

2. N. S. Prostakov, A. T. Soldatenkov, and V. O. Fedorov, *Khim. Geterotsikl. Soedin.*, No. 8, 1101 (1979).
3. N. S. Prostakov, A. V. Varlamov, G. A. Vasil'ev, O. G. Kesarev, and G. A. Urbina, *Khim. Geterotsikl. Soedin.*, No. 1, 124 (1977).
4. N. S. Prostakov, K. D. Matthew, and V. A. Kurichev, *Khim. Geterotsikl. Soedin.*, No. 5, 876 (1967).
5. N. S. Prostakov, M. É. Sintra, S. A. Soldatova, V. P. Shalimov, V. P. Zvolinskii, and A. A. Savina, *Khim. Geterotsikl. Soedin.*, No. 9, 1231 (1976).
6. M. A. Ryashentseva, Kh. M. Minachev, V. V. Dorogov, and N. S. Prostakov, *Khim. Geterotsikl. Soedin.*, No. 1, 88 (1972).

QUANTUM-CHEMICAL INTERPRETATION OF RECYCLIZATION REACTIONS.

6.* ISOELECTRONIC ANALOGS OF THE BENZYL CATION. AZA DERIVATIVES

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The scheme, developed within the framework of the π -electron approximation, of the quantum-chemical description of recyclization reactions in the case of the isomerization of benzyl and methylenenaphthyl cations to tropylium ions and their benzo derivatives is extended to rearrangements with ring expansions. The effect of aza substitution on these recrystallizations is examined on the basis of the bonded variant of perturbation theory. The results of the calculations are in good agreement with the mass-spectrometric data.

In the preceding papers of this series we have developed a scheme for the quantum-chemical description of Kost-Sagitullin [1-4], Dimroth [4], and Schmidt [5] rearrangements. A similar approach, but, to be sure, without allowance for interelectronic interaction, has been used together with other reactivity indexes to explain the intramolecular aminolysis of 2-methylamino-N-{1-[3,6-di(2-pyridyl)-4-pyridazinyl]ethylidene}aniline [6, 7] and the conversion of a spiro-fused pyridazine to pyrazole derivatives [7]. Some predictions of this scheme such as the direction of recyclization of 1,2,3-trimethylisoquinolinium iodide [3] have been confirmed experimentally [8]. Experimental verification [9] has also been obtained for the fact that the introduction of an electron-acceptor substituent in the 2 position of 1-methylene-1,2-dihydroisoquinoline substantially hinders its recyclization (see Table 3 in [3]).

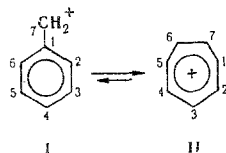
At the foundation of the approach under consideration lies the assumption of the possibility of the description of the reactivities of molecules with conjugated bonds in recyclization and cyclization reactions within the framework of the π -electron approximation on the basis of the bonded variant of perturbation theory [1]; the orders of the bonds between the chemically unbonded atoms (the "long-range" or "nonvalent" bond orders) and the bond-bond self-consistent self-polarizabilities of the starting substrate (see [1]) will be the reactivity indexes in these reactions within the approximation of an isolated molecule (a static reactivity model) in this case. Let us note that the residual π -electron charges and atom-atom self-polarizabilities, which are widely used for the interpretation of aromatic substitution reactions, are the reactivity indexes in the similar examination of the latter.

The rearrangements of isoelectronic heteroanalogs of the benzyl cation are examined in the present communication within the framework of the method being developed in this research. A peculiarity of these reactions as compared with the previously described reactions [1-5] is the fact that they generally proceed with ring expansion (for example, see [10-12]).

*See [1] for Communication 5.

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TABLE 1. Bond Orders in the Aza Derivatives of the Benzyl Cation and the Tropylium Cation



π -Electron bond orders*	Compound I					Compound II				
	benzyl cation	2N	3N	4N	7N	tropylium cation	1N	2N	3N	4N
P_{1-2}	0,486	0,472	0,486	0,473	0,535	0,642	0,621	0,621	0,669	0,623
P_{1-6}	0,486	0,465	0,488	0,473	0,535	0,159	0,176	0,125	0,149	0,149
P_{2-7}	0,283	0,323	0,298	0,263	0,272	0,159	0,208	0,176	0,125	0,149
P_{6-7}	0,283	0,236	0,261	0,263	0,272	0,642	0,669	0,623	0,666	0,623

*The numbering of the atoms in the schemes of Tables 1 and 2 does not correspond to the rearrangement of the carbocycles and the IUPAC nomenclature but was selected for convenience in the discussion of the results of the calculations.

