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## On Additivity of Heats of Atomization of Benzenoid Hydrocarbons

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An additivity scheme, based on Clar's formulae, is proposed for calculating heats of atomization of benzenoid hydrocarbons. Comparison between the results obtained by this scheme, by the semiempirical SCF MO model, and experimental findings is good. This satisfactory agreement, thus, presents independent support for Clar's notion of isolated  $\pi$  sextets and migrating sextets.

### INTRODUCTION

Several papers have described semi-empirical MO treatments of conjugated systems which allow heats of atomization,  $\Delta H_a$  to be calculated with appreciable accuracy<sup>1-6</sup>. The results of these calculations agreed with experimental values within a few tenths of eV, with the exception of strained molecules where the observed heats of atomization were smaller than those calculated; the difference being attributed to the ring strain. Although sufficiently correct values were obtained, it remained unclear if, and how, the individual results for structurally similar systems are related. In particular the question of possible additivity of the contributions associated with the constituting fragments could not be recognized in the treatment which considered each molecule individually. Here we describe a procedure which reduces the evaluation of heats of atomization of conjugated hydrocarbons to a summation of a few distinctive contributions to be associated with fragments such as benzene and naphthalene carbon skeletons joining C—C single bonds and isolated C=C double bonds and finally the contributions from all the C—H bonds present. This approach differs from a number of other earlier considerations of the additivity of heats of atomization of conjugated systems<sup>7-11</sup> in one important aspect. Earlier works aimed at the full fragmentation of a molecule into constituting bonds and were then concerned with the search for an adequate number of parameters to characterize the expressions which will yield  $\Delta H_a$ . To obtain more satisfactory results such schemes required additional parameters to characterize details of environmental variations. In contrast we propose here a partial fragmentation of a system, not necessarily breaking it up completely, but retaining as fragments isolated benzene and naphthalene skeletons. The remaining bonds, which comprise C—C bonds joining such fragments, additional C=C isolated bonds, and the totality of C—H bonds add to the total heat of atomization. We, thus, have

all *five* parameters in an empirical scheme which determines the additivity scheme, which as will be seen later provides quite accurate heats of atomization of conjugated hydrocarbons. This approach can be used for predictive purposes with considerable reliability for molecules in which there is marked alternation of bond lengths.

The approach is essentially based on consideration of only selected Kekulé-type formal structural formulae of benzenoid hydrocarbons assumed to represent molecules adequately. The particular structures selected are those which upon superposition produce the structural formulae introduced by Clar<sup>12,13</sup>, in which one has isolated pi-electron sextets, isolated C=C double bonds and possibly two or more such structures which upon superposition point to migration of pi-electron sextets. The latter, in fact, stand for isolated formal naphthalene pi-electrons, and also higher  $(4m + 2)$  linear acene systems. Clar's structural formulae have their empirical justification in accumulated data from NMR and UV spectra<sup>14</sup>, some aspects of which can consistently be interpreted as manifestation of local aromatic properties of such systems. This means that not all rings in benzenoid systems are equivalent in their aromatic character which Clar symbolized with isolated pi-electron sextets. The qualitative arguments of Clar have found recently quantitative formulations<sup>15</sup>. The relative magnitudes of the local aromaticity, more appropriately called benzenoidity<sup>16</sup>, as one tries to characterize departure of a particular pi-ring from the properties of an isolated benzene ring, can be described through an extension of the concept of Kekulé-indices recently introduced for conjugated hydrocarbons<sup>17,18</sup>, or through an extension of the concept of Pauling bond order<sup>19</sup> from individual bonds to individual six-membered rings<sup>20</sup>. The results of this work provide yet further quite independent support for the concept of the aromatic sextet of Clar.

