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MOLECULAR AND CRYSTAL STRUCTURE OF 4,6-DIMETHYL-2-(PHENYLHYDRAZINO)PYRIMIDINE AND ITS ALKYLATION AND PROTONATION PRODUCTS

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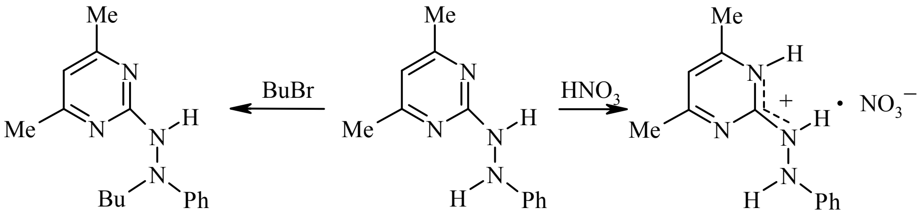
It was established by X-ray crystallographic analysis that 4,6-dimethyl-2-(phenylhydrazino)pyrimidine forms 2-(2-n-butyl-2-phenylhydrazino)-4,6-dimethylpyrimidine during alkylation with n-butyl bromide and 4,6-dimethyl-2-(phenylhydrazino)-1(H)-pyrimidinium nitrate during protonation with nitric acid. The structure of the molecules and the system of intermolecular hydrogen bonds in the crystals of the compounds are discussed.

Keywords: 4,6-dimethyl-2-(phenylhydrazino)pyrimidine, pyrimidine, alkylation, protonation, X-ray crystallographic analysis.

2-Hydrazinopyrimidine and its derivatives, like other α-hydrazinoazahetaryls, belong to the series of compounds in which there are several nucleophilic centers localized at nitrogen or carbon atoms contained in a ring or a side chain. As a rule the results from the reactions of such compounds with electrophilic reagents depend on the nature of the substituents in the heterocycle and in the hydrazine fragment. In the case of the alkylation and protonation of 4,6-dimethyl-2-(phenylhydrazino)pyrimidine (**1**) we found that the reactions in such systems can be directed at one or the other nucleophilic center, depending on the nature of the electrophilic reagent. Complications arose during determination of the structure of the product from the reaction of phenylhydrazinopyrimidine **1** with *n*-butyl bromide and also the product from its reaction with nitric acid by NMR and IR spectroscopy. Therefore, in order to determine the structure of the isolated compounds unambiguously compound **1**, the product from its alkylation with *n*-butyl bromide (2-(2-*n*-butyl-2-phenylhydrazino)-4,6-dimethylpyrimidine (**2**), and the product from its protonation with nitric acid (4,6-dimethyl-2-(phenylhydrazino)-1(H)-pyrimidinium nitrate (**3**) were submitted to X-ray crystallographic analysis.



2 **1** **3**

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TABLE 1. The Lengths (*l*) of Certain Bonds in Compounds **1-3**

Bond	*l*, Å				
	1A	**1B**	**1C**	**2**	**3**
$N_{(1)}$–$N_{(2)}$	1.384(5)	1.413(5)	1.43(1)	1.417(5)	1.398(2)
$N_{(2)}$–$C_{(9)}$	1.409(5)	1.371(5)	1.506(9)	1.397(6)	1.417(2)
$N_{(1)}$–$C_{(3)}$	1.347(5)	1.489(7)	1.15(1)	1.382(7)	1.334(2)
$N_{(4)}$–$C_{(3)}$	1.380(4)	1.326(7)	1.60(1)	1.350(6)	1.342(2)
$N_{(8)}$–$C_{(3)}$	1.377(5)	1.243(8)	1.35(1)	1.319(6)	1.340(2)
$N_{(8)}$–$C_{(7)}$	1.269(6)	1.438(6)	1.13(1)	1.326(7)	1.356(2)
$C_{(7)}$–$C_{(6)}$	1.377(7)	1.338(6)	1.40(1)	1.372(7)	1.357(3)
$C_{(6)}$–$C_{(5)}$	1.48(1)	1.317(9)	1.48(1)	1.357(8)	1.389(2)
$C_{(5)}$–$N_{(4)}$	1.346(5)	1.386(7)	1.55(1)	1.330(7)	1.340(2)