1. The isomerization of the benzyl cation (I) to a tropylium cation (II) is one of the key rearrangements in mass spectrometry [13] and has therefore been studied thoroughly (for example, see [10, 13-16]). The randomization of the hydrogen and carbon atoms, which has been proved by means of ^2H [14] and ^{13}C labels [15], constitutes evidence of the existence of the I \rightleftharpoons II equilibrium. As a consequence of the high stability of the tropylium cation as compared with the benzyl cation (as indicated, in particular, by its greater aromatic character: compare $\chi^\pi(\text{I}) = -16.6 \text{ cm}^3/\text{mole}$ and $\chi^\pi(\text{II}) = -49.9 \cdot 10^{-6} \text{ cm}^3/\text{mole}$), this equilibrium is shifted to favor the formation of cation II, which is also confirmed by calculations of the potential surfaces by the MINDO/3 method [17-19]. This is also indicated by the results of calculations of the orders of the cleaved and newly formed bonds of I and II, which are presented in Table 1. In fact, the existence in I of a positive order of the 2-7 bond, which is greater than $P_{2,7}$ of cation II, and the fact that the order of 1-2 bond ($P_{1,2}$) cleaved in cation I is smaller than in cation II are in agreement with the shift of the equilibrium of the isomerization to favor II.

The I \rightarrow II rearrangement may proceed via two pathways: a) The 1-2 bond is initially cleaved, after which the new 2-7 bond is formed; b) the 1-2 bond is cleaved after the formation of the new 2-7 bond. In the latter case the rearrangement proceeds through a step involving the formation of a bicyclo[4.1.0]hepta-2,4-diene cation (III).



In the π -acyclic analog of cation I ($\beta_{1,2} = 0$) the long-range order of the 2-7 bond (+0.251) is somewhat smaller than in the starting cation. This fact, together with the relatively larger order of the cleavable 1-2 bond of I, indicates the preferableness of recyclization through intermediate III, in agreement with calculations by the MINDO/3 method [19].

2. Allowance for the effect of the substituents on the investigated reaction was made within the framework of the bonded variant of perturbation theory in the self-consistent-field (SCF) MO LCAO method, as described in [1-4]. An analysis of the mutual atom-bond polarizabilities of cation I shows that, as expected, the adjacent functional groups have the greatest effect on the cleaved and newly formed bonds. Thus electron-acceptor substituents in the 2 position intensify the interaction of the 2-7 atoms and weaken the interaction of the 6-7 atoms and somewhat decrease the orders of the bonds undergoing cleavage ($\pi_{2,2-7} = 0.127$, $\pi_{2,6-7} = -0.146$, $\pi_{2,1-2} = -0.046$, $\pi_{2,1-6} = -0.067$), whereas their effect is just the opposite in the 7 position ($\pi_{7,2-7} = -0.031$, $\pi_{7,1-2} = 0.151$). The introduction of electron-acceptor groups in the 3 position has virtually no effect on the strengths of the bonds undergoing cleavage, inasmuch as it strengthens the 6-7 bond and weakens the 2-7 bond ($\pi_{3,1-2} = 0.000$, $\pi_{3,1-6} = 0.004$, $\pi_{3,6-7} = 0.049$, $\pi_{3,2-7} = -0.067$). The same substituents in the 4

position weaken both the cleaved and newly formed bonds ($\pi_{4,1-2} = -0.043$, $\pi_{4,2-7} = -0.060$). The effect of electron-donor substituents is just the opposite.

One should bear in mind that the given scheme for allowance for chemical substitution does not describe transformations in which the atoms of the functional groups participate in rearrangements. In these cases the effect of substituents on the recyclization reaction must not be described by means of the atom-bond mutual polarizabilities, but rather one must introduce the π -electron centers of the functional groups directly into the computational scheme. Then, within the framework of the approach being developed here, the long-range bond orders will serve as reactivity indexes for both the initial recyclization and for the rearrangements with the participation of the substituents.

