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Energetics of intramolecular hydrogen bonds and conformations of ω-diphenylphosphoryl- and ω-diphenylthiophosphoryl-substituted aliphatic alcohol molecules

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A series of conformationally heterogeneous and energetically multilevel ω-diphenylphosphoryl- and ω-diphenylthiophosphoryl-substituted aliphatic alcohols, whose molecules are capable of five-, six-, and seven-membered ring closure due to the OH...O=P or OH...S=P intramolecular hydrogen bond (IHB), was studied by IR spectroscopy and molecular mechanics. The data obtained confirm the previous assumption that the energy of IHB (E_{IHB}) differs substantially from the difference of enthalpies of conformers with and without IHB ($-\Delta H$), which is experimentally determined by the temperature dependence of the intensity of IR ν(OH) absorption bands. The measured $-\Delta H$ values can be positive, zero, or negative at significant IHB effects, *i.e.*, at high Δν(OH) and E_{IHB} values.

Key words: intramolecular hydrogen bond, energetics, ω-diphenylphosphoryl- and ω-diphenylthiophosphoryl-substituted aliphatic alcohols, IR spectroscopy, conformations, molecular mechanics calculations.

Intramolecular hydrogen bonds (IHB) play an important role in biological systems and determine the course of many chemical reactions and structures of molecules. Nevertheless, the problem of correct determination of the IHB energy remains unsolved so far.

The enthalpy of formation ($-\Delta H$) of intermolecular hydrogen bonds (IMHB) can be measured by IR spectroscopy from the temperature dependence of the relative intensity of bands of stretching vibrations of free ($\nu(\text{XH})^{\text{f}}$) and bonded ($\nu(\text{XH})^{\text{b}}$) XH groups using van´t Hoff´s equations.[1—3] The use of this approach to measuring the enthalpy of IHB sometimes results in unclear situations when the $-\Delta H$ values are not parallel to the shifts of the ν(XH) frequency but often are zero and even negative[4—6] in spite of bright manifestations of IHB formation.

We previously[7—10] showed for the series of diols, halosubstituted alcohols, and phenols that these anomalies can be explained by a complicated character of the $-\Delta H_{\text{IHB}}$ values obtained by this method. These values do not coincide with the "true" energy of IHB (E_{IHB}), because they additionally include conformational components. Unlike IMHB, the formation of IHB is always accompanied by conformational transformations, which independently change the energy-component of the IHB energy (Fig. 1). To determine E_{IHB}, we proposed[7] to subtract the corresponding difference of conformational energies (ΔE_{conf}) calculated by the molecular mechanics method ignoring the formation of IHB from the experimental value ($-\Delta H_{\text{IHB}}$) (see Fig. 1).

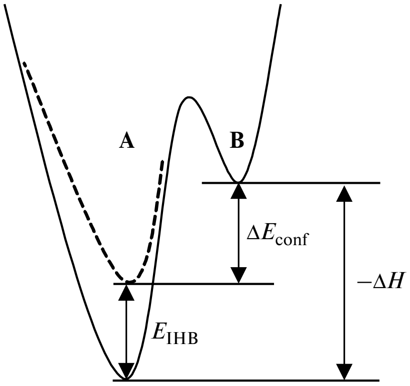

Fig. 1. Potential internal rotation curve for the equilibrium of two conformers **A** and **B**, of which **A** is stabilized by intramolecular hydrogen bonds (IHB). Experiment is solid, and molecular mechanics calculation ignoring IHB is dotted (ignoring the latter results in the situation where the calculated difference in energies of the conformers (ΔE_{conf}) is lower than the experimentally measured value ($-\Delta H$), and $E_{\text{IHB}} = -\Delta H - \Delta E_{\text{conf}}$).