The general form, the system of hydrogen bonds, and the numbering of the atoms in the molecules of compounds **1-3** are shown in Fig. 1-6. The values of some of the bond lengths and the bond and torsion angles are given in Tables 1-3. The lengths of the corresponding bonds in the molecules of compounds **1-3** coincide within the experimental error limits. The bond lengths in the individual fragments of the molecules of the hydrazines **1-3** are comparable with the lengths typical of mono- and disubstituted hydrazines and hydrazides, like the lengths of the $N_{(1)}$–$N_{(2)}$ bonds 1.384(5), 1.417(5), and 1.398(2) (compounds **1-3** respectively). They are close to the values obtained in other types of substituted hydrazines ([1, 2] and references therein), including 5-acetyl-2-hydrazino-4-methylpyrimidine (**4**) and its hydrazone **5** [3]. The introduction of phenyl at the terminal nitrogen atom in the system of 2-hydrazinopyrimidines has little effect on the length of the $N_{(1)}$–$N_{(2)}$ bond. The length of the $N_{(1)}$–$N_{(2)}$ bond in the salt **3** has an intermediate value among the investigated compounds and is closer to the value in 1,2-diformylhydrazine (1.387(2)) [4] than in acylhydrazinium salts, e.g., in 4-phenylthiosemicarbazidodiacetic acid (1.434(9)) [5].

In the molecule of **2** (Fig. 2) the butyl fragment is at the aniline nitrogen atom. This in turn indicates primarily that the $N_{(2)}$ atom has the highest nucleophilicity in alkylation reactions. A different pattern is observed when the proton of nitric acid is used as electrophilic reagent. The reaction center in this case becomes the nitrogen atom of the heterocycle (Fig. 3). All the hydrogen atoms in the molecule of compound **3**, including those at the $N_{(8)}$ atom, were revealed in the electron density difference series and were refined in isotropic approximation.

TABLE 2. Some Bond Angles (ω) in Compounds **1-3**

Angle	ω, deg				
	1A	**1B**	**1C**	**2**	**3**
$N_{(4)}C_{(3)}N_{(8)}$	125.8(3)	135.7(5)	121.7(7)	128.9(5)	123.0(1)
$N_{(1)}C_{(3)}N_{(4)}$	112.6(3)	109.2(5)	104.4(9)	111.9(4)	117.3(1)
$N_{(1)}C_{(3)}N_{(8)}$	121.4(3)	114.1(4)	133.6(8)	119.3(4)	119.7(2)
$C_{(3)}N_{(1)}H_{(1)}$	117.(3)	117.(3)	121(1)	118.(4)	121.(1)
$N_{(2)}N_{(1)}C_{(3)}$	120.5(3)	118.0(4)	129.(1)	122.6(4)	120.0(1)
$N_{(2)}N_{(1)}H_{(1)}$	117.(3)	117.(3)	110.(1)	118.(4)	118.(1)
$N_{(1)}N_{(2)}X$*	121.(3)	107.(3)	122.3(6)	116.4(4)	111.(1)
$N_{(1)}N_{(2)}C_{(9)}$	115.8(3)	115.7(3)	116.8(6)	119.5(4)	114.5(1)
$C_{(9)}N_{(2)}X$*	122.(3)	123.(3)	120.5(5)	119.9(4)	114.(2)
$N_{(2)}C_{(9)}C_{(14)}$	121.6(4)	125.3(4)	128.3(7)	114.4(4)	121.8(2)
$N_{(2)}C_{(9)}C_{(10)}$	118.8(4)	118.1(4)	106.3(7)	126.4(4)	117.8(2)

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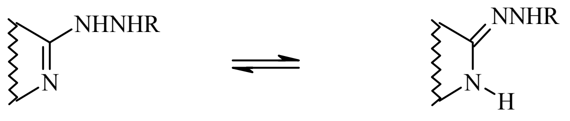
* **1A-1C**, **3** X = $H_{(N2)}$, **2** X = $C_{(17)}$.

