*1*); [e] in 10^{-3} kg s^{-2}; [f] the least significant term in the respective regression.

and the implicit exponents of 1 at γ and at $(T_c - T)$. Comparison of the two respective lines reveals that the multiple regressions are generally not so much better to justify an improvement of exponents. This improvement would be relatively most significant for the exponent at d, *i.e.* $-0{\cdot}56$ to $-0{\cdot}64$ instead of $-0{\cdot}667$, but even in this case it is not of decisive importance. Considering that the exponents $-2/3$ and $2/3$ have been derived theoretically when defining the molar surface energy, we do not recommend to amend these values on an empirical basis. In any case the suggested exponents[3,16] at d and M, -1 and $+1$, respectively, are inacceptable. The values of the exponents also affect the value of constant k which is obtained from simple regressions in agreement with the first estimate as $2{\cdot}16 \pm 0{\cdot}04$. Multiple regressions yield much lower and less constant values.

Table I reveals further that the Eötvös equation is fulfilled with a uniform acceptable accuracy, irrespective of which quantity is chosen as the dependent variable (see the values of ψ). It means that any of the quantities γ, d, M, and T_c can be predicted from the remaining three. Actually, the best precision is obtained for γ, *viz.* 0·0089 log units, corresponding to a relative error of 2·1%. Least suitable would be the equation for determining the molecular weight (0·013 log units, *i.e.* 3%). The most significant result from this part of our investigation is that all the quantities involved are of comparable importance and none can be omitted. This follows from the similar values of ψ in all the correlations, and is confirmed by the statistical significance of individual terms of the multiple regression (F-test – the values are not tabulated). The least significant term in each regression is indicated in Table I but the differences are not marked. The least significant of the whole table is the term in $\log (T_c - 293)$ in line *7* but even in this case the F-test is positive at the 0·005 level. Note still that the logarithmic transformation does not impair the results obtained because the range of the variables γ, d, M, and $(T_c - T)$ is relatively narrow (approximately ±40% from the average for any quantity). This conclusion is corroborated by the essential agreement of lines *1* and *2* of Table I, see in particular the correlation coefficients and slopes k.

The last point at issue is a possible stochastic dependence of the quantities involved. For instance it is known from the investigation of the parachor[7] that higher values of surface tension are more often encountered together with higher values of density, this makes a prediction of γ less efficient. As regards the four variables of Eq. (*1*), the highest value in the correlation matrix ($r = 0{\cdot}915$) was found for the pair $\log \gamma$ and $\log (T_c - T)$. Since the correlation coefficient is positive, a partial compensation could take place within the quotient $\gamma/(T_c - T)$, making it less variable and less dependent on the remaining quantities. This hypothesis was tested by the correlations of this quotient, lines *10* and *11*. Table I. The correlation is worse thena both for $\log \gamma$ (lines *4* and *5*) and for $\log (T_c - T)$ (lines *2* and *3*) alone but it is though highly significant. As expected the combined expression $\gamma/(T_c - T)$ is less variable (see the standard deviation s_y) but is still far from being constant. Hence the stochastic dependence does not impair the validity of Eq. (*1*).

In conclusion, our appreciation of the Eötvös equation is between the utmost scepticism[2] and undue belief[3,4]: It holds as an empirical relationship at a fixed temperature and within its rather narrow range of validity as regards the structure. The accuracy is not high and the equation cannot be recommended for reliable estimation of one of the quantities involved when the others are determined experimentally; such procedure[4] would be in particular impracticable for the molecular weight. Even so, an approximate relationship between the four quantities undoubtedly exists for many simple molecules and a task arises to explain it in terms of structural theory.