Earlier attempts to elucidate the complicated character of the $-\Delta H_{\text{IHB}}$ values obtained from the IR spectroscopic data have been published.[4] However, this problem

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was not solved for a long time. At the same time, after publication of the work,[7] other authors also used hypothetical situations to estimate the energy of specific intramolecular interactions.[11—13] The empirical correlations for the estimation of the IHB energy were also considered.[14]

The purpose of this work is to extent the previously proposed approach to studies of the energetics of IHB of the conformationally heterogeneous, *i.e.*, energetically multilevel, organoelement molecules. The phosphorylated aliphatic alcohols $Ph_2P(O)(CH_2)_nOH$ (n = 1 (**1**), 2 (**2**), 3 (**3**)) and $Ph_2P(S)(CH_2)_nOH$ (n = 2 (**4**), 3 (**5**)) were chosen as objects for the study. As it has been shown previously,[6,15,16] the O—H...X=P IHB are formed in molecules of these compounds accompanied by closure of five- (in the case of **1**), six- (in the case of **2** and **4**), and seven-membered (in the case of **3** and **5**) hydrogen-bonded rings.

Experimental and calculation procedures

Compounds **1**—**5** were synthesized by previously described procedures.[15,16] IR absorption spectra of dilute solutions of substances in CCl_4 were recorded in the ν(OH) wave number interval from 3000 to 3700 cm^{-1}. Two main ν(OH) bands remained in each spectrum of solutions diluted to a concentration of $\approx 5 \cdot 10^{-4}$ mol L^{-1}, and the ratio of intensities of these bands remained unchanged with further dilution. The high-frequency band ($\nu_{max} \approx 3630$ cm^{-1}) corresponds to the $\nu(OH)^f$ vibrations of free OH groups. The low-frequency band (ν_{max} ~3340—~3570 cm^{-1}) was assigned to the $\nu(OH)^b$ vibrations of IHB-bonded groups. The $-\Delta H$ values for the average experimental temperature T_{av} were determined by the graphical method from the temperature plot of the ratio of surface areas of the absorption bands ($B_{OH}{}^b/B_{OH}{}^f$) in the 300—340 K interval using van´t Hoff´s equation

$$\ln \frac{B^f_{OH}}{B^b_{OH}} = -\Delta H \frac{1}{RT} + \text{const}.$$

The energies of possible conformers of molecules of the alcohols under study, which correspond to minima in the potential internal rotation surface, were calculated by the molecular mechanics method in the variant that takes into account the lone electron pairs (LEPs) of heteroatoms.[17,18] The potential energy of molecules was presented as

$$U = \sum_{i>j}\sum f(r_{i,j}) + 1/2\sum_i F_i(l_i - l_i^0)^2 + 1/2\sum_i K_i(\alpha_i - \alpha_i^0)^2 + $$
$$+ 1/2\sum_i V_i(1 + m_i \cos n_i\phi_i) + \sum_{i>j}\sum S(r_{i,j})q_iq_j/r_{i,j},$$

where $f(r_{i,j}) = -A/r_{i,j}{}^6 + B\exp(-Cr_{i,j})$ are the interaction potentials of valent-unbonded atoms i and j remote at the $r_{i,j}$ distance from each other; A, B, and C are empirical parameters. The second and third terms in the formula describe the energy of deformation of bond lengths l_i and bond angles α_i, *i.e.*, their deviations from the ideal values $l_i{}^0$ and $\alpha_i{}^0$, respectively. The ϕ_i value in the fourth term is the dihedral angle between the certain bond vectors; $m_i = \pm 1$. The $l_i{}^0$, F_i, K_i, $\alpha_i{}^0$, m_i, V_i, and n_i values for different atoms are published.[17,18] The q_i and q_j values in the last term are the charges of "effective" LEPs (ELEPs) estimated by the formula $q = 0.1eN$, where N is the number of LEP of the given atom, and e is the electron charge. Here $r_{i,j}$ is the distance between the ith and jth ELEPs, and $S(r_{i,j})$ is the dimensionless correction factor.[17,18] The parameters of the model were calibrated by the conformational energies of organic and organophosphorus molecules in which IHBs were absent or negligible. Calculations by this procedure make it possible to estimate all factors determining the energetics of conformational transitions of molecules **1**—**5**, except E_{IHB}.

Potential internal rotation hypersurfaces were calculated at unchanged bond angles and bond lengths. Then the structure was refined in the vicinities of the potential surface minima using the energy minimization by bond and dihedral angles.