Determination of the structure of the investigated compounds is also of interest in connection with their tautomerism. As known, α-hydrazinoazaheterocycles can exist in various tautomeric forms, e.g., hydrazine **6** or hydrazone **7**. Thus, the derivatives of pyridine exist in the hydrazine form, while hydrazinophthalazine exists in the hydrazone form ([6-8] and references therein). In solutions these forms can exist in equilibrium [5, 6].



6 **7**

According to the results from X-ray crystallographic analysis, in the crystalline state compounds **1** and **2** exist in the hydrazine form (Figs. 1 and 2). This is demonstrated by the independent effects of the position of the proton at the $N_{(1)}$ atoms and also of the proton at $N_{(8)}$ in the salt **3** and by the nature of the structural parameters (the bond lengths, torsion angle at the $N_{(1)}–N_{(2)}$ bond in compound **1** and others, Tables 1-3). After comparison of the structural parameters of compounds **1-3** it is possible to state that the cation in the nitrate **3** is a protonated form of the hydrazine tautomer of compound **1**.

The length of the $N_{(1)}–C_{(3)}$ bond in the investigated compounds **1-3** and in the hydrazines **6** and **7** is somewhat shorter than the length of the $N–C(_{sp^2})$ bond, indicating that the unshared electron pair of the $N_{(1)}$ atom is conjugated with the pyrimidine ring.

The bond lengths and the bond and torsional angles in the benzene rings are normal. The benzene rings are planar within 0.1(5) Å, i.e., within the experimental error limits. In the pyrimidine ring of compounds **1-3** a small deviation from planarity is observed (e.g., see the values of the torsion angle $C_{(5)}N_{(4)}C_{(3)}N_{(8)}$ in Table 3).

Passing on to a description of the conformation of compounds **1-3** it should be mentioned that the molecules as a whole are non-planar as a result of rotation about the $N_{(1)}–N_{(2)}$ bond (the angles of rotation about the $N_{(1)}–N_{(2)}$ bond in compounds **1-3** are 68, -90, and 85° respectively) and differ substantially. The $N_{(1)}$ atom in all the molecules is close to a coplanar arrangement with the pyrimidine fragment, whereas the $N_{(2)}$ atom is deflected significantly from the plane of the phenyl ring (Table 3). Moreover, the torsion angle $N_{(1)}N_{(2)}C_{(9)}C_{(10)}$ in the alkylated derivative **2** differs from the analogous torsion angle in the molecules of **1** and **3** by approximately 40° (Table 3) as a result of steric hindrances arising from the presence of the butyl substituent at the $N_{(2)}$ atom in the molecule of compound **2**. The unshared electron pairs of the $N_{(1)}$ and $N_{(2)}$ atoms in compounds **2** and **3** are practically orthogonal (Table 3), as in the molecules of unsubstituted hydrazine and also methyl-substituted and 1,1- and 1,2-dimethyl-substituted hydrazines and 1,2-diacyl-1-arylhydrazines (87-89°)