The deficiency of the earlier approaches to detect the additivity of heats of atomization in the results of MO calculations is mainly due to attempts to fragment all the bonds in a molecule. For classical polyenes which are represented by a single formula and for which the supposition of localized bonds seems natural such a complete fragmentation gives satisfactory interpretation of the heat of atomization in terms of energies of C—C, C=C, and C—H bonds. No satisfactory simple results could be obtained for benzenoid hydrocarbons if such a fragmentation is made. However, if similar considerations are made on individual Kekulé formal structures of conjugated benzenoid systems, one can, under certain conditions, obtain acceptable values for the heats of atomization. The approach requires retaining the benzene C<sub>6</sub> skeleton as a unit, not to be fragmented, and similarly higher  $(4m + 2)$  isolated linear groupings should not be fragmented into the constituting C—C and C=C bonds. There appears to be a constant increment in the heats of atomization associated with the carbon skeleton going from benzene to naphthalene, or from naphthalene to anthracene, or from anthracene to tetracene, etc. However, since the correlation is a *linear* one, not a proportionality, there is no appropriate value to be associated with a single aromatic CC bond. In this work we adopted the set of parameters established by Dewar and Gleicher<sup>21</sup> associated with the contributions of C—H, isolated C=C and joining C—C bonds. The parameters used were as follows:  $E(\text{C—C bond}) = 4.3499$  eV,  $E(\text{C=C bond}) = 5.5378$  eV, and  $E(\text{C—H bond}) = 4.4375$  eV. The value for the heat of atomization of benzene carbon skeleton, has been taken as 30.535 eV. This value is derived

from the experimental value for the heat of atomization of benzene<sup>22</sup> 57.16 eV from which the contribution of six C—H bonds has been subtracted. Similarly from  $-\Delta H_a$  (naphthalene) = 90.61 eV<sup>23</sup>, by subtracting C—H bond contributions, one obtains for the naphthalene  $C_{10}$  skeletal part a contribution of 55.11 eV. The difference between the skeletal contributions of naphthalene and benzene, 24.575 eV indicates the additional stabilization which can be associated with the pi-electron sextet migration. Extending these considerations to higher linear acenes one observes that the increment in the skeletal part of the heat of atomization is approximately constant. The mean value of the increment for the first four members in the series (for which experimental data are available<sup>22-24</sup>) is 24.592 eV. Thus, from the value for the skeletal contribution to the heat of atomization in benzene one obtains a contribution of 5.089 eV per aromatic CC bond, but the increment on addition of each  $C_4$  fragment producing an additional five aromatic CC bonds gives only 4.913 eV per bond. The bonds in an isolated benzene skeleton are more stabilized by  $\sim 0.1$  eV and the difference is sufficient to cause departure from the simple additivity which is based on the complete fragmentation of a benzenoid hydrocarbon system to contributions of individual bonds.

In our scheme, which retains the contributions of benzene and naphthalene skeletal fragments, the heat of atomization of benzenoid hydrocarbons can be written in the form,

$$-\Delta H_a = n_{CH} E_{CH} + n_{C-C} E_{C-C} + n_{C=C} E_{C=C} + \sum E_N \quad (1)$$

where  $n_{CH}$  is the number of CH bonds of bond energy  $E_{CH}$ ,  $n_{C-C}$  is the number of formally single C—C bonds in Clar's structural formula, joining isolated benzene or higher linear acene rings, of energy  $E_{C-C}$ ,  $n_{C=C}$  is the number of isolated formal C=C double bonds of energy  $E_{C=C}$ , and  $E_N$  is the bonding part of the energy of the CC skeleton of fragment N which can be a  $(4m + 2)$  benzenoid linear system. The summation goes over all the contributing units.  $E_N$  can, in fact, be given as,

$$E_N = E_1 + N \Delta E \quad (2)$$

in view of the constancy of the increment in the skeletal part of the heat of atomization when the number of rings (N) changes in linear polyacenes.  $E_1$  is the energy of the CC skeleton of benzene, while  $\Delta E$  is the increment.

## RESULTS

Although we investigate here conjugated systems which typify delocalized systems, we will show that if one considers only selected Kekulé-type structural formulae, which symbolize localised distribution of C—C and C=C bonds, if taken isolated, satisfactory values for the heats of atomization can be obtained. Thus, the additivity of heats of atomization found in classical polyenes<sup>1</sup> can be extended to conjugated systems, providing one recognizes the specificity of benzene and other linear polyacenes. We obtain heats of atomization from selected Kekulé-type formulae and the crucial problem is the selection of important classical valence bond structures for this purpose. One should realize that superposition of a few Kekulé-type structures leads to representation of a system in which some molecular regions may be delocalized while others

still remain described by isolated C=C and C—C bonds. This is illustrated for the case of chrysene (Figure 1). There are eight distinct Kekulé-type structures