The IBM PC computer programs developed by A. Kh. Plyamovatyi, R. Sh. Khadiullin, and A. E. Vandyukov (A. E. Arbuzov Institute of Organic and Physical Chemistry, Kazan Research Center of the Russian Academy of Sciences) were used in calculations.

Results and Discussion

The experimental data for the IR spectroscopic and thermodynamic parameters of the compounds under study[6,15,16] are presented in Table 1. It is of interest that for compounds **1** and **5** the $-\Delta H$ values are approximately equal to zero. In the case of compound **5**, the divergence of the $-\Delta H$ and E_{IHB} values is especially noticeable, because the IHB in this compound induces the significant shift Δν(OH) (220 cm^{-1}).

For compounds with IHB, the $-\Delta H$ value, which was experimentally measured from the temperature plot of the ratio of intensities (surface areas of absorption bands of bonded and free XH groups (B^b/B^f)), is the difference between the weighted average energies of conformers with the free hydroxyl group ($\bar{E}_i$) and conformers

Table 1. Spectral (ν(OH), Δν(OH)) and thermodynamic (ΔH) parameters[6,15] of molecular equilibrium involving intramolecular hydrogen bonds (IHB) for ω-diphenylphosphoryl- and ω-diphenylthiophosphoryl-substituted aliphatic alcohols in CCl_4 solutions and the corresponding calculated IHB energies (E_{IHB})

Compound	m*	$\nu(OH)^f$	$\nu(OH)^b$	Δν(OH)	$-\Delta H$ ($-\Delta E_{exp}$)	E_{IHB}	$-\Delta H$**
		cm^{-1}			kJ mol^{-1}		
1	5	3617	3570	47	~0	2.1	3.8
2	6	3635	3446	189	15.5	10.0	16.7
3	7	3643	3340	303	19.7	11.3	22.2
4	6	3628	3450	178	7.1	11.7	16.3
5	7	3640	3420	220	~0	5.9	18.6

* Number of atoms in the cycle.

** The ΔH values obtained by the Iogansen correlation.[3]

with IHB ($\bar{E}_n$) at the average temperature of measurement T_{av} [7—10,19,20]

$$-\Delta H\,(-\Delta E_{\exp}) = \left[\frac{\sum E_i W_i \exp(-E_i/RT_{\text{av}})}{\sum W_i \exp(-E_i/RT_{\text{av}})} - \frac{\sum E_n W_n \exp(-E_n/RT_{\text{av}})}{\sum W_n \exp(-E_n/RT_{\text{av}})}\right] = (\bar{E}_i - \bar{E}_n), \qquad (1)$$

where W are the statistical weights of the conformers. Equation (1) was derived using van´t Hoff´s equation in the approximation of the temperature-independent ΔH and ΔS values and the ratio of absorption coefficients $\alpha^{\text{f}}/\alpha^{\text{b}}$, as well as the $\Delta H = \Delta E$ equality valid for liquid.[2] When the molecular system contains several groups of conformers with IHBs of different types, a series of equations (1) can be written. For diols (polyols), the number of hydroxy groups is taken into account by the corresponding statistical weights W_n.[9,10]

If the E_n values for the conformers with IHB are presented as two components

$$E_n = E_n{}' - E_{\text{IHB}},$$

where $E_n{}'$ are the hypothetical values of energies of the corresponding conformers "undisturbed" by intramolecular hydrogen bonds, and E_{IHB} is the energy of IHB (E_{IHB} is positive by definition), then

$$\begin{aligned}-\Delta E_{\exp} &= \left[\frac{\sum E_i W_i \exp(-E_i/RT_{\text{av}})}{\sum W_i \exp(-E_i/RT_{\text{av}})} - \frac{\sum (E_n' - E_{\text{IHB}}) W_n \exp[-(E_n' - E_{\text{IHB}})/RT_{\text{av}})]}{\sum W_n \exp[-(E_n' - E_{\text{IHB}})/RT_{\text{av}})]}\right] = \\ &= \left[\frac{\sum E_i W_i \exp(-E_i/RT_{\text{av}})}{\sum W_i \exp(-E_i/RT_{\text{av}})} - \frac{\sum E_n' W_n \exp(-E_n'/RT_{\text{av}})}{\sum W_n \exp(-E_n'/RT_{\text{av}})} + E_{\text{IHB}}\right] = \\ &= (\bar{E}_i - \bar{E}_n') + E_{\text{IHB}}. \qquad (2)\end{aligned}$$