TABLE 3. Some Torsion Angles θ in Compounds **1-3**

Angle	θ, deg.				
	1A	**1B**	**1C**	**2**	**3**
$C_{(9)}N_{(2)}N_{(1)}C_{(3)}$	78.95(0.43)	77.96(0.48)	81.49(1.34)	76.49(0.55)	-114.98(0.17)
$C_{(9)}N_{(2)}N_{(1)}H_{(1)}$	-123.57(0.39)	-71.72(0.46)	-95.26(0.97)	-99.43(0.52)	56.21(1.34)
$XN_{(2)}N_{(1)}C_{(3)}$*	-101.36(0.41)	-65.01(0.46)	-90.77(1.19)	-80.25(0.57)	114.39(1.54)
$N_{(2)}N_{(1)}C_{(3)}N_{(4)}$	-177.66(0.30)	179.82(0.36)	-169.77(0.82)	-167.15(0.37)	174.95(0.14)
$N_{(2)}N_{(1)}C_{(3)}N_{(8)}$	-3.56(0.51)	9.19(0.62)	17.10(1.36)	13.78(0.64)	-6.81(0.24)
$N_{(1)}N_{(2)}C_{(9)}C_{(10)}$	-155.36(0.36)	-160.50(0.38)	-138.29(0.96)	-115.08(0.54)	-151.88(0.15)
$C_{(5)}N_{(4)}C_{(3)}N_{(8)}$	5.83(0.50)	-13.47(0.81)	0.54(0.61)	1.15(0.72)	3.67(0.24)
$C_{(10)}C_{(9)}C_{(14)}C_{(13)}$	3.32(0.64)	-2.47(0.74)	-10.20(1.17)	-0.03(0.92)	1.39(0.29)

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* **1A-1C**, **3** X = $H_{(N2)}$, **2** X = $C_{(17)}$.

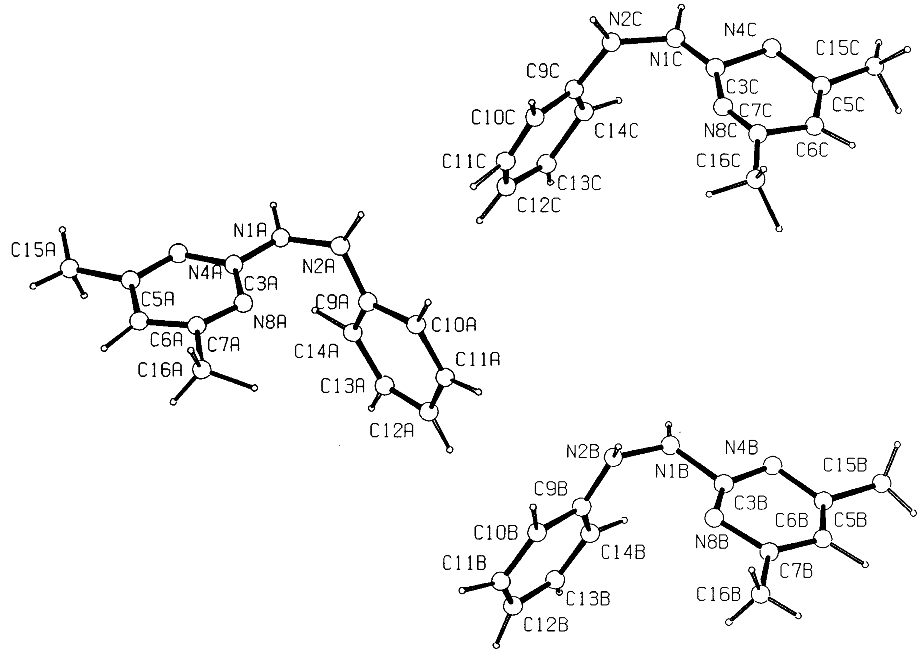


Fig. 1. The geometry of the molecule of 4,6-dimethyl-2-(phenylhydrazino)pyrimidine (**1**).

[2, 9-13]. However, even in compound **1** conditions for the formation of a single conjugated system are absent. Conjugation of the *n*-π electrons is only realized with the limits of the aminopyrimidine and, separately, the aniline fragments, whereas the molecule of 1,2-diformylhydrazine (uniform π-*n*–*n*-π conjugation), for example, is planar [4].

