

Weighted Self-Returning Walks for Structure-Property Correlations*

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Two algorithms are developed for the weighting of self-returning walks in molecular graphs. Algorithm I modifies the adjacency matrix of a graph by a parameter accounting for the Kekule structure's Pauling bond orders of conjugated molecules. Algorithm II specifies a two-parameter function which is a weighted sum of the self-returning walks of different lengths. The two procedures are shown to improve the structure-property correlations for benzenoid hydrocarbons compared to the cases with non-weighted walks and with molecular connectivity indices. Five properties of benzenoids are studied: heats of atomization and formation, gas-chromatographic retention indices, *n*-octanol/water partition coefficients and melting points. The best result has been found for the heats of atomization (with statistical parameters $r = .9995$, $\sigma = 1.11$, $F = 9442$, $N = 12$).

1. INTRODUCTION

The search for graph-invariants that better match molecular structure and mirror molecular properties has led to the development of a series of powerful topological indices. Among them, the number of self-returning walks of fixed lengths has attracted increasing attention during the last decade. The reason for this growing popularity of the self-returning walks (SRWs) is due, in part, to their direct link with the quantum mechanical method of moments. As shown by Burdett *et al.*¹⁻⁷ and others,^{8,9} this link is of great importance in understanding the topological influences of molecular structure. The SRWs of different length in alkanes have been enumerated by Knop *et al.*¹⁰ up to $C_{16}H_{34}$. The SRW counts have been applied in specifying isomer comparability.¹¹

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Their relation to the Wiener index,¹² one of the better known topological indices, has been studied.¹³ Some correlations with molecular^{14,15} and atomic^{16,17} properties have also been reported. Graphs (called endospectral) with non-equivalent vertices which have the same number of SRWs (isocodal vertices) have been studied.^{10,16,18-21} Also, random walks of various types have been applied in statistical mechanics.²¹⁻²⁵

A link of SRWs with quantum mechanics is by way of the spectral moments of Hückel-type models. When using a convenient weighting of these walks, one simulates the different values of the interaction integrals between neighbouring atomic orbitals, thence arriving at an extended Hückel-type treatment.

The purpose of this paper is to introduce and examine two specific procedures for weighting the SRWs in molecular graphs with the hope to offer a way towards optimized structure-property correlations.

2. SELF-RETURNING-WALK GENERATING FUNCTIONS

Freedom of variation in the self-returning-walk (SRW) model may be elegantly gained through introduction of appropriate generating functions. First, we may weight each possible SRW in accordance with bond orders for the bonds traversed. For a graph G with at least one Kekule structure (perfect matching), the *Pauling bond order*²⁶ P_{ij} for a bond between sites i and j of G is commonly defined to be the ratio of the number of Kekule structures where this bond is double to the total number of Kekule structures. (In a more graph-theoretic language, P_{ij} is the probability that a perfect matching of G contains the edge $\{i,j\}$.) We define a *Pauling weight* $W_{\xi}(s)$ for a SRW ξ to be the product over factors $sP_{ij} + (1-s)$ for each step from a site i to j traversed in ξ . (Multiple crossings of a bond give powers of such factors.) Thence, we introduce a polynomial

$$\text{SRW}_n(s) = \sum_{\xi}^{(n)} W_{\xi}(s) \quad (2.1)$$

where the summation is over all walks of length n . As $s \rightarrow 0$, the value of this n th degree polynomial approaches the number of SRWs of length n . At $s = 1$, $\text{SRW}_n(s)$ again is an integer which breaks into factors each associated to a separate graphical component of the graph arising as the union of all perfect matchings of G .