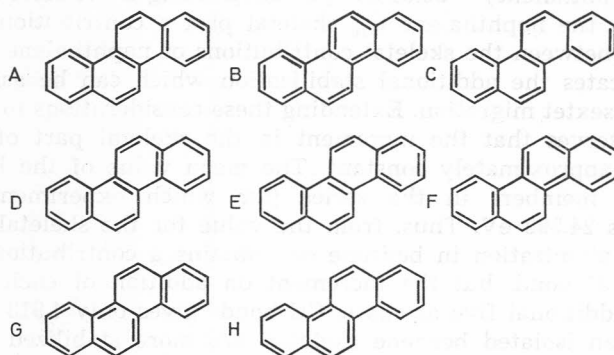


Figure 1. Kekulé structures of chrysene.

of chrysene, indicated by letters A—H. When two structures at a time are superimposed from 28 possible combinations (see Figure 2) several result in an identical superposition. The resulting structures have delocalized regions, indicated by a formal benzene ring, a naphthalene perimeter, a phenanthrene perimeter, and finally, a chrysene perimeter. Among these structures only three structures have *two* isolated benzene rings, which in this case represent stru-

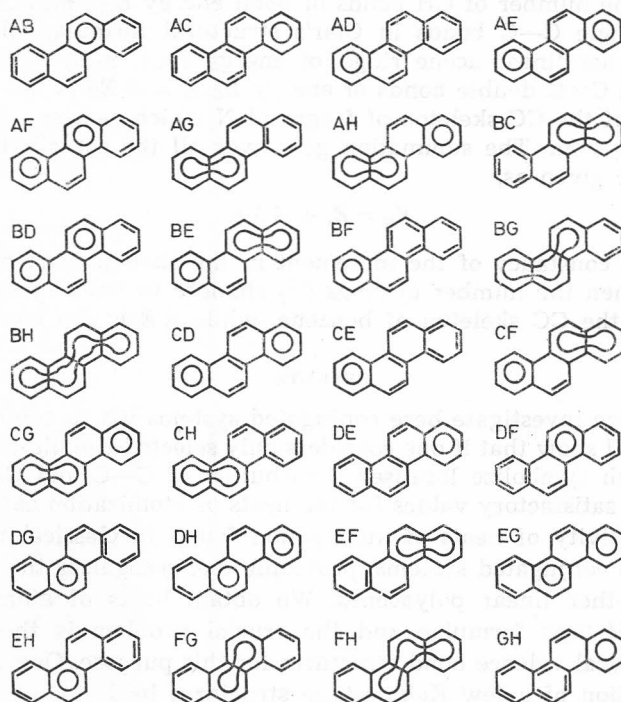


Figure 2. Combinations of Kekulé structures of chrysene.



tures with the maximal number of benzene sextets. According to Clar<sup>13,25</sup>, these structures are adequate for discussion of the experimental properties of benzenoid systems and are retained in our approach to the problem of evaluation of heats of atomization and their additivity. The basis of this treatment is, therefore, essentially empirical and the validity of it depends on the quality of the consequences. However, in addition we have constrained the selected parameters to semi-empirical values. Thus, at the same time we show not only that the additivity of heats of atomization among the benzenoid systems is valid, but also that this is achieved with parameters characterizing the classical structures.

We represent chrysenes as a superposition of Clar-type structural formulae I—III (Figure 3). A superposition of structures (I) and (II), for instance, intro-

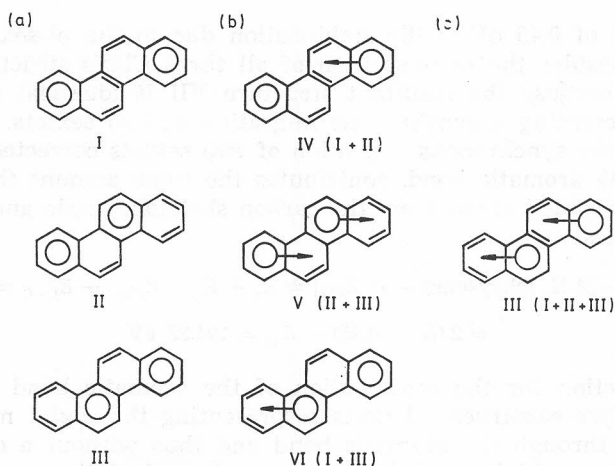


Figure 3. Clar-type structural formulae of chrysenes (a) and their superpositions (b and c).