Here $\bar{E}_i - \bar{E}_n{}'$ is the difference of weighted average conformational energies. Denoting the latter as $\Delta E^{\text{f,b}}{}_{\text{conf}} = -\Delta E^{\text{b,f}}{}_{\text{conf}} = -\Delta E_{\text{conf}}$, we obtain for the average experimental temperature

$$-\Delta E_{\exp} = -\Delta E_{\text{conf}} + E_{\text{IHB}}$$

or

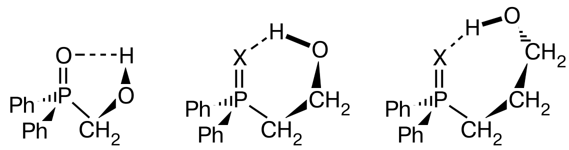
$$E_{\text{IHB}} = -\Delta E_{\exp} + \Delta E_{\text{conf}}. \qquad (3)$$

Thus, the IHB energy turns out to be equal to the measured difference in enthalpies $-\Delta H$, except for the conformational component (taking into account the sign of the corresponding values).

Equation (3) and schemes in Fig. 2 illustrate the difference between the $-\Delta H$ ($-\Delta E_{\exp}$) and E_{IHB} values and their physical meaning. In principle, the following variants are possible (see Fig. 2): (I) the weighted average energy level of conformers with the free hydroxyl group ($\bar{E}_i$) lies higher, (II) lower, or (III) coincides with the weighted average level of conformers with IHB ($\bar{E}_n$). In the case I, two variants are possible when the hypothetical weighted average energy level of conformers of the n family, calculated assuming the absence of IHB ($\bar{E}_n{}'$), is between $\bar{E}_i$ and $\bar{E}_n{}'$ (I´) or above them (I″). In the cases II and III, there is the only variant of arrangement of levels, because $\bar{E}_n{}'$ is always higher than $\bar{E}_n$. The calculated formulas demonstrating that the absolute value of E_{IHB} can be lower than (I´), higher than (I″), or equal to $-\Delta H$ (III, $\bar{E}_n = \bar{E}_i$) are given in Fig. 2.

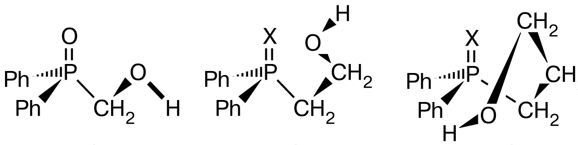
As can be seen, basically the $-\Delta H$ value can be either positive (in variant I), negative (II), or equal to zero (III) at different positive E_{IHB} values. Thus, low $-\Delta H$ values at significant $\nu(\text{OH})^{\text{b}}$ shifts can naturally be explained.

The calculation performed in this work shows that the potential internal rotation surface of molecule **1** contains 20 minima corresponding to spectroscopically discernible conformers, which can be unified in five families of conformations. Two families of the most energetically favorable conformers (with and without IHB) are shown below. Inside each family, the conformers differ only by the orientation of Ph rings. On account of geometric conditions for the mutual arrangement of the OH group and O=P moiety, the IHB is possible only in conformations of the *gg´* family. The number of similar families of conformers with and without IHB is 2 and 20, respectively,