A convenient computational formula for the polynomial of (2.1) is available. It is well-known (and fairly straightforwardly seen) that the number of SRWs of length n is simply the trace of the n^{th} power of the graph's adjacency matrix \mathbf{A} . Further, it is known²³ that the weighted count with W_{ij} for each step from i to j is given as the trace of the n^{th} power of the matrix \mathbf{W} . Thence, for the present case with \mathbf{P} the matrix of nearest neighbor *Pauling bond orders*, we identify

$$\mathbf{W} = s\mathbf{P} + (1-s)\mathbf{A} \quad (2.2)$$

so that

$$\text{SRW}_n = \text{Tr } \mathbf{W}^n \quad (2.3)$$

The indicated computation is further facilitated for *benzenoid* graph G for which²⁷

$$P_{ij} = \begin{cases} (A^{-1})_{ij} & , i \sim j \\ 0 & , \text{otherwise} \end{cases} \quad (2.4)$$

where $i \sim j$ indicates that i and j are nearest neighbors in G . This computational procedure is referred to in the following section as Algorithm I.

A further more general SRW generating function may be introduced

$$\text{SRW}(s, t) \equiv \sum_{n \geq 0} \text{SRW}_n(s) t^n \quad (2.5)$$

At $s = 0$, this reduces to the already well-known^{22,23} ordinary SRW generating function. In combination with (2.2), one has

$$\text{SRW}(s, t) \equiv \sum_{n \geq 0} \text{Tr } \mathbf{W}^n t^n = \sum_{n \geq 0} \sum_a \{ \lambda_a(s) \}^n t^n \quad (2.6)$$

where the $\lambda_a(s)$ are eigenvalues of \mathbf{W} . Thence, if t is less in magnitude than the inverse of the maximum eigenvalue of \mathbf{W} , one has

$$\text{SRW}(s, t) = \sum_a \{ 1 - t \lambda_a(s) \}^{-1} = \text{Tr} \{ \mathbf{I} - t \mathbf{W} \}^{-1} \quad (2.7)$$

This is referred to as Algorithm II.

Since the elements of \mathbf{W} monotonically decrease in magnitude as s increases from 0 to 1, so do the magnitudes of the maximum and minimum eigenvalues decrease, and thence (2.7) applies if t is less than the inverse of the maximum-magnitude eigenvalue of \mathbf{A} (this being the matrix \mathbf{W} at $s = 0$). For the generating function $\text{SRW}(s, t)$, the average length of the walks making significant contribution increases as t increases from 0, with the length diverging as t approaches the inverse of the maximum $\lambda_a(s)$. This average length (or number of steps) of a SRW is

$$\begin{aligned} \langle n \rangle &= \frac{\sum_{n \geq 0} n t^n \text{SRW}_n(s)}{\sum_{n \geq 0} t^n \text{SRW}_n(s)} = \frac{t \frac{\partial}{\partial t} \text{SRW}_n(s, t)}{\text{SRW}_n(s, t)} = \\ &= \frac{\text{Tr } t \mathbf{W} (\mathbf{I} - t \mathbf{W})^{-2}}{\text{Tr } (\mathbf{I} - t \mathbf{W})^{-1}} \end{aligned} \quad (2.8)$$

where the last equality gives the convenient computational form.

A further point of some interest is that the graph invariant $\text{SRW}(s, t)$ is »asymptotically additive«, in the sense that this invariant for a graph comprised of disjoint components is the sum of the corresponding invariants for the individual components. Such additivity has been argued²⁸ to correlate to a type of scaling behavior, which we associate with a variety of properties such as dealt with in the next section. For asymptotically »constantive«, »multiplicative« or »derivative« invariants, correlations to yet other sets of properties would be anticipated.