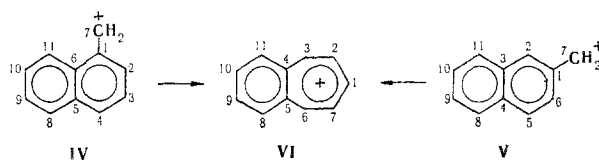
3. Since the $\Delta\alpha$ value for a nitrogen atom of the pyridine type is small [4], aza substitution in the benzyl cation should not markedly change the long-range bond orders and hinder the isomerization. In fact, as one can see from Table 1, in which the results of calculations of aza derivatives of cations I and II on the basis of perturbation theory are presented, the introduction of nitrogen atoms in the 2 or 3 position facilitates rearrangement, whereas the introduction of nitrogen atoms in the 4 or 7 position hinders it somewhat. The orders of the bonds undergoing cleavage change slightly, except for the 7-azabenzyl cation, in which they increase, thereby hindering recyclization. The presented bond orders of the azatropylium cation constitute evidence that the introduction of nitrogen atoms in cation I upon the whole does not affect the shift of the (I) \rightleftharpoons (II) equilibrium to favor the formation of II, and the rearrangement should proceed more readily for the 2-aza and 3-aza derivatives (compare with $P_{2,7}$).

This method of quantum-chemical description of recyclization reactions may prove to be extremely useful for the interpretation of mass-spectrometric data. As an example, let us examine the fragmentation of picolines, in which the investigated cations are formed (for example, see [13]). The intensity of the M-1 peak in the mass spectra of picolines is determined primarily by three factors: a) the stability of the resulting cation; b) the rate of its formation from M^+ ; c) competitive fragmentation of the molecular ion with detachment of a methyl group. Let us now deal with the quantum-chemical interpretation of each of these factors. In the case of azabenzyl cations their stability is intimately bound up with rearrangements to the azatropylium cation (see above) and, as follows from the results of the calculations, should be maximal for the 2-aza and 3-aza derivatives of I. In analogy with [1, 2], the rate of detachment of a hydrogen atom from the cation radicals of picolines can be linked with the magnitudes of the charges on the methylene groups in the aza derivatives of cation I (+0.455, +0.483, and +0.451 for the α , β , and γ isomers, respectively). This process should then take place more readily for the β isomer. The rate of competitive detachment of a methyl group from M^+ can be characterized (as in [20]) by the charges in the unsubstituted pyridine cation radical. Calculation within the framework of the unrestricted Hartree-Fock method (for example, see [21, 22]) gives the following charge distribution in the pyridine cation radical: $q_N = -0.216$, $q_2 = +0.342$, $q_3 = +0.232$, and $q_4 = +0.067$. Spin projection does not change this distribution, inasmuch as it affects primarily the spin densities. It follows from the results of the calculation that detachment of a methyl group should be most characteristic for the α isomer, and in the mass spectrum of α -picoline one may therefore expect an increase (as compared with the other isomers) in the intensity of the M-15 peak and, as a consequence, a decrease in the intensity for the M-1 peak. Thus in the picoline series the maximum intensity in the mass spectra of the M-1 peak should be observed for the β isomer, whereas the maximum intensity of the M-15 peak should be observed for the α isomer, and this is in agreement with the experimental data [23, 24].

4. Calculation of the cations of α - and β -methylenenaphthalenes (IV, V) showed that annelation of the benzyl cation has virtually no effect on any of the long-range bond orders for IV but increases the magnitude in the case of cation V (see Table 2), and these compounds should therefore undergo rearrangement to a benzotropylium cation (VI); this is in agreement with the experimental data [13]. The effect of aza substitution on this rearrangement is the same as in the case of the benzyl cation.