duces a migration of one of the pi-sextets and it can be represented by a formula with isolated benzene and naphthalene fragments (IV). Similarly, the superposition of (I) and (III) produces another such structure (VI) with a naphthalene fragment, while the superposition of (II) and (III) leads to synchronous migration of *two* pi-electron sextets producing (V). The contribution to the heats of atomization of an individual Clar's structure involving the isolated benzene moiety and C=C bonds is evaluated by adding the contributions to the heat of atomization of the constituent parts. A superposition of *two* such structures which results in an isolated naphthalene segment (e. g., superposition of structures (I) and (II) in chrysenes) can be similarly evaluated by adding the contributions of the individual parts, where additional stabilization is accounted for through the inclusion of the increment in the skeletal part of the molecule, going from benzene to the naphthalene skeleton. The net gain, i. e. the difference between the heat of atomization associated with the naphthalene C<sub>10</sub> skeleton and the Clar single formula for the naphthalene skeleton, which is 0.488 eV (approximately 0.5 eV), signifies the additional stabilization produced by the pi-sextet migration. A superposition of *three* (or more) Clar's structures should result in an additional contribution to the heat of atomization, but the

precise characterization of such a contribution introduces some difficulties, as will be discussed later.

The calculation of the heats of atomization is illustrated for chrysene. There are three Clar's structural formulae for chrysene (Figure 3). The existence of several formulae indicates that each of the isolated  $\pi$ -electron sextets is delocalized. The following relations represent the contributions to the heat of atomization of hypothetical structures given by (a) a *single* Clar's formula,

$$-\Delta H_a(\text{chrysene}) = 12 E_{\text{C-H}} + 6 E_{\text{C-C}} + 3 E_{\text{C=C}} + 2 E_1 = 157.03 \text{ eV}$$

and (b) a superposition of *two* Clar's formulae,

$$-\Delta H_a(\text{chrysene}) = 12 E_{\text{C-H}} + 3 E_{\text{C-C}} + E_{\text{C=C}} + E_1 + E_2 = 157.48 \text{ eV}$$

The increment of 0.45 eV is the stabilization due to the pi-sextet migration. We did not consider the superposition of all *three* Clar's structural formulae for chrysene, because the resultant structure VII is identical to V, i. e. the structure representing a synchronous migration of two sextets. It appears in this case that the synchronous migration of two sextets corrected for the contribution of the aromatic bond, contributes the same amount that one sextet migration plus contributions from the carbon skeleton, single and double bond fragments makes,

$$\begin{aligned} -[\Delta H_a(\text{chrysene}) - 12 E_{\text{CH}}] &= E_1 + E_2 + E_{\text{C=C}} + E_{\text{C-C}} = \\ &= 2(E_2 - 0.45) - E_{\text{Ar}} = 104.23 \text{ eV} \end{aligned}$$

The correction for the contribution of the aromatic bond is due to the fact that the two substructural units representing the sextet migration in V are annelated through the aromatic bond and thus without a correction this bond fragment would be included twice in the calculation.

The experimental heat of atomization of chrysene is 157.73 eV<sup>26</sup>, while the SCF MO calculating produced a value (157.77 eV) very close to the measured one.

Table I shows heats of atomizations calculated for several benzenoid hydrocarbons together with available experimental values and theoretical values based on SCF MO calculations.

The agreement between the calculated values obtained by the simple superposition of additive contributions of C=C, C—C, C—H, C<sub>6</sub> and C<sub>10</sub> fragments, and the values obtained by the more elaborate semiempirical MO methods is excellent. We have not tried to alter the parametrization and the only additional parameters are the *experimental* heats of atomization of benzene and naphthalene. In the most cases studied the agreement between the theoretical results and experimental findings is good. In some instances the difference between the theoretical and experimental results is rather large (e. g. pyrene). Here one may suspect that perhaps the choice of the Kekulé and Clar's structures is not quite satisfactory or is not unique and that the additional factors which may influence the molecular properties of such systems have to be considered such as, for example, the overcrowdedness (non-bonded hydrogen interactions) or the ring strain.