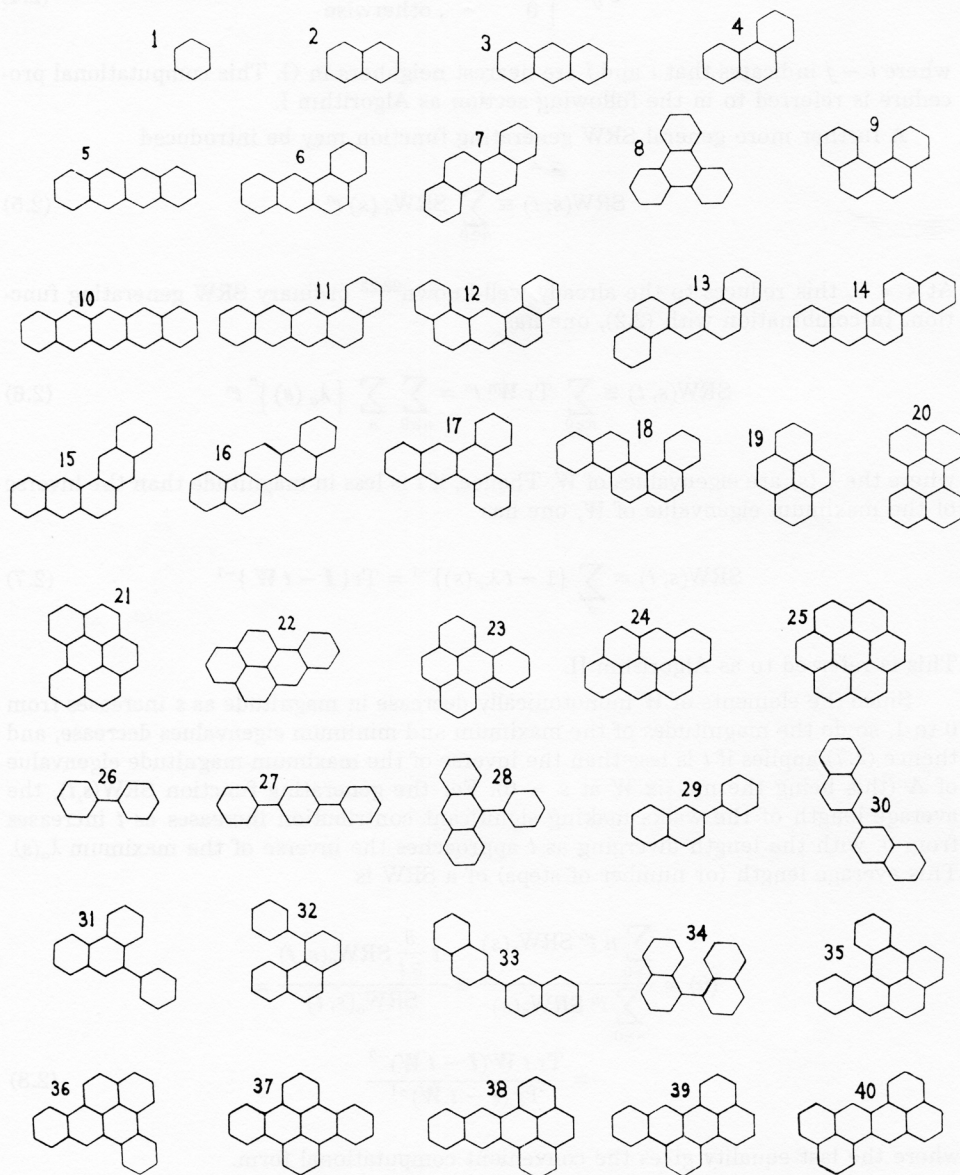


Figure 1. Molecular graphs of 40 benzenoid hydrocarbons whose properties are correlated with SRWs.

3. RESULTS

Correlation Samples

Unlike the numerous calculations, the experimental data for benzenoids are rather scarce. In Figure 1 we show 34 benzenoid hydrocarbons for which sufficient experimental data were found to provide statistically significant correlations. The data are shown in Table I and include 25 gas-chromatographic retention indices RI,³¹ 20 hydrophobicity constants $\log P$ (logarithm of the *n*-octanol/water partition coefficients),³¹ 12 heats of atomization ΔH_a ,³³ and 9 heats of formation ΔH_f .³⁴ The precision of some of the data may not have been sufficiently high, as reflected in the correlations obtained. Thus, nine of the melting points were given²⁹ within only one interval of 1–2 °C and the average of the interval had to be taken. On the other hand, the mean deviation of the $\log P$ data was estimated to 0.41.