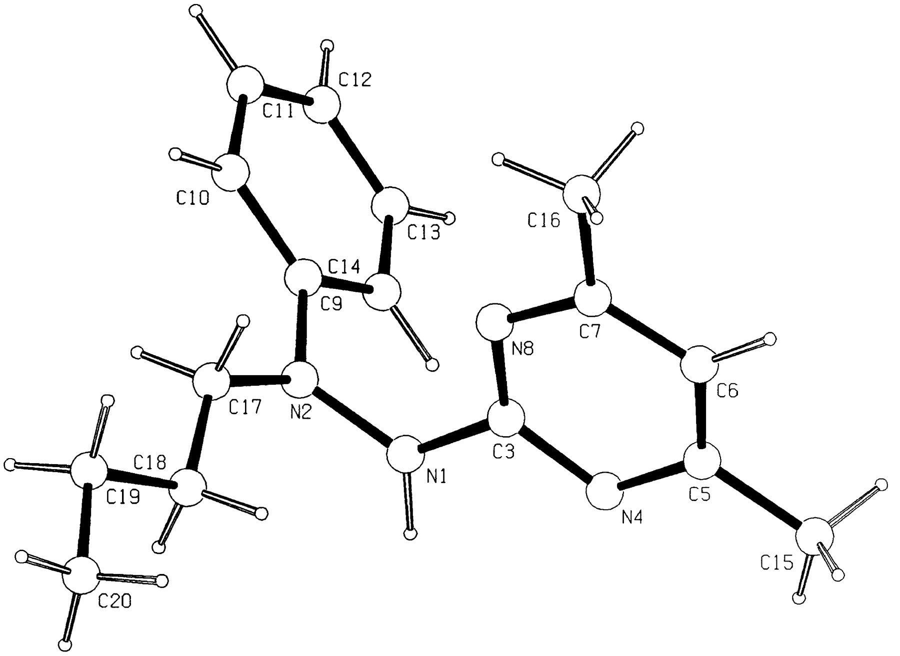


Fig. 2. The geometry of the molecule of 2-(2-butyl-2-phenylhydrazino)-4,6-dimethylpyrimidine (**2**).

TABLE 4. The Parameters of the Hydrogen Bonds in Compounds **1-3**

Compound	Bond	Symmetry operation	d (H–A), Å	d (D–A), Å	∠(DHA), deg.
1	$N_{(1A)}–H_{(1A)}\cdots N_{(4B)}$	$1 - x$, $1/2 + y$, $1/2 - z$	2.20	3.026(4)	155
	$N_{(1B)}–H_{(1B)}\cdots N_{(4A)}$	$1 - x$, $-1/2 + y$, $1/2 - z$	1.84	2.991(4)	157
	$N_{(2B)}–H_{(2B)}\cdots N_{(4B)}$	$1 - x$, $-y$, $1 - z$	2.60	3.416(5)	127
2	$N_{(1)}–H_{(1)}\cdots N_{(4)}$	$-x$, $2 - y$, $-z$	2.08	3.049(6)	165
3	$N_{(2)}–H_{(2)}\cdots O_{(2)}$	$1/2 + x$, $1/2 - y$, $1/2 + z$	2.19(2)	2.972(2)	148.4(2)
	$N_{(1)}–H_{(1)}\cdots N_{(4)}$	$1 - x$, $-y$, $2 - z$	2.14(2)	3.019(2)	179.1(2)
	$N_{(8)}–H_{(8)}\cdots N_{(2)}$	BMC	2.37(2)	2.701(2)	101.8(2)
	$N_{(8)}–H_{(8)}\cdots O_{(1)}$	$1/2 - x$, $1/2 + y$, $3/2 - z$	1.95(2	2.800(2)	157(2)

The asymmetric part of the crystal cell of compound **1** contains three independent molecules (**A**, **B**, and **C**), and one of the molecules (molecule **C**) is disordered. Molecules **A** and **B** have approximately the same conformation (see Fig. 1), but the numerical values of the geometric parameters in them differ somewhat (see Tables 1-3). It should be noted that in structures with disordered fragments the real error in the measurement of the geometric parameters is often higher than the value calculated in the refinement procedure. It should be noted that the fairly large differences in the geometric parameters of molecules **A** and **B** (see Table 1) may also be due to the real error of the measurement. The mutual arrangement of the aromatic rings in the molecules is identical, and the values of the dihedral angles between the benzene ring and the pyrimidine heterocycle are 91.2(2) and 92.0(2)° for molecules **A** and **B** respectively. The conformation of the molecules in relation to the $N_{(1)}–N_{(2)}$ bond is identical. (The torsion angles are close in absolute value at -78.9 and -71.9° respectively). The $N_{(2)}N_{(1)}C_{(3)}N_{(8)}$ fragment has an eclipsed conformation; the torsion angles are -3.6 and -9.19° respectively for molecules **A** and **B**.