Relationship at Variable Temperature

Two of the variables (d, γ) involved in Eq. (1) are functions of temperature. Hence the equation can express the temperature dependence of one of them provided the other is known; more natural would be to predict the values of γ from known d. In this function the Eötvös equation is to be confronted with the parachor expression. Eq. (2). If both equations are solved according to γ and temperature is considered as the only variable, one gets:

$$(\forall T) \quad \gamma = k d^{2/3} M^{-2/3} (T_c - T - \vartheta) \qquad (1a)$$

$$(\forall T) \quad \gamma = P^4 d^4 M^{-4} \qquad (2a)$$

If these equations are to be tested, one must take into consideration that P in Eq. $(2a)$ varies with structure and must be optimized for each particular compound. In order to obtain comparable conditions, even k in Eq. $(1a)$ was given an *ad hoc* value for each compound; then both equations describe purely the dependence on temperature. Some typical results are shown in Fig. 3; the data were restricted to the existence of the liquid phase at atmospheric pressure. For non-polar compounds either equation is satisfactory, while for strongly polar compounds the deviations can be quite big. We do not believe that a decision between Eqs $(1a)$ and $(2a)$ would be possible on the basis of relationships like Fig. 3; one reason is also lack of experimental data for other compounds than for parafins. Still more important, such a decision would not be of much value since a more efficient relationship can be inferred as follows.

Let us assume that Eqs $(1a)$ and $(2a)$ are not exclusive but both valid simultaneously. Then by eliminating d or γ we get respectively:

$$(\forall T) \quad \gamma = \text{const}\,(T_c - T)^{6/5} \qquad (3)$$

$$(\forall T) \quad d = \text{const}\,(T_c - T)^{3/10} \qquad (4)$$

when the values of k, P, and M were contracted into one constant and $\vartheta = 0$ was assumed. These relationships are not completely new. Eq. (3) was derived in a modified form by Lövgren[17] but is written more often with an empirical exponent[18] the optimum values of which is given to 1·23. Fig. 3 reveals that Eq. (*3*) is as good as Eqs (*1a*) and (*2a*) while it is much simpler; if necessary even T_c can be empirically estimated as an adjustable constant. Eq. (*4*) with an empirical exponent is also known[19] and will not be tested here. We conclude that neither the Eötvös equation nor the parachor yield useful relationship for the temperature dependence since simpler equations can be derived.

Relationship with Variable Temperature and Structure

There is a lack of systematic data varying both in structure and in temperature simultaneously, as required by Eq. (*1*) in its general form. Hence we must be satisfied

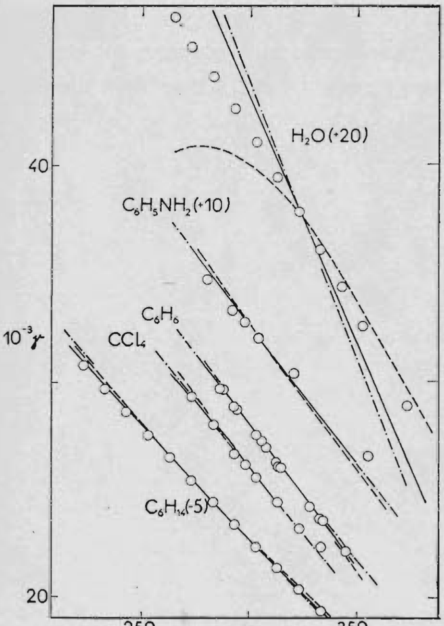

FIG. 3

The dependence of the surface tension $10^{-3}\gamma$ on temperature T (K). Full lines — the Eötvös equation (*1a*) with k adjusted for every compound, broken lines — the parachor equation (*2a*) with P adjusted, dot-and-dashed lines — Eq (*3*). The curves for three compounds are shifted as indicated in parentheses