TABLE I

Four molecular properties of the benzenoid hydrocarbons from Figure 1.

	$\frac{\Delta H_a}{\text{eV}}$	$\frac{\Delta H_f^o(g)}{\text{kcal mol}^{-1}}$	RI	$\log P$
1	57.16	20.0		2.13(0.10)
2	90.61	36.0	200.00	3.35(0.10)
3		54.9	301.69	4.50(0.15)
4	124.20	49.4	300.00	4.52(0.15)
5		68.6	408.30	5.76(0.30)
6	157.49	63.1	398.50	5.91(0.40)
7	157.73	63.9	400.00	5.86(0.40)
8	157.76	69.3	400.00	5.49(0.50)
9			391.39	—
10			486.81	7.19(0.70)
11				6.81(0.70)
12				—
13	191.24		495.45	6.75(0.40)
14			497.66	—
15				—
16			500.00	—
17	191.35		495.01	7.19(0.70)
18				
19	138.88		351.22	5.00(0.20)
20	172.04		456.22	6.25(0.50)
21	186.38		501.32	6.90(0.40)
22			453.44	6.35(0.40)
23			450.73	
24			503.89	
25	200.65			6.50(1.0)
26			233.96	3.98(0.10)
27				6.03(0.50)
28			396.38	6.04(0.50)
29			332.59	
30			405.38	
31			406.90	
32			421.61	
33			423.91	
34			388.38	

Correlations with Non-Weighted SRWs

In order to evaluate the significance of the weighting of the SRWs by means of the parameters s and t described in the preceding section, calculations were initially performed with non-weighted (integer) counts $SRW_n(0)$. The linear correlations thus obtained are given in Eqs. (3.1), (3.5), (3.9), (3.13) of Table II.

As seen, the SRWs of length 2 or 4 correlate highly ($r = 0.99$) with the heats of atomization, heats of formation, and gas-chromatographic retention indices, as well as with hydrophobicity constants ($r > 0.96$). The correlations with melting points were poor ($r = 0.64$ to 0.72) and so will not be reported in detail here. Presumably, the differences in the crystal structure of the compounds prevent the obtaining of a more pronounced dependence of melting points on the conjoined molecular structures, at least when characterized by our present topological indices.

Correlations with Pauling-Bond-Order Weighted SRWs

The potential importance of the Kekule structures for molecular properties under consideration was examined with Algorithm I via a computer program. A systematic search was performed for the optimal values of the step-weight s -parameter. The SRWs with lengths 2, 4 and 6 ($SRW_n(s) = 0$ for alternants with $n = \text{odd}$) were studied. With the exception of melting points, the properties studied are sensitive to the variations of the s -parameter. This is illustrated by Table III, where the ranges of the correlation coefficients r , standard deviation σ , and Fischer ratio F values are presented for the scale of s -parameter variation from $s = 0$ to 1.

The best correlations, as measured by r and obtained with optimized values of the s -parameter, are presented in Eqs. (3.2), (3.6), (3.10), (3.14) of Table II. The comparison of these equations and Eqs. (3.1), (3.5), (3.9), (3.13) shows that the accounting

TABLE II

Linear Correlations of the Four Benzenoid Properties with Non-Weighted and Weighted SRWs, as well as with Molecular Connectivities