The orders of the cleaved and newly formed bonds for monoaza-substituted cations IV and V are presented in Table 2. In all of these compounds there are large positive long-range bond orders, which indicates the possibility of recyclization of these systems to aza derivatives of the benzotropylium cation. In fact, these rearrangements are observed in the mass spectra of methylquinolines and isoquinolines [23]. It follows from Table 2 that the

TABLE 2. Effect of Benzo Annellation on the Rearrangement of Azabenzyl Cations



Compound	π - electron bond orders			
	P_{1-2}	P_{1-6}	P_{2-7}	P_{6-7}
IV	0,497	0,408	0,283	0,126
2-Aza-IV	0,477	0,393	0,312	0,089
3-Aza-IV	0,497	0,409	0,254	0,140
4-Aza-IV	0,479	0,398	0,257	0,112
7-Aza-IV	0,554	0,432	0,294	0,153
8-Aza-IV	0,496	0,406	0,272	0,113
9-Aza-IV	0,497	0,408	0,290	0,117
10-Aza-IV	0,491	0,405	0,267	0,129
11-Aza-IV	0,495	0,406	0,283	0,090
V	0,512	0,447	0,349	0,159
2-Aza-V	0,503	0,487	0,377	0,095
5-Aza-V	0,514	0,399	0,373	0,124
6-Aza-V	0,492	0,445	0,316	0,212
7-Aza-V	0,575	0,450	0,332	0,186
8-Aza-V	0,513	0,449	0,355	0,145
9-Aza-V	0,505	0,458	0,335	0,151
10-Aza-V	0,511	0,436	0,351	0,162
11-Aza-V	0,507	0,462	0,344	0,137
VI	0,629	0,193	0,087	0,677

order of the 1-6 bond is somewhat smaller in all cases than the order of the 1-2 bond, whereas the long-range order of the 2-7 bond is greater than the order of the 6-7 bond, and recyclization of IV and V may therefore take place with the formation of a new bond between both the 2-7 and 6-7 atoms. A similar conclusion was obtained experimentally by means of ^{13}C labels [23]. Let us also note that as in the case of aza derivatives of cation I, the greatest long-range $P_{2,7}$ bond orders are observed for 5-aza cation V (+0.373) and for 2-aza cation V (+0.377).

The existence in cation VI of a long-range positive order of the 3-5 bond (0,144) constitutes evidence for the possibility of rearrangement with interconversion of the rings with one another. For the aza derivatives of cation VI this recyclization is no longer degenerate and may lead to migration of the nitrogen atom from the six-membered ring to the seven-membered ring and vice versa, in agreement with the concepts in [23].

5. Compounds I, IV, and V are odd-numbered alternant hydrocarbons for which, by virtue of the theorem of parity (for example, see [25]), the density matrixes of anions and cations with identical multiplicity differ only with respect to the sign of their "atomic" component. In other words, the orders of the bonds between atoms from different alternant sets for the anions and cations are correspondingly equal among themselves, and the residual π -electron charges on the atoms and the orders of the bonds between the atoms of the same alternant set have opposite signs. Thus the orders of the 1-2 and 1-6 bonds presented in Tables 1 and 2 for cations I, IV, and V will coincide with those in the case of the benzyl and methylenenaphthalene anions, respectively, and the long-range orders of the 2-7 and 6-7 bonds become negative for the latter. Ring expansion should therefore not be observed for these anions.

Let us note that the orders of the 4-7 bonds, which in cations I, IV, and V were negative (-0.294, -0.289, and -0.179, respectively), will be positive in the anions. According to the scheme being developed in this research, this should lead to ring contraction reactions in the investigated compounds after cleavage of the 1-2 or 1-6 bonds with the formation of derivatives of the cyclopentadienyl anion. It is apparent from these data that the rearrangement of the β -methylenenaphthalene anion should be the most hindered process as compared with the other anions under consideration by virtue of the smallness of $P_{4,7}$. These conclusions do not contradict the data of mass spectrometry of negative ions; for example, the predicted rearrangement of the benzyl anion to a vinylcyclopentadienyl anion is in agreement with the results in [26].

It is not difficult to demonstrate that the mutual atom-atom polarizabilities for the cations and anions with identical multiplicity of odd-numbered alternant hydrocarbons are correspondingly equal, whereas the mutual atom-bond polarizabilities coincide for the bonds between the atoms that correspond to the same alternant set and have the opposite sign for the bonds between the atoms of different sets. For example, for the benzyl anion the $\pi_{1,2-7}$ values coincide with the values for the benzyl cation, whereas the $\pi_{1,1-2}$ values undergo a change in sign (see above; $i = 1-7$), i.e., the effect of the substituents on the strengths of the 1-2 and 1-6 bonds is the same as in I, whereas the effect of the 2-7 and 6-7 bonds is just the opposite.