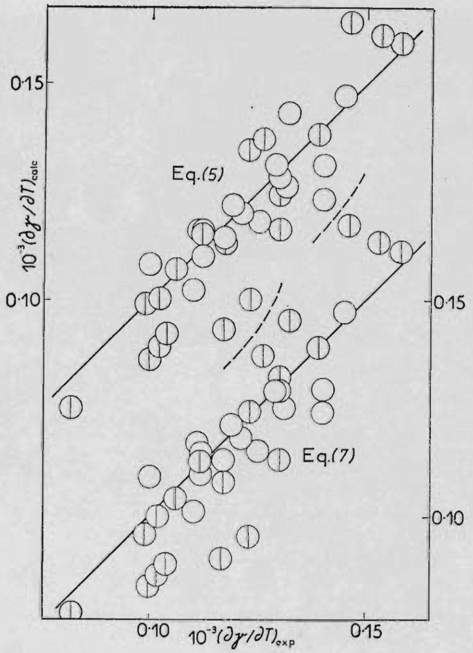

FIG. 4

Test of the Eötvös equation (*1*) at variable temperature (in the vicinity of 293 K). The calculated values of $-10^{-3}\,\partial\gamma/\partial T$ are plotted against the experimental ones; at the top Eq. (*5*), at the bottom Eq. (*7*); ○ original basic set of compounds, ⦶ additional compounds with a restriction to the neighbourhood of a given temperature, say again 293 K. By differentiating Eq. (*1*) with respect to temperature and substituting for T_c we get:

$$(\forall X)\quad \frac{\partial \gamma}{\partial T} = \frac{2}{3}\gamma d^{-1}\frac{\partial d}{\partial T} - kd^{2/3}M^{-2/3} \tag{5}$$

Similar expressions can also be derived containing T_c instead of one of the variables γ, d, or M, but we preferred to eliminate just T_c which is still the most difficult to find in the literature. Eq. (*5*) can be tested in a single graph (Fig. 4). The accuracy is evidently quite low and is not further lowered by including some additional compounds which partly even exceed the original range of validity as defined in the case of Figs 1 and 2 (see the Experimental). The pertinent statistics are listed in Table II. According to the value of ψ, Eq. (*5*) cannot be considered a valid empirical relationship. It is true that a part of the observed scatter may be due to the experimental uncertainty in $\partial\gamma/\partial T$; nevertheless the size of points in Fig. 4 corresponds to the standard deviation of 0·03 (10^{-3} N m^{-1}) in measuring the surface tension and this accuracy should be attainable (see[13]).

TABLE II

Statistical test of the Eötvös equation and of the parachor at variable temperature and structure

	Equation	(*5*)	(*6*)	(*7*)	(*7a*)	(*8*)	(*8a*)
1	dependent variable y	$\frac{\partial\gamma}{\partial T}$	$\frac{\partial\gamma}{\partial T}$	$\frac{\partial\gamma}{\partial T}$	$\frac{\partial\gamma}{\partial T}$	$\frac{\partial d}{\partial T}$	$\frac{\partial d}{\partial T}$
2	mean value of $y - y_{calc}$	$9{\cdot}7^a$	$8{\cdot}0^a$	$10{\cdot}9^a$	$8{\cdot}4^a$	132^b	69^b
3	ψ	0·57	0·47	0·64	0·54	0·24	0·14
	linear regression:						
4	correlation coefficient	0·897	0·897	0·894	0·870	0·979	0·993
5	slope^c	0·719	0·916	0·673	0·907	0·908	0·934
6	standard deviation	$7{\cdot}6^a$	$7{\cdot}6^a$	$7{\cdot}7^a$	$7{\cdot}9^a$	116^b	55^b
7	ψ	0·45	0·45	0·46	0·52	0·21	0·12
8	number of points	35	35	35	23	35	23

[a] In 10^{-6} N m^{-1} K^{-1}; [b] in 10^{-6} K^{-1}; [c] the theoretically expected value of the slope is unity in all cases since the coefficients $\frac{2}{3}$, 4, $-\frac{6}{5}$, $-\frac{3}{10}$ have been included into the independent variable.