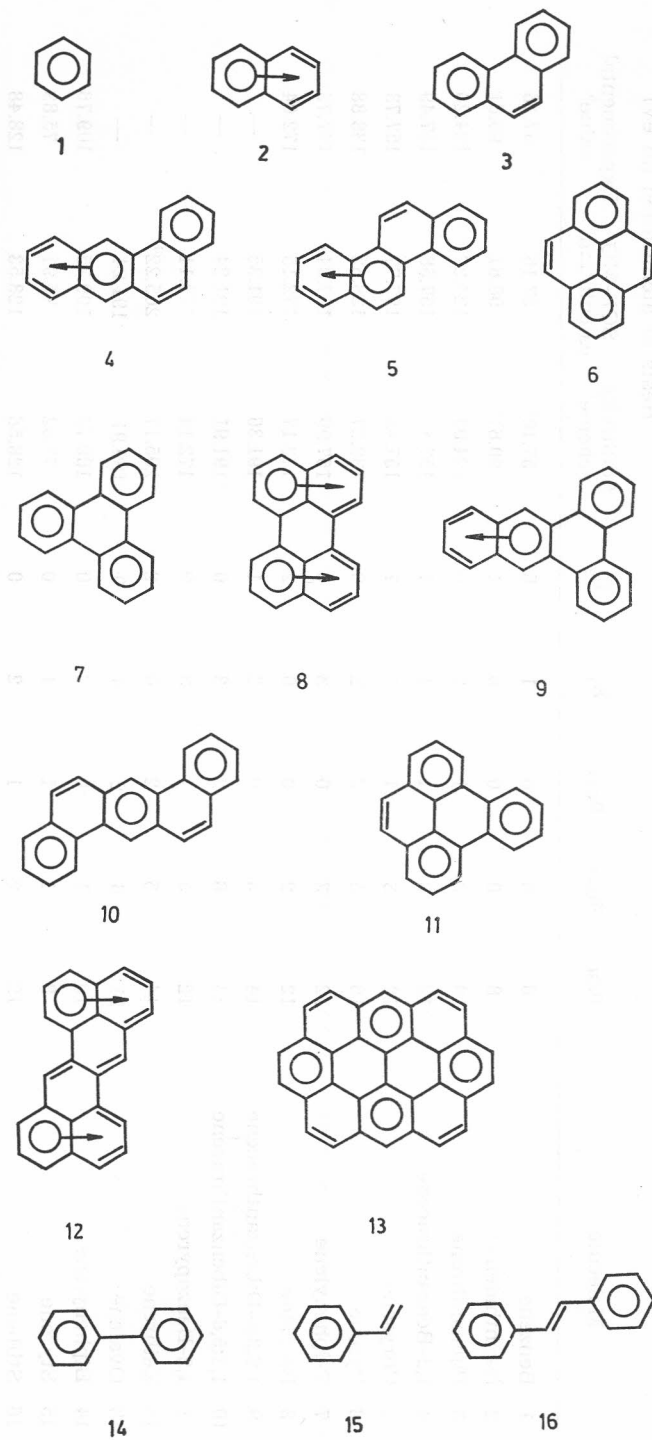


Figure 4. Clar's formulae of studied molecules in Table I.

TABLE I  
 Calculated Heats of Atomization for Several Benzenoid Hydrocarbons Using the Additivity Scheme