gg´ *gg´g* *gg´g´g*

Conformers with IHB



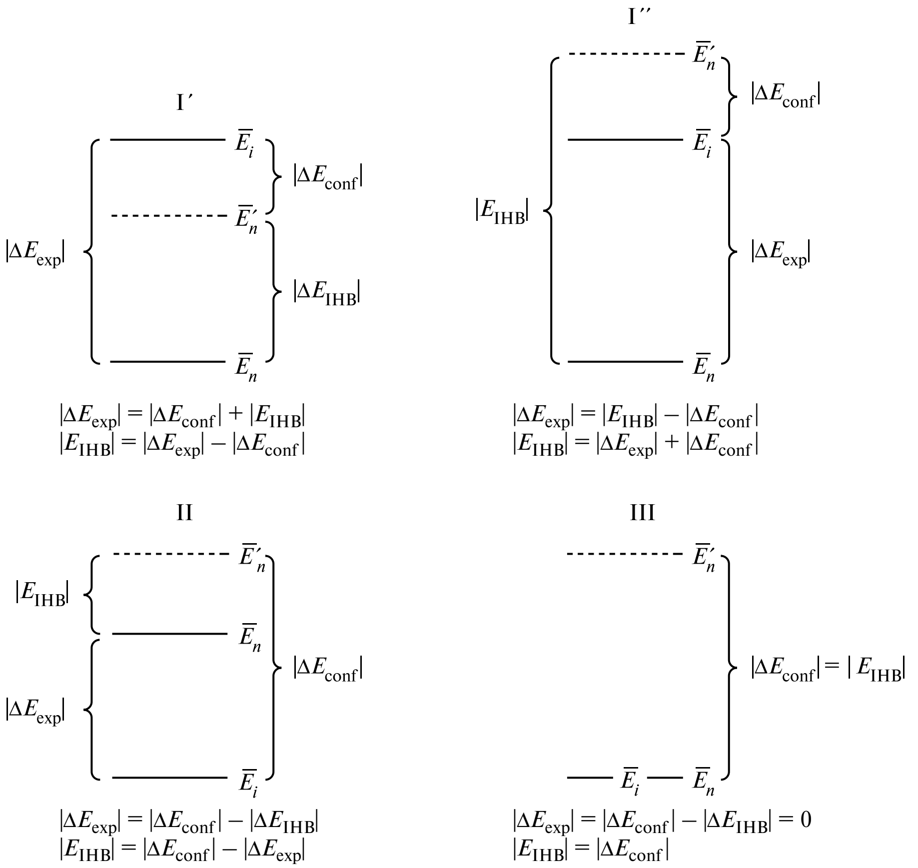
gt *ggt* *gggt*

1 **2**, **4** **3**, **5**

Conformers without IHB

X = O (**2**, **3**), S (**4**, **5**)

Note. The *g* or *g´* symbols designate *gauche*-orientations in the alcohol moiety (anti- or clockwise, respectively), and *t* designate *trans*-orientations. The first letter describes the conformation about the P—CH$_2$ bond (relatively to the P=X moiety), and the subsequent letters describe those about the bonds more and more remote from the P atom.



$$|\Delta E_{\rm exp}| = |\Delta E_{\rm conf}| + |E_{\rm IHB}|$$
$$|E_{\rm IHB}| = |\Delta E_{\rm exp}| - |\Delta E_{\rm conf}|$$

$$|\Delta E_{\rm exp}| = |E_{\rm IHB}| - |\Delta E_{\rm conf}|$$
$$|E_{\rm IHB}| = |\Delta E_{\rm exp}| + |\Delta E_{\rm conf}|$$

$$|\Delta E_{\rm exp}| = |\Delta E_{\rm conf}| - |\Delta E_{\rm IHB}|$$
$$|E_{\rm IHB}| = |\Delta E_{\rm conf}| - |\Delta E_{\rm exp}|$$

$$|\Delta E_{\rm exp}| = |\Delta E_{\rm conf}| - |\Delta E_{\rm IHB}| = 0$$
$$|E_{\rm IHB}| = |\Delta E_{\rm conf}|$$

Fig. 2. Schemes of possible combinations of weighted average energy levels: E_i are the levels of conformers without IHB, E_n are the levels of conformers taking into account IHB, and $E_n{}'$ are the hypothetical levels ignoring IHB (for clarification, see text).

for molecules of compounds **2**, 2 and 36 for **3**, 2 and 22 for **4**, and 2 and 36 for **5**.