Of greatest interest in the investigated anions is the effect of substitution on the long-range order of the 4-7 bond. It follows from the calculation that electron-acceptor substituents in the 2 and 7 positions of the benzyl anion strengthen this bond, whereas such substituents in the 3 and 4 positions weaken it ($\pi_{2,4-7} = +0.035$, $\pi_{3,4-7} = -0.040$, $\pi_{4,4-7} = -0.019$, $\pi_{4,4-7} = +0.113$). Electron-donor substituents have the opposite effect. Since the effect of aza substitution is similar to the effect of a nitro group [4], the given scheme describes the rearrangements of azabenyl anions also. For example, the calculation of the 2-azabenzyl anion gives negative orders of the 3-7, 2-7, and 6-7 bonds vis-à-vis large positive orders of the 2-3 and 1-2 bonds and the long-range orders of the 4-7 and 2-6 bonds (-0.099 , -0.292 , -0.252 , $+0.699$, $+0.482$, $+0.292$, and $+0.176$, respectively; see [2]), which within the framework of the developed scheme constitutes evidence for the impossibility of recyclization of this anion both with retention of the number of atoms in the ring and with ring expansion. However, reactions involving contraction of the heteroring are extremely likely for this anion. A similar pattern is observed for the IV and V systems.

6. Since in the zero approximation of perturbation theory the charge density matrix of odd-numbered alternant radicals is defined as the average of the matrixes of the bond orders — the residual charges of the corresponding anions and cations [25] — the charges and orders of the bonds between the atoms of different alternant sets in the radicals are equal to zero. This result is also retained in the precise solution of the equations of the unrestricted Hartree-Fock method. Thus in the benzyl and methylenenaphthalene radicals the long-range $P_{2,7}$, $P_{4,7}$, and $P_{6,7}$ bond orders are equal to zero, and these radicals are stable [27] and do not undergo recyclization with either ring expansion or contraction. The introduction of functional groups or aza substitution does not alter this conclusion.

LITERATURE CITED

1. Yu. B. Vysotskii, B. P. Zemskii, T. V. Stupnikova, and R. S. Sagitullin, *Khim. Geterotsikl. Soedin.*, No. 6, 779 (1981).
2. Yu. B. Vysotskii, B. P. Zemskii, T. V. Stupnikova, R. S. Sagitullin, A. N. Kost, and O. P. Shvaika, *Khim. Geterotsikl. Soedin.*, No. 11, 1496 (1979).
3. Yu. B. Vysotskii, B. P. Zemskii, T. V. Stupnikova, and R. S. Sagitullin, *Khim. Geterotsikl. Soedin.*, No. 3, 381 (1980).
4. Yu. B. Vysotskii and B. P. Zemskii, *Khim. Geterotsikl. Soedin.*, No. 7, 934 (1980).
5. Yu. B. Vysotskii, N. A. Kovach, and O. P. Shvaika, *Izv. Sibirsk. Otd. Akad. Nauk SSSR, Ser. Khim. Nauk*, No. 2, 3 (1980).
6. G. L. Rusinov, I. Ya. Postovskii, and E. G. Kovalev, *Dokl. Akad. Nauk SSSR*, 253, 1392 (1980).
7. G. L. Rusinov, Master's Dissertation, Sverdlovsk (1980).
8. A. N. Kost, V. I. Terenin, L. G. Yudin, and R. S. Sagitullin, *Khim. Geterotsikl. Soedin.*, No. 9, 1272 (1980).
9. T. V. Stupnikova and Z. M. Skorobogatova, No. 12, 1662 (1979).
10. I. T. Bursey, M. M. Bursey, and D. C. I. Kingston, *Chem. Rev.*, 73, 191 (1973).
11. H. C. van der Plas, *Ring Transformation of Heterocycles*, Vol. 1, Academic Press, New York-London (1973), p. 484.
12. S. D. Sampl, D. A. Gightner, O. Buchardt, and C. Djerassi, *J. Org. Chem.*, 32, 997 (1967).
13. P. B. Terent'ev, *Mass Spectrometry in Organic Chemistry [in Russian]*, Vysshaya Shkola, Moscow (1979).
14. P. N. Rylander, S. Meyerson, and H. M. Grubb, *J. Am. Chem. Soc.*, 79, 842 (1967).
15. S. Meyerson and P. N. Rylander, *J. Chem. Phys.*, 27, 901 (1957).
16. A. S. Siegel, *J. Am. Chem. Soc.*, 92, 5277 (1970).
17. C. Cone, M. I. S. Dewar, and D. Landman, *J. Am. Chem. Soc.*, 99, 372 (1977).
18. M. I. S. Dewar and D. Landman, *J. Am. Chem. Soc.*, 99, 4633 (1977).

