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G.A. Tolstikov on his 75th anniversary

## Synthesis and Steric Structure of *N''*-(Arylmethylidene)-*N'''*-(1-methyl-3-phenylprop- 2-yn-1-ylidene)thiocarbonohydrazides

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Received May 30, 2007

**Abstract**—Thiocarbonohydrazones derived from aromatic aldehydes reacted with 4-phenylbut-3-yn-2-one in aqueous acetic acid (20°C, 2 h) to produce previously unknown *N''*-[(*E-s-cis*)-arylmethylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazides with high chemo-, regio-, and stereoselectivity. The steric structure of *N''*-[(*E-s-cis*)-benzylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide in crystal and in solution was studied by X-ray analysis and IR and NMR spectroscopy.

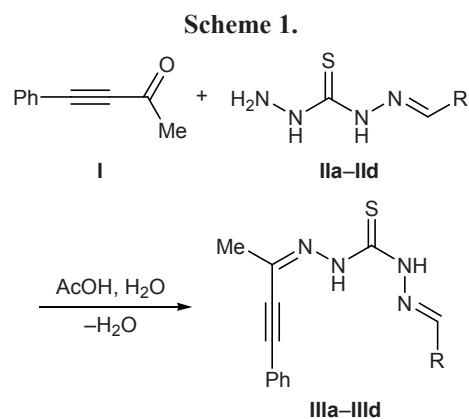
**DOI:** 10.1134/S1070428008010144

Thiocarbonohydrazones and their complexes with transition complexes exhibit a broad spectrum of biological activity (antituberculostatic, antitumor, antiviral, fungicidal) [1–5]. Hydrazones having unsaturated fragments, including acetylenic, attract interest as highly reactive building blocks for organic synthesis. Such N,S-multident conjugated systems are promising as ligands for the preparation of polynuclear complexes [3–8] possessing specific properties (such as redox and magnetic) [3, 7, 9–11]. A convenient approach to such compounds may be based on reactions of accessible  $\alpha$ -acetylenic ketones with thiocarbonohydrazide (thiocarbonic acid dihydrazide) and its derivatives [2, 12].

We previously reported that terminal  $\alpha$ -acetylenic ketones react with aromatic aldehyde thiocarbonohydrazones to give, depending on the reactant ratio, 2-(2-acylvinyl)- or 2,2-bis(2-acylvinyl)thiocarbonohydrazones [13] and that the reactions of benzaldehyde thiocarbonohydrazone with 1-benzoyl- and 1-thenoyl-2-phenylacetylenes involve the triple C≡C bond and

the carbonyl group to produce functionally substituted dihydropyrazoles [14].

In continuation of our studies on reactions of bi-electrophilic  $\alpha$ -acetylenic ketones with thiocarbonohydrazones and their synthetic potential, we examined the reactions of 4-phenylbut-3-yn-2-one (**I**) with thiocarbonohydrazones **IIa–IIId** derived from aromatic

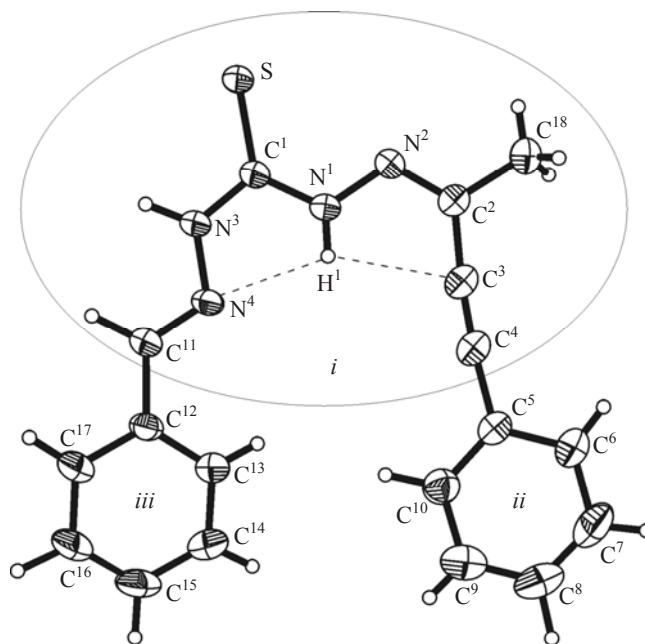


aldehydes. The reactions were carried out in aqueous acetic acid at room temperature (reaction time 2 h), and they involved exclusively the carbonyl group in molecule **I** to give the corresponding *N'''*-[(*E-s-cis*)-arylmethylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazides **IIIa–III d** in 78–82% yield (Scheme 1).