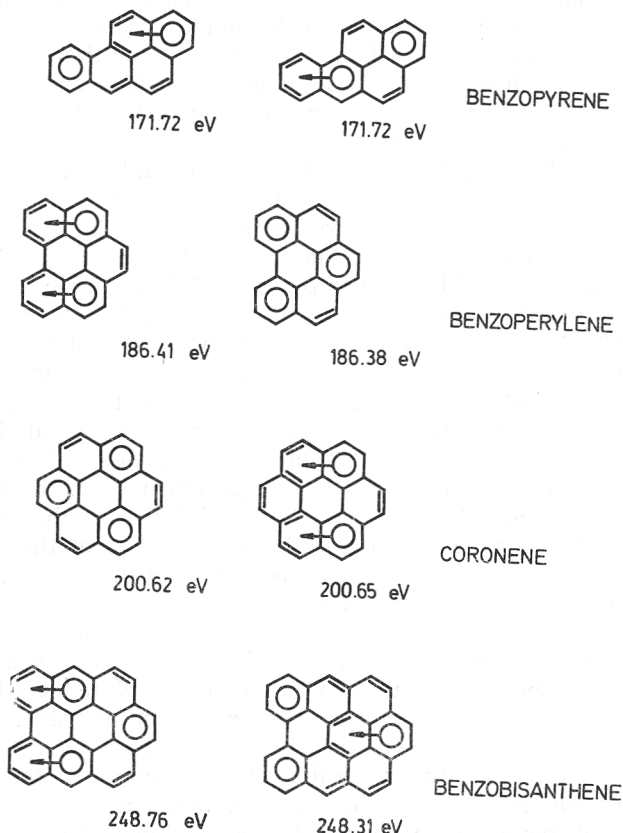
Molecule <sup>a</sup>	$n_{\text{CH}}$	$n_{\text{C-C}}$	$n_{\text{C=C}}$	$E_1$	$E_2$	Heats of atomization (in eV)		
						Additivity scheme	SCF-MO calculation <sup>b</sup>	Experimental value <sup>b</sup>
1 Benzene	6	0	0	1	0	57.10 <sup>c</sup>	57.16	57.16
2 Naphthalene	8	0	0	0	1	90.61 <sup>c</sup>	90.61	90.61
3 Phenanthrene	10	3	1	2	0	124.03	124.22	124.20
4 1,2-Benzanthracene	12	3	1	1	1	157.48	157.58	157.49
5 Chrysene	12	3	1	1	1	157.48	157.77	157.73
6 Pyrene	10	5	2	2	0	138.27	138.62	138.88
7 Triphenylene	12	3	0	3	0	157.90	157.94	157.76
8 Perylene	12	2	0	0	2	172.17	172.15	172.04
9 1,2,3,4-Dibenzanthracene	14	3	0	2	1	191.36	191.35	—
10 1,2,5,6-Dibenzanthracene	14	6	2	3	0	191.91	191.24	—
11 1,2-Benzopyrene	12	5	1	3	0	172.14	172.11	—
12 Zethrene	14	5	2	0	2	205.17	205.22 <sup>d</sup>	—
13 Ovalene	14	6	2	3	0	190.91	191.24	—
14 Biphenylene	10	1	0	2	0	109.79	109.75	109.76
15 Styrene	8	1	1	1	0	75.92	75.91	75.83
16 Stilbene	12	2	1	2	0	128.56	128.53	128.48

<sup>a</sup> Clar's formulae are given in Figure 4. <sup>b</sup> Ref. 1. (Table 5.2, pp. 170-171). <sup>c</sup> Used in determining the empirical parameters. <sup>d</sup> Ref. 2.

In Table II we list the results for several benzenoid hydrocarbons which may be represented by non-equivalent Clar's structures and for which Clar's postulate of the maximal number of isolated benzene-like rings does not necessarily define the system uniquely. In several instances, in fact in the original work of Clar, there is an indication that additional structures may contribute to the description of the system. The idea of superimposing two, not even necessarily equivalent, Clar's structures is implied in the definition of migrating sextets. What has not been fully characterized is a superposition of several structures with isolated benzene sextets and the quantitative differentiation of such a situation from simpler cases when the resulting structure is obtained from a superposition of *two* structural formulae of Clar (e.g., naphthalene). Examples in Table II will illuminate this point to some extent.

TABLE II

*Examples of Non-equivalent Clar's Formulae for the Same Conjugated Hydrocarbon*



In benzopyrene, for instance, there are only two (non-equivalent) Clar's formulae. One involves a migrating sextet within the pyrene part of the molecule and the other involves an additional benzene peripheral ring. Each of these structures is the result of a superposition of two Clar's formulae involving isolated benzene fragments, so that in fact there would be initially four Clar's formulae, which are then contracted to two structures involving naphthalene

fragments. Both structures are equivalent from the count of fragments, thus both lead to the same value for the heat of atomization.

$$-\Delta H_a(\text{benzopyrene}) = 12 E_{\text{CH}} + 2 E_{\text{C=C}} + 5 E_{\text{C-C}} + E_1 + E_2 = 171.72 \text{ eV}$$