The departures of the bond angles from the standard values are small (Table 2), and the angles themselves coincide with those observed in the molecules of hydrazines similar in the type of substitution. To judge from the value of the bond angles (Table 2) the configuration of the bonds with the $N_{(1)}$ and $N_{(2)}$ atoms in hydrazines **1-3** is planar-trigonal, which is characteristic of compounds of this type. The only exception is the $N_{(2)}$ atom in the nitrate **3**. Its configuration departs substantially from planar, and the $N_{(2)}$ atom has degree of pyramidality $C_p^N = 0.55$ [14]. This occurs as a result of the formation of an intramolecular hydrogen bond $N_{(8)}–H_{(N8)}\cdots N_{(2)}$, which brings about a change in the bond angles (Table 2) at the $N_{(2)}$ atom. The decrease of the $N_{(2)}C_{(9)}C_{(14)}$ angle and the increase of the $N_{(2)}C_{(9)}C_{(10)}$ angle in the alkylated hydrazine **2** compared with the angles in the hydrazine **1** and the salt **3** is explained by increase in the angle of rotation about the $N_{(2)}–C_{(9)}$ bond on account, as already mentioned above, of steric factors.

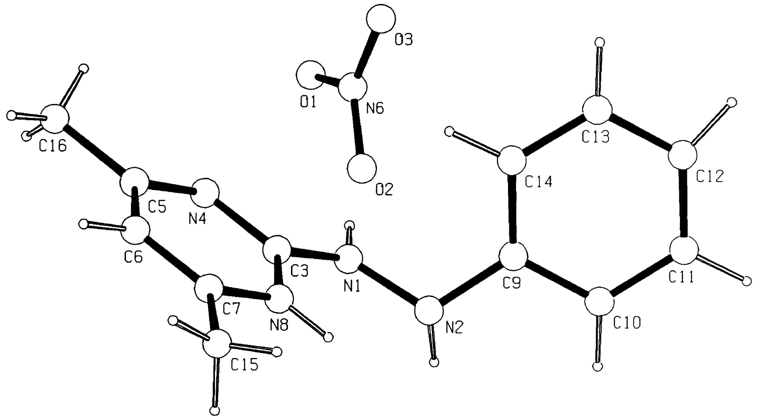


Fig. 3. The geometry of the molecule of compound **3**.