			a	b	r	σ	F	N	$\langle n \rangle$	s	t
ΔH_a	3.1	SRW_n	23.142	3.1205	.9940	2.04	821	12	2	-	-
	3.2	$SRW_n(s)$	9.5294	10.377	.9987	1.39	3873	12	2	-	-
	3.3	$SRW(s, t)$	6.4950	5.3518	.9995	1.11	9442	12	1.5	0.92	0.71
	3.4	n_χ	8.4990	16.579	.9989	1.33	4618	12	1	-	-
ΔH_f	3.5	SRW_n	1.9456	1.5464	.9889	1.45	311	9	2	-	-
	3.6	$SRW_n(s)$	-2.4576	5.6103	.9936	1.27	539	9	2	0.84	-
	3.7	$SRW(s, t)$	-2.0872	3.3542	.9934	1.27	528	9	0.3	1.34	0.66
	3.8	n_χ	3.2389	8.012	.9918	1.35	422	9	2	-	-
RI	3.9	SRW_n	37.663	2.1780	.9880	3.40	1022	27	4	-	-
	3.10	$SRW_n(s)$	25.572	3.5258	.9883	3.40	1053	27	4	0.18	-
	3.11	$SRW(s, t)$	-0.7826	13.354	.9848	3.63	805	27	2.1	0.16	0.38
	3.12	n_χ	-47.141	48.959	.9696	4.31	393	27	1	-	-
$\log P$	3.13	SRW_n	1.1069	10.832	.9659	.567	521	20	2	-	-
	3.14	$SRW_n(s)$.3402	.4130	.9875	.443	707	20	6	0.86	-
	3.15	$SRW(s, t)$.2974	.1592	.9875	.442	708	20	2.6	0.98	-
	3.16	n_χ	.5348	.5774	.9783	.507	400	20	1	-	-

for the Kekule structures in the benzenoids under examination alters only marginally the statistics for retention indices. The other three properties, however, are mirrored considerably better. Their correlation coefficients rise up to 0.999, 0.994 and 0.988 for the heats of atomization and formation and the hydrophobicity constants, respectively. The standard deviations of these properties decrease while the Fischer ratios increase.

A further potential improvement of the correlations with the five benzenoid properties might be expected when using nonlinear functions. This possibility was partially studied by testing six other functions, including hyperbolic, power, and exponential functions. No single one of these provided better statistics, but the power function produced correlations very close to those given above, *e.g.*

$$\Delta H_a = 1.1427\{\text{SRW}_4(0.8)\}^{0.91311}$$

$$r = 0.9986, \sigma = 1.43, F = 3492$$

$$\Delta H_f^0(g) = 0.62595\{\text{SRW}_2(0.84)\}^{1.086}$$

$$r = 0.9933, \sigma = 1.28, F = 517$$

Inspection of the four best correlations (Eqs. (3.2), (3.6), (3.10), (3.14)) reveals also that weighted SRWs with length two prevail although those with lengths four and six can also be of importance. Examination of $\text{SRW}_n(s)/\log P$ and $\text{SRW}_n(s)/\text{RI}$ correlations for $n > 6$ resulted in worse statistics. Thus, it seems that only the SRWs of lengths 2, 4 and 6 are of importance for the properties under consideration. However, it can be studied in further detail by Algorithm II (section 2) using the additional parameter t .

Correlations with the $\text{SRW}(s,t)$ Functions

The calculations based on the averaged and weighted SRW function (Eq. 2.7) were performed with the weighting parameter s within the 0.0 to 2.0 range and $0 < t < 1/\lambda_{\max} < 1$. The $\text{SRW}(s,t)$ function clearly shows a specified optimum for the heats of atomization

TABLE III
Sensitivities upon variation in s

	n	r	σ	F
ΔH_a	2	.994 - .999	1.40 - 2.04	820 - 2495
	4	.985 - .999	1.40 - 2.57	320 - 3798
	6	.972 - .998	1.45 - 2.99	170 - 3187
$\Delta H_f^0(g)$	2	.989 - .993	1.27 - 1.45	311 - 533
	4	.987 - .993	1.31 - 1.52	260 - 471
	6	.986 - .988	1.49 - 1.54	246 - 283
RI	2	.934 - .985	3.6 - 5.2	172 - 816
	4	.914 - .988	3.5 - 5.6	126 - 1052
	6	.978 - .987	3.5 - 4.5	318 - 923
$\log P$	2	.966 - .983	.47 - .57	250 - 510
	4	.945 - .985	.46 - .64	150 - 590
	6	.917 - .986	.45 - .70	95 - 626