19. M. I. S. Dewar, Faraday Discussions of the Chemical Society, No. 62, 197 (1977).
20. T. V. Stupnikova, B. P. Zemskii, Yu. B. Vysotskii, R. S. Sagitullin, and Kh. Ya. Lopatinskaya, Khim. Geterotsikl. Soedin., No. 7, 959 (1980).
21. M. M. Mestechkin, Yu. B. Vysotskii, L. S. Gutyrya, and G. E. Vaiman, Teor. Éksp. Khim., 11, 362 (1975).
22. V. É. Umanskii and A. V. Luzanov, Problems of Computer Mathematics and Technology [in Russian], Naukova Dumka, Kiev (1976), p. 74.
23. Q. N. Porter and J. Baldes, Mass Spectrometry of Heterocyclic Compounds, Interscience, New York-London (1977).
24. R. Johnston, Handbook of Mass Spectrometry for Organic Chemists [Russian translation], Mir, Moscow (1975).
25. M. M. Mestechkin, The Density-Matrix Method in Molecular Theory [in Russian], Naukova Dumka, Kiev (1977).
26. V. I. Khvostenko, The Mass Spectrometry of Negative Ions in Organic Chemistry [in Russian], Nauka, Moscow (1981).
27. A. L. Buchachenko and A. M. Vasserman, Stable Radicals [in Russian], Khimiya, Moscow (1973).

FREE-RADICAL HYDROXYMETHYLATION OF BENZIMIDAZOLE

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Several benzimidazole derivatives were subjected to free-radical hydroxymethylation. Benzimidazole displays lower activity than quinoline in the first stage of the transformation, viz., in the addition of the hydroxymethyl radical. The yields of hydroxymethylation products are also determined by the ability of the intermediate benzimidazolium cation radicals to undergo oxidation and are increased in the presence of a catalyst, viz., silver ions.

Benzimidazole, like other heteroaromatic compounds, undergoes free-radical substitution in the imidazole ring [1]. It therefore seemed of interest to realize its hydroxymethylation by a method that gives excellent results in the 2-methylquinoline series: 4-Hydroxymethylquinoline is formed in 86% yield when quinaldine is refluxed with ammonium persulfate in aqueous methanol in the presence of sulfuric acid (method A) [2].

In the present research we have shown that this reaction with 1-methyl- and 1-phenylbenzimidazoles (Ia, c) gives 2-hydroxymethyl derivatives (IIa, c) in considerably lower yields, viz., 28 and 8%, respectively. However, the yield of carbinol IIa increases to 43% when the temperature is raised to 100°C (in an ampul) (method B).

The previously observed [3] decrease in the yields of homolytic substitution products in the benzimidazole series as compared with pyridine and quinoline can be explained either by the lower susceptibility of the benzimidazolium cation to attack of the radical or by the relative difficulty of the second step in the process, viz., oxidative dehydrogenation.

Considering the correlation of the relative rates of homolytic alkylation [4] and acylation [5] with the chemical shifts of the protons in the substitutable positions of the pyridine ring it may be assumed that the lower yields in the benzimidazole series are due to the difficulty involved in the first step of the process, since the chemical shifts of the 2-H protons in the 1-methyl- and 1-phenylbenzimidazolium cations (8.50 and 8.66 ppm, respectively) are lower than the chemical shift of the 4-H proton in the quinaldinium cation (8.99 ppm).

However, 1-methyl-5-nitrobenzimidazole (If), which in an acidic medium has a chemical shift of the 2-H proton that is almost the same (8.94 ppm) as that of the quinaldinium cation, undergoes virtually no hydroxymethylation under these conditions. Similar results were ob-

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