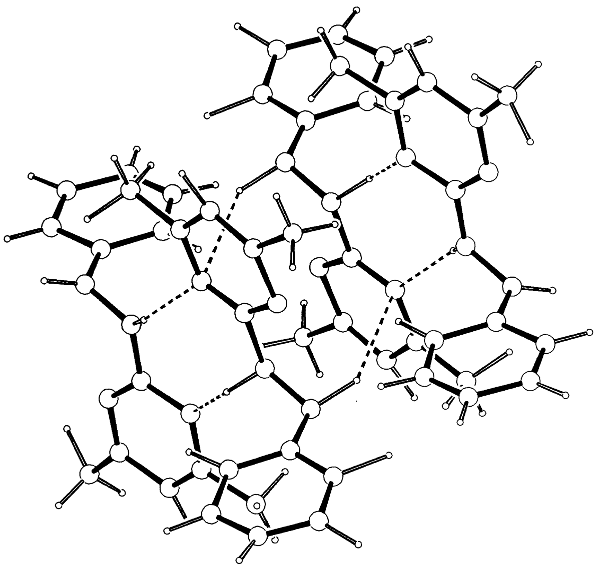
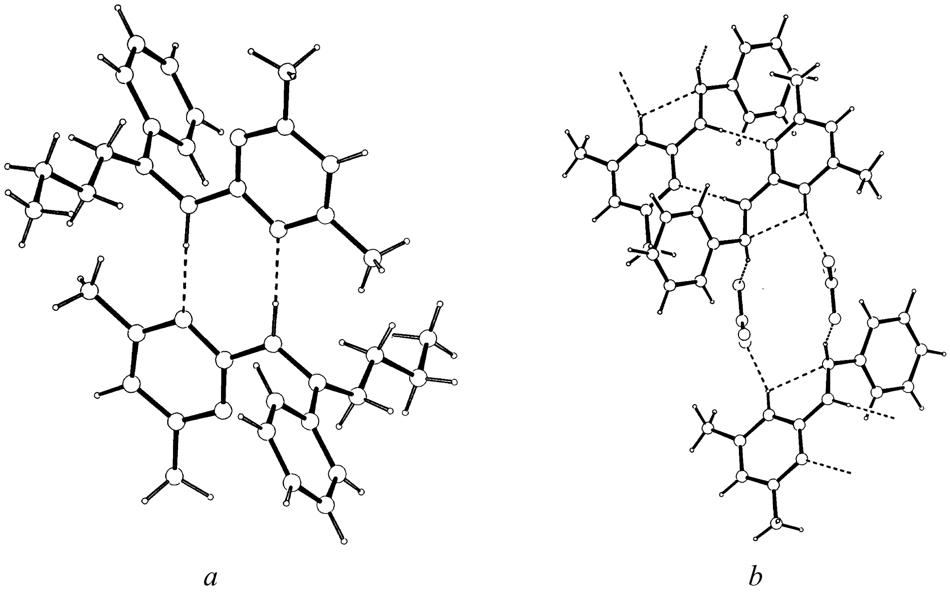


Fig. 4. The system of hydrogen bonds in the crystals of compound **1**.

Analysis of intermolecular contacts of the D–H···A type using the PLATON software [15] indicates the presence of hydrogen bonds of the N–H···N type in the crystal of the hydrazine **1** (Fig. 4). The parameters of the hydrogen bonds are given in Table 4. With the participation of the amidine fragments molecule **A** forms a pseudodimer with molecule **B**, and two such pseudodimers are linked to each other into a centrosymmetric dimer of the pseudodimers. Classical hydrogen bonds with the participation of molecule **C** were not found, and this may be due to its disordering.

The molecules of the butyl derivative **2** and of the cation of salt **3** in the crystals are linked into centrosymmetric dimers, formed as a result of intermolecular hydrogen bonds $N_{(1)}–H_{(N1)}···N_{(4)}$ with the amidine fragments of the molecule (Fig. 5). Similar dimers are observed in the crystals of compounds **4** and **5**. The



a *b*

Fig. 5. The system of hydrogen bonds in the crystals of compounds **2** (*a*) and **3** (*b*).

TABLE 5. The Parameters of the Crystals of Compounds **1-3** and the Conditions of the X-ray Crystallographic Analysis*

Parametr	Compound		
	1	**2**	**3**
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	$P2_1/c$	$P2_1/c$	$P2_1/n$
Unit cell parameters			
a, Å	10.748(1)	8.359(1)	11.0774
b, Å	21.802(6)	9.971(6)	10.4760
c, Å	14.830(2)	18.914(2)	12.3049
β, deg.	95.44(1)	96.72(1)	107.02
V, Å^3	3559(1)	1565(1)	1365.40
Z	12	4	4
M	214.27	270.38	277.28
ρ_{calc}, g/cm^3	1.23	1.15	1.15
Absorption coefficient, F(000), cm^{-1}	0.73	0.66	7.995
Radiation, λ, Å	MoKα, λ 0.71073	MoKα, λ 0.71073	CuKα, λ 1.54184
Range of θ	$2.12 \leq \theta \leq 26.3$	$2.12 \leq \theta \leq 26.3$	$3.12 \leq \theta \leq$
Scan	ω/2θ	ω/2θ	ω/2θ
Angle of scan	0.68 + 0.4 tgθ	0.68 + 0.4 tgθ	1.2 + 0.35 tgθ
Standard reflections	Two control reflections for orientation and three for intensity for each 200 reflections		
Index variation range	$-13 \leq h \leq 13$ $-27 \leq k \leq 0$ $0 \leq l \leq 18$	$-8 \leq h \leq 4$ $-12 \leq k \leq 9$ $-18 \leq l \leq 18$	$0 \leq h \leq 14$ $0 \leq k \leq 12$ $-15 \leq l \leq 14$
Reflections measured	3271	3992	3093
Number of observed reflections with $I > 3\sigma(I)$	3145	756	2345
Conditions of assignment and refinement of hydrogen atoms*^{2}	Revealed from difference series, not refined, included with fixed parameters	Revealed from difference series, not refined, included with fixed parameters	Revealed from difference series, refined isotropically
Final values of divergence factors			
R	0.087	0.048	0.054
R_W	0.098	0.051	0.069
Adjustment parameter	3.009	1.439	2.324
Δ/σ_{max}	0.1	0.2	0
Ratios of number of reflections to number of refined parameters	6.26	6.022	8.37