The actual experimental value can be expected to be larger, however, it is not available for comparison. Dewar and de Llano<sup>3</sup> calculated it to be 172.38 eV some 0.66 eV higher. But, an additional superposition of the two structures should, as in the case of chrysene, produce another increase of 0.45 eV in the heat of atomization, improving the agreement between the two approaches. The parallelism between chrysene and benzopyrene is not however complete. It is true that in both cases we have Clar's structures in each step of successive superposition, but in one case the structures are equivalent and in the other the superimposed structures are not equivalent. The observed differences might be related to this additional structural features. Such discussions however would be outside the scope of the present work, and are somewhat speculative in view of the lack of experimental data to support one or the other value calculated by different methods. In the case of benzoperylene and benzobisanthene there is some ambiguity since one can construct alternative structures with the isolated naphthalene fragments rather than with isolated benzene units and it is not immediately clear which approach should be followed. In benzoperylene one of the two structures has the maximal number of formally isolated benzene rings and should, according to Clar's postulate, be the dominant structure. However, the other structure has a smaller number of isolated C=C bonds and may be suspected to be of importance. When the heats of atomization are evaluated a slight preference is given to the latter structure having an increase of 0.03 eV in the heat of atomization. This situation indicates the necessity to characterize more fully Clar's postulate, regarding structures with the maximal number of isolated  $\pi$ -electron sextets. If the sextets can migrate a choice has to be made between the large number of isolated sextets or a lesser number of migrating sextets. The principle that determines the structures in such cases may be supplemented with a requirement that the representative structure has the minimal number of isolated C=C double bonds, as the fixation of C=C bonds means no delocalization and loss of the extra stability of non-classical structure systems. For instance, if benzene is assumed to be a collection of C=C, C-C, and C-H bonds, the calculated heat of atomization is lower by 0.889 eV than the experimental value indicates. Similarly, when naphthalene is represented by a *single* Clar's symbol, i. e., by a benzene ring skeleton, two C=C bonds and three C-C bonds (and ten C-H bonds), the heat of atomization is again lower than the experimental value, the difference being half of that found for benzene when represented by a single classical structure. The difference is therefore due to stabilization of the system when  $\pi$ -electron migration is allowed. Thus, a superposition (resonance) of two or more Clar's structures gives an appreciable correction to heats of atomization. The results for coronene and benzobisanthene are consistent with such an interpretation. In benzobisanthene the considered structures differ by having either two migrating  $\pi$ -sextets and  $\pi$ -sextet and naphthalene portions. The latter structure has a larger heat of atomization, again in agreement with its having fewer isolated C=C bonds. Coronene is another interesting example. There are two equivalent Clar's structures. The Clar's structure with three



benzene rings and three double bonds has a heat of atomization  $-\Delta H_a = 200.62$  eV. The Dewar-de Llano value<sup>3</sup> is higher by 0.90 eV ( $-\Delta H_a$  (Dewar-de Llano) = 201.53 eV). The difference of 0.90 eV comes from the resonance between two Clar's structures in the same way that two benzene classical structures contribute 0.889 eV to the heat of atomization of benzene. In fact the similarity of the numerical magnitudes is not fully coincidental, as in both cases we have a superposition of two structures with three C=C bonds, although in the case of coronene the situation is more complex involving other formal benzene rings. We may, however, attribute 0.90 eV to the additional stabilization associated with what Clar termed "superaromaticity", that is to an additional aromatic component in a system. Clar's proposal is based on an interpretation of properties of coronene such as reduced reactivity in comparison to phenanthrene, the structure which is contained in a single structure of coronene and the position of the proton signal in NMR which appears at an unusually low field.

#### CONCLUSIONS

The following results are achieved for benzenoid hydrocarbons:

(i) Additivity of heats of atomization of benzenoid hydrocarbons is based on the partial fragmentation of the system. The benzene and naphthalene carbon skeletons are retained as constituting blocks.

(ii) Formally the structural representations of benzenoid hydrocarbons which serve as the basis for the evaluation of heats of atomization are formulae introduced by Clar. The satisfactory agreement between the additivity schemes based on Clar's formulae and the experimental data, thus, presents independent support for Clar's notion of isolated  $\pi$ -sextets and migrating sextets.

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### SAŽETAK

#### Aditivnost toplina atomiziranja benzenoidnih ugljikovodika

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Predložena je aditivna shema, temeljena na Clarovim formulama, za izračunavanje toplina atomiziranja benzenoidnih ugljikovodika. Postignuto je vrlo dobro slaganje s vrijednostima polučenim pomoću semiempirijskih MO teorija na SCF razini i s eksperimentalnim mjerenjima. Taj rezultat nezavisno podupire Clarovu koncepciju o izoliranim i migrirajućim pi-elektronskim sektetima.

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