and hydrophobicity constant. For the other three properties there is a wide range of optimal (s,t) values providing nearly the same best statistics. Thus, for the heats of formation, the maximal correlation coefficient $r = r_{\max} = 0.9934$, the minimal standard deviation $\sigma = \sigma_{\min} = 1.27$ and the Fisher ratio F in the range from 526.4 to 527.7 are all realized by many (s,t) -pairs: (1.30, 0.68), (1.32, 0.67), (1.34, 0.66), (1.36, 0.65), (1.38, 0.65), (1.40, 0.64), (1.42, 0.63), (1.44, 0.62), 1.46, 0.61 and (1.48, 0.60). Similarly, the optimal range of (s,t) -values providing the highest correlation ($r_{\max} = 0.9848$) with gas-chromatographic retention indices is from $(s = 0.02, t = 0.34)$ to $(s = 0.26, t = 0.41)$. This might be regarded as an indication of a possible interrelation between the two parameters.

The four examined properties have been best reproduced by equations (3.3), (3.7), (3.11), and (3.15) in Table II.

In comparing the correlations produced by the SRW (s,t) function with those obtained for SRW $_n(s)$ (Eqs. (3.2), (3.6), (3.10), and (3.14), respectively) one may firstly mention the significant improvement of the results for the heats of atomization. The correlation coefficient increased from 0.9887 to 0.9995, the standard deviation decreased from 1.39 to 1.11 and the Fischer ratio jumped from 3873 to 9442. Evidently, SRW (s,t) produces the best linear correlation with the energy of atomization as compared with any parameters examined here for this purpose.

The other molecular properties under examination do not show such improved correlation with SRW (s,t) . Equation for $\log P$ and $\Delta H_f^0(g)$ have practically the same statistics with Eqs. (3.14) and (3.6): $r=0.9875, 0.9875$ and $r = 0.9936, 0.9934$, respectively. Surprisingly enough, the RI/SRW correlation is not only worse than that with RI/SRW (s,t) but even worse than the correlation with the unweighed SRWs ($r = 0.9848 < 0.9883, 0.9880$).

Some note may be paid to the non-integer n values in Table II in the case of the SRW (s,t) generating functions. These are the mean values $\langle n \rangle$ of the length of SRWs as calculated by Eq. (2.8). As seen, $\langle n \rangle \approx 0.3$ for heats of formation where also the optimal value of s is unusually large. The optimal equation (3.3) for the heats of atomization still corresponds to a $\langle n \rangle$ value less than two ($\langle n \rangle \approx 1.5$) while for retention indices and $\log P$, the average length of the SRWs is within the 2 to 3 range. Thus, once again, as in the case of optimized SRW $_n(s)$ functions (Eqs. (3.2), (3.6), (3.10), and (3.14), the higher lengths of SRWs play no significant role.

Resuming the comparison of the results obtained by Algorithm I and Algorithm II, we may list the best equations found for calculating the examined properties of benzenoid hydrocarbons: Eq. (3.3) (heats of atomization), Eq. (3.6) (heats of formation), Eq. (3.10) (gas-chromatographic retention indices), and Eq. (3.15) (logarithm of the octanol/water partition coefficient).