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*Empirical inclusion of absorption; correction for the decrease in the intensities of the control reflections was not made; software employed PCA MoIEN [16], Alpha Station 200; direct method of interpretation, SIR software [17]; refinement, full-matrix least squares; minimized function $\Sigma_\omega(|F_0| - |F_c|)_2$; allowance was not made for extinction; weighting scheme $4F_0^2 / [\sigma(I)^2 + (0.04F_0^2)]^2$.

*^{2} From the thermal vibrations of the nitrogen atoms of molecule **C** of compound **1**, and also from the large number of peaks in the electron density difference series around the central part of molecule **C** it is possible to suppose that it is statistically disordered. Unfortunately, it was not possible to distinguish between the individual positions of the disordered atoms of this molecule, and as a result the geometric parameters of molecule **C** were established with large experimental error. Subsequently the hydrogen atoms of molecules **A** and **B** were revealed from the electron density difference series, and the H atoms of molecule **C** were placed at the calculated positions.

parameters of the hydrogen bonds are given in Table 4. In addition to the intermolecular hydrogen bonds, in the cation of the salt **3** there is a short nonbonding $N_{(8)}\cdots N_{(2)}$ contact of 2.701(2) Å, indicating the existence of an intermolecular hydrogen bond $N_{(8)}–H_{(N8)}\cdots N_{(2)}$ with parameters: $N_{(8)}–H_{(N8)}$ 0.90(2), $H_{(N8)}\cdots N_{(2)}$ 2.37(2) Å, angle $N_{(8)}–H_{(N8)}\cdots N_{(2)}$ 102(2)°.

In the crystal of the hydrazine **1** there are also π–π interactions between the pyrimidine rings. Molecules **A** and **C'** participate in this type of interactions with the corresponding molecules **C** and **A'** linked by a symmetry operation (2 - *x*, 1/2 + *y*, 1/2 - *z*), on account of which pairs of molecules are formed. The parameters of the short contacts for the interacting pairs are identical, i.e., the distances between the centers of the rings 4.39(2) Å, the dihedral angle between the planes 1.2(3)°, and the angle between the normal to one of the planes and the vector linking the centers of the rings 39.6(2)°.

EXPERIMENTAL

X-ray Crystallographic Investigation. Compounds **1-3** were investigated on an Enraf-Nonius CAD-4 automatic four-circle diffractometer. The parameters of the crystals of compounds **1-3** and the conditions of X-ray crystallographic analysis are given in Table 5.

The crystals of compound **1** ($C_{12}H_{14}N_4$; mp 166-169°C) were grown from heptane, compound **2** ($C_{16}H_{22}N_4$; mp 138°C) from a mixture of DMF and hexane, compound **3** ($C_{12}H_{15}N_5O_3$; mp 78-79°C) from a 1:1 mixture of 2-propanol and water.

The methods for the synthesis of compounds **2** and **3**, the spectral characteristics of compounds **1-3**, and the elemental analyses will be presented in the next paper.

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