The results of calculations for the lowest-energy conformers of molecules **1**—**5** are presented in Table 2. The list of all calculated levels would be too long and, hence, is not presented in this publication.

It should be emphasized that for molecular conformers of compounds **1** and **5** the $\bar{E}_n{}'$ levels are, in fact,

Table 2. Lowest-energy conformers for molecules of compounds **1**—**5** and their relative energies (E/kJ mol^{-1})

1	E	**2**	E	**4**	E	**3**	E	**5**	E
tg	0	*gtt*	0	*tgg*	0	*gggt*	0	*gggt*	0
tt	1.05	*gtg*	0.25	*tgt*	0	*gggg*	0.79	*gggg*	0.96
gt	2.51	*ggg*	0.46	*ggt*	0.42	*gggg′*	1.88	*gggg′*	2.38
gg	4.14	*ggt*	0.50	*gtt*	0.50	*gtg′g′*	2.15	*gtg′g′*	2.85
gg′	2.89	*gtg′*	0.50	*ggg*	0.54	*gtg′t*	2.22	*gtg′t*	3.10
		ggg′	1.55	*gtg*	0.79	*gtg′g*	2.47	*gtg′g*	3.39
		gg′t	2.55	*gtg′*	1.09	*ggtt*	2.76	*tttt*	3.39
		gg′g	7.45	*ttt*	1.30	*ggtg*	3.10	*ttgg*	3.56
				ttg	1.46	*gttg*	3.22	*ggtt*	3.68
				ggg′	1.51	*gtgg*	3.22	*tttg*	3.85
				gg′g	4.48	*gttt*	3.22	*ggtg*	4.18
						gg′g′g	5.02	***gg′g′g***	8.49
						gg′gg′	13.85	***gg′gg′***	17.11

Note. Symbols *g* and *g′* designate *gauche*-orientations of bonds in the alcohol moiety (anti- or clockwise, respectively), and *t* designates *trans*-orientations. Conformers with IHB but ignoring it in the calculation of *E* are emphasized by bold.

higher than $\bar{E}_i$, as it is required, according to variant III (see Fig. 2), for the $-\Delta H$ values to be approximately equal to zero.[6]

Using the E_i and E_n' values found by calculation and solving Eq. (2) relatively to $E_{\rm IHB}$ (under the assumption that these values are equal for different conformers with IHB in molecules of one compound and at $T_{\rm av}$ = 320 K),[6] we obtained the $E_{\rm IHB}$ values presented in Table 1. Note that IHB of the OH...π(Ph) type can be formed in molecules of compounds **1**—**5**. However, the proton-withdrawing ability of the aromatic moiety is much lower than those of the phosphoryl and thiophosphoryl groups,[6] and the corresponding Δν(OH) shifts are small. As a result, the ν(OH)...π absorption peaks are beyond the $\nu({\rm OH^f})$ contours and only complicate the latter. Based on these concepts, in calculations we included the corresponding group of conformers into free conformers.

As can be seen from the data in Table 1, the applied method really results in positive $E_{\rm IHB}$ values. It is especially important that a reasonable, non-zero estimate is obtained for compound **5**. It was natural to attempt to compare the obtained $E_{\rm IHB}$ values with those estimated by other methods, in particular, using the correlation proposed[3] for IMHB in CCl_4 solutions

$$(\Delta H)^2 = 1.92[(\Delta\nu) - 40].$$

This correlation can also be applied, most likely, to IHB closing rings larger than five-membered.[3] Therefore, the last column in Table 1 contains the results of these estimations. The divergences between $E_{\rm IHB}$ and ΔH^* (see Table 1) can be explained by systematical experimental errors,[6,16] differences in the nature of IMHB and IHB, and disadvantages of parametrization of the mechanical model of the molecule used.[17,18] The latter was not refined for the phenylphosphoryl and phenylthiophosphoryl compounds studied in this work.

Thus, the example considered illustrates possibilities of the approach developed for the solution of problems of IHB energetics in rather complicated molecular systems by a combination of IR spectroscopy and molecular mechanics for revealing the physical meaning of the measured values and searching for correlations between the spectroscopic and thermodynamic parameters of IHB.

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