Comparison with Molecular Connectivity Calculations

Having obtained a number of high structure-property dependencies by virtue of the weighted SRWs (Algorithm I and Algorithm II), a question may arise: to what extent is this a novel result? or, otherwise, are not these results a consequence of the connection between SRWs and molecular connectivity?^{34,35} In fact, it is well known that the number of non-weighted SRWs of two steps through a vertex i in molecular graph G equals the vertex degree a_i , which is a basic component in calculating the first-order molecular connectivity index $\chi = \sum_{i=j} (a_i a_j)^{-1/2}$ where the summation is taken

over all bonds. Hence, although some correlation between the total number of SRWs of length $2k$ and the k^{th} order molecular connectivity might be expected, the two graph invariants are not the same, and any correlation might even be worse when weighted SRWs are used. Yet, for a more explicit test of the independency of the result obtained, we performed comparative correlation studies with molecular connectivity indices of order 0 through 4, as well as with³⁶ cluster and path-cluster connectivity indices. The equations with the best statistics are also shown in Table II (Eqs. (3.4), (3.8), (3.12), (3.16)). Albeit a high correlation is obtained for the four benzenoid properties, the statistics are somewhat poorer than that of the best equations with weighted SRWs. More specifically, for the heats of atomization and formation, retention indices, and the hydrophobicity constant of the benzenoids under examination, the use of weighted SRWs improved the statistics as follows: correlation coefficient: 0.9989 \rightarrow 0.9995, 0.9918 \rightarrow 0.9936, 0.9696 \rightarrow 0.9883, and 0.9783 \rightarrow 0.9875, respectively; standard deviation: 1.33 \rightarrow 1.11, 1.35 \rightarrow 1.27, 4.3 \rightarrow 3.4, and 0.507 \rightarrow 0.442, respectively; Fischer ratio: 4618 \rightarrow 9442, 422 \rightarrow 539, 393 \rightarrow 1053, and 400 \rightarrow 708, respectively.

4. CONCLUSION

Novel types of graph invariants based on weighted self-returning walks have been introduced and found to correlate very well with certain properties of our data set of benzenoids. Particularly well correlated were the heat of atomization and chromatographic retention index, to a lesser extent, the heat of formation and octanol/water partition coefficient. Neither the new invariants nor the others compared to them correlate well with the melting points. The new self-returning walk invariants often improve (though often just slightly and sometimes at the »expense« of an additional parameter) over the edge count (SRW₂) and may also be of use in other QSAR/QSPR studies.

REFERENCES

1. J. Burdett, S. Lee, and W. C. Sha, *Croat. Chem. Acta* **57** (1984) 1193.
2. J. K. Burdett and S. Lee, *J. Amer. Chem. Soc.* **107** (1985) 3050; 3063.
3. J. K. Burdett, in *Graph Theory and Topology in Chemistry*, R. B. King, D. H. Rouvray Eds., Elsevier, Amsterdam 1987, p. 302.
4. J. K. Burdett, S. Lee, and W. C. Sha, *Nouv. J. Chim.* **90** (1985) 757.
5. J. K. Burdett, *Acc. Chem. Res.* **21** (1988) 189.
6. J. K. Burdett, *Struct. & Bonding (Berlin)* **65** (1987) 10.
7. J. K. Burdett, *Chem. Rev.* **88** (1988) 1.
8. Y. Jiang, A. Tang, and R. Hoffmann, *Theor. Chem. Acta* **66** (1984) 183.
9. Y. Jiang, H. Zhu, H. Thang, and I. Gurman, *Chem. Phys. Lett.* **159** (1989)
10. J. V. Knop, W. R. Müller, K. Szymanski, M. Randić, and N. Trinajstić, *Croat. Chem. Acta* **56** (1983) 405.
11. D. Bonchev, V. Kamenska, and O. Mekenyan, *J. Math. Chem.* **5** (1990) 43.
12. H. Wiener, *J. Amer. Chem. Soc.* **69** (1947) 17; *J. Chem. Phys.* **15** (1947) 766.
13. D. Bonchev, O. Mekenyan, and O. E. Polansky, Ref. [3], p. 209.
14. M. Barysh, J. V. Knop, S. Pejaković, and N. Trinajstić, *Polish J. Chem.* **53** (1985) 405.
15. M. Barysh, D. Bonchev, and O. Mekenyan, *MATCH* **20** (1986) 125.
16. D. Bonchev, L. B. Kier, and O. Mekenyan, *Int. J. Quant. Chem.* (in press).
17. D. Bonchev and L. B. Kier, *J. Math. Chem.* **9** (1992) 75.
18. M. Randić, *J. Comput. Chem.* **1** (1980) 386.
19. M. Randić, W. L. Woodworth, and A. Graovac, *Int. J. Quant. Chem.* **24** (1983) 435.
20. Y. Jiang, *Sci. Sinica, Ser. B* **27** (1984) 236.