When Eq. (*2*) is treated in a similar manner as above. one gets:

$$(\forall X) \quad \frac{\partial \gamma}{\partial T} = 4\gamma d^{-1} \frac{\partial d}{\partial T} \tag{6}$$

This equation was derived and tested by us previously[7] but its precision was possibly somewhat overestimated. According to Table II it is not more precise than Eq. (*5*) and one cannot say which of them is better. As shown in lines *4*–*8*, the precision is not evidently improved by a linear regression, it means by applying linear corrections in Eqs (*5*) or (*6*). Because of this low precision we have not carried out a more detailed statistical investigation as it was done in the case of Eq. (*1*).

Another possible treatment is again to consider Eqs (*5*) and (*6*) as simultaneously valid like in the preceding part with Eqs (*3*) and (*4*). Then the following expressions can be derived for $\partial\gamma/\partial T$ and $\partial d/\partial T$ separately:

$$(\forall X) \quad \frac{\partial \gamma}{\partial T} = -\frac{6}{5} k d^{2/3} M^{-2/3} \tag{7}$$

$$(\forall X) \quad \frac{\partial \gamma}{\partial T} = -\frac{6}{5} \gamma/(T_c - T) \tag{7a}$$

$$(\forall X) \quad \frac{\partial d}{\partial T} = -\frac{3}{10} k d^{5/3} M^{2/3} \gamma^{-1} \tag{8}$$

$$(\forall X) \quad \frac{\partial d}{\partial T} = -\frac{3}{10} d/(T_c - T) \tag{8a}$$

A graphical test of Eq. (7) is shown in Fig. 4, the statistics are given in Table II for all the equations. The only one which may be viewed as valid with a fair accuracy is Eq. (*8a*), but even it cannot be of practical importance either for predicting $\partial d/\partial T$ from T_c or *vice versa*. The somewhat higher precision of Eqs (*8*) and (*8a*) compared to (*5*)–(*7*) may be partly due to better experimental accuracy of $\partial d/\partial T$ than $\partial \gamma/\partial T$. In any case the conclusion seems safe that the Eötvös equation does not postulate any useful and valid relationship if both structure and temperature are varied.

REFERENCES

1. Eötvös R.: Ann. Phys. (New York) *27*, 448 (1886).
2. Jellinek K.: *Lehrbuch der Physikalischen Chemie*, Vol. I, p. 862. F. Enke, Stuttgart 1928.
3. Partington J. R.: *An Advanced Treatise on Physical Chemistry*. Vol. 2, p. 157. Longmans, Green & Co., London 1955.

4. Brdička R.: *Základy fyzikální chemie*, p. 171. Published by Přírodovědecké vydavatelství, Prague 1952.
5. Hála E., Reiser A.: *Fysikální chemie*, Vol. 1, p. 156. Published by Nakladatelství ČSAV, Prague 1960.
6. Jaeger F. M.: Z. Anorg. Allg. Chem. *101*, 1 (1917).
7. Exner O.: This Journal *32*, 24 (1967).
8. Exner O.: This Journal *31*, 3222 (1966).
9. Macleod D. B.: Trans. Faraday Soc. *19*, 38 (1923).
10. Sugden S.: J. Chem. Soc. *1924*, 1177.
11. Ref.[3], p. 144.
12. Timmermans J.: *Physico-Chemical Constants of Pure Organic Compounds*, Vol. 1 and 2. Elsevier, Amsterdam 1950 and 1965.
13. Rossini F. D., Pitzer K. S., Arnett R. L., Braun R. M., Pimentel G. C.: *Selected Values of Physical and Thermodynamic Properties of Hydrocarbons and Related Compounds*. Carnegie Press, Pittsburgh 1953.
14. Wells P. R., Ehrenson S., Taft R. W.: Progr. Phys. Org. Chem. *6*, 147 (1968).
15. Hamilton W. C.: Acta Crystallogr. *18*, 502 (1965).
16. Walden P.: Z. Phys. Chem. *65*, 129 (1908).
17. Lövgren N.: Sv. Kem. Tid. *53*, 359 (1941).
18. Ref.[3], p. 141.
19. Ref.[3], p. 47.

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