21. M. Randić and A. F. Kleiner, *Ann. N.Y. Acad. Sci.* **555** (1988) 320.
22. E. W. Montroll, *Amer. Math. Soc. 16th Symp. Appl. Math.* (1964) 193.
23. P. W. Kasteleyn, in *Graph Theory and Theoretical Physics*, F. Harary Ed., Academic Press, New York 1967. p. 43.
24. G. H. Weiss and R. J. Rubin, in *Advances in Chemical Physics* **52** (1983) 363.
25. E. W. Montroll and M. F. Schlesinger, in *Nonequilibrium Phenomena II* J. L. Lebowitz, E. W. Montroll, Eds., Elsevier, Amsterdam 1984, p. 2.
26. See, e.g. L. Pauling *The Nature of the Chemical Bond*, 3rd Ed. Cornell University Press, Ithaca, New York 1960.
27. N. S. Ham, *J. Chem. Phys.* **29** (1959) 1229.
28. D. J. Klein, *Int. J. Quant. Chem.* **S20** (1986) 153.
29. J. R. Dias, *Handbook of Polycyclic Hydrocarbons, Part A, Benzenoid Hydrocarbons*, Elsevier, Amsterdam 1987.
30. *Spectral Atlas of Polycyclic Aromatic Compounds*, W. Karcher, R. J. Fordham, J. J. Dubois, P. G. J. Glaude, J. A. M. Lighthart, Eds., Reidel. Dordrecht 1985.
31. D. J. Futoma, S. R. Smith, T. E. Smith, and J. Tanaka, *Polycyclic Hydrocarbons in Water Systems*, CRC Press, Boca Raton FL. 1981.
32. J. Sangster, *J. Phys. Chem. Ref. Data* **18** (1988) 1125.
33. M. Kh. Karapet'yantz and L. Karapet'yantz, *Thermodynamic Constants of Inorganic and Organic Compounds*, Humphrey Science Pub., Ann Arbor 1970.
34. W. C. Herndon, D. A. Connor, and P. Lin, *Structure-Enthalpy Relationships in Polycyclic Cata-Condensed Aromatic Hydrocarbons*, University of Texas at El Paso, El Paso 1989.
35. M. Randić, *J. Amer. Chem. Soc.* **97** (1975) 6609.
36. L. B. Kier and L. H. Hall, *Molecular Connectivity in Structure-Activity Analysis*, Research Studies Press, Chichester, U.K., 1986.

SAŽETAK

Vagane povratne šetnje za korelaciju strukture i svojstava

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Razvijena su dva algoritma za vaganje povratnih šetnji u molekularnim grafovima. Algoritam I modificira matricu susjedstva grafa parametrom zasnovanim na Paulingovu redu veza konjugirane molekule. Algoritam II koristi se dvo-parametarskom funkcijom koja predstavlja vaganu sumu povratnih šetnji različitih duljina. Pokazano je da te dvije procedure poboljšavaju korelaciju između strukture i svojstava benzenoidnih ugljikovodika u usporedbi s vaganim šetnjama i indeksima molekularske povezanosti. Proučavano je pet svojstava benzenoida: topline atomizacije i nastajanja, plinsko-kromatografski retencijski indeksi, particijski koeficijenti *n*-oktanol/voda i tališta. Najbolji rezultati dobiveni su za topline atomizacije (sa statističkim parametrima $r = .9995$, $\sigma = 1.11$, $F = 9442$, $N = 121$).