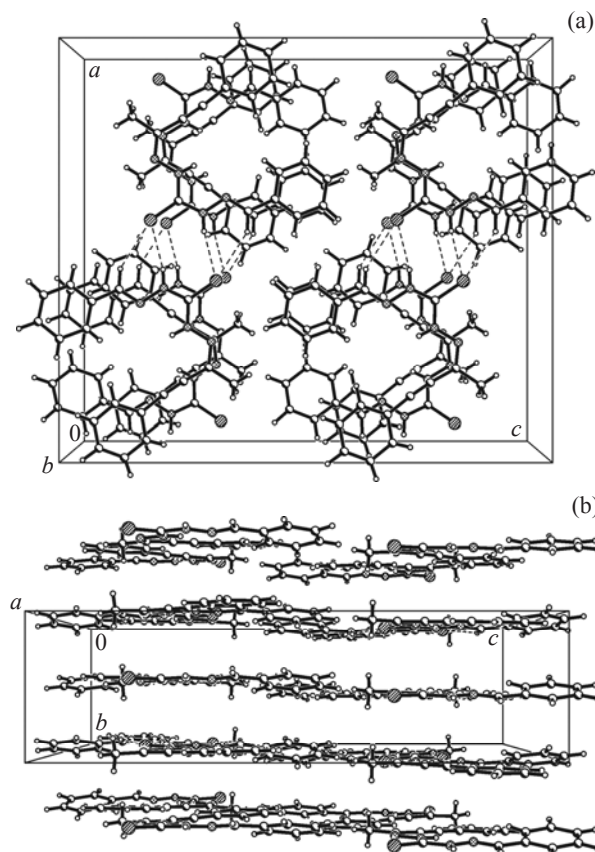
The molecular and crystalline structure of compound **IIIa** was determined by X-ray analysis of its single crystal. Conformational behavior of compounds **IIIa–III d** in solution was studied by IR spectroscopy and  $^1\text{H}$ ,  $^{13}\text{C}$ , and  $^{15}\text{N}$  NMR using two-dimensional homo- and heteronuclear correlation techniques. We also performed quantum-chemical calculations of normal vibration frequencies for molecule **IIIa** with the geometric parameters determined by X-ray analysis.

The crystalline structure of **IIIa** is represented by one crystallographically independent  $\text{C}_{18}\text{H}_{16}\text{N}_4\text{S}$  molecule (Fig. 1) located in the general position. The molecule has almost planar structure: the maximal deviation of non-hydrogen atoms from the mean-square plane does not exceed 0.44 Å ( $\text{C}^{14}$ ). The maximal deviation from the plane formed by all non-hydrogen atoms (except for carbon atoms in the benzene rings; plane *i* in Fig. 1) is observed for the  $\text{N}^3$  atom (0.06 Å). The distance between the  $\text{N}^3$  and  $\text{N}^1$  atoms is equal to 2.23(2) Å; it suggests formation of intramolecular hydrogen bond  $\text{N}\cdots\text{H}$  (the sum of the van der Waals radii of the nitrogen and hydrogen atoms is 2.75 Å [15]). Also, formation of a weak hydrogen bond like  $\text{N}^1\text{--H}^1\cdots\text{C}^3$  is possible, as was reported in [16, 17]. The  $\text{H}^1\cdots\text{C}^3$  distance is 2.37(2) Å; the sum of the corresponding van der Waals radii (C and H) is 2.9 Å [15]. The dihedral angle between the benzene ring planes (*ii* and *iii*) is 163.0°, and the dihedral angles between the *i* and *ii* and *i* and *iii* planes are 176.3° and 166.6°, respectively. The torsion angles  $\text{N}^1\text{N}^2\text{C}^2\text{C}^{18}$ ,  $\text{C}^1\text{N}^1\text{N}^2\text{C}^2$ ,  $\text{N}^2\text{N}^1\text{C}^1\text{S}$ ,  $\text{N}^2\text{N}^1\text{C}^1\text{N}^4$ ,  $\text{SC}^1\text{N}^4\text{N}^3$ ,  $\text{N}^1\text{C}^1\text{N}^4\text{N}^3$ ,  $\text{C}^1\text{N}^4\text{N}^3\text{C}^{11}$ ,  $\text{C}^{18}\text{C}^2\text{C}^3\text{C}^4$ , and  $\text{C}^2\text{C}^3\text{C}^4\text{C}^5$  are 179.7(2), 177.4(2), 0.4(2), 178.7(1), 177.2(1), 3.6(2), 177.4(1), 3(2), and 3(4)°, respectively.

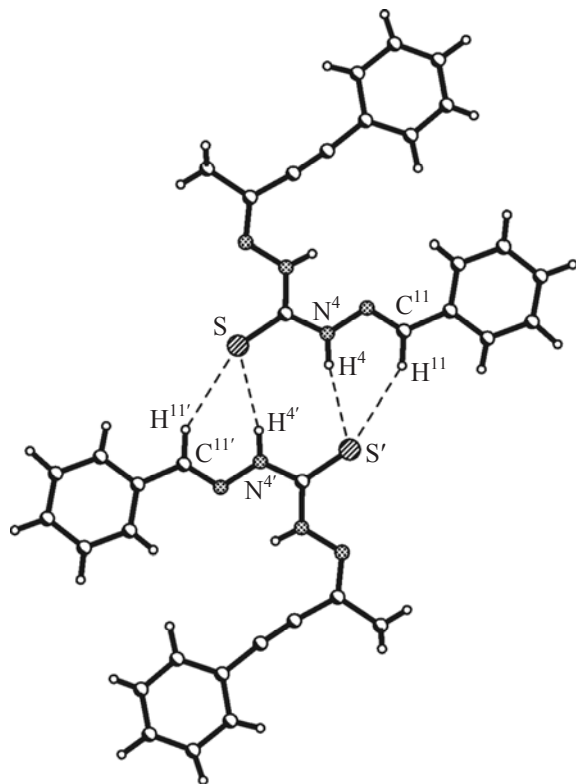
Molecules **IIIa** in crystals are arranged in layers parallel to the *ac* plane (Figs. 2a, 3), and the layers are stacked along the *b* axis (Fig. 2b). No shortened intermolecular contacts were revealed in the stacks thus formed. Molecules in neighboring stacks are linked in pairs through shortened intermolecular interactions  $\text{S}\cdots\text{H}^4$  2.53(2) Å [the  $\text{N}^4\text{--H}^4$  bond length is 0.91(2) Å, and the angle  $\text{SH}^4\text{N}^4$  is 168(2)°] and  $\text{S}\cdots\text{H}^{11}$  2.88(2) Å [ $\text{C}^{11}\text{--H}^{11}$  1.03(2) Å,  $\angle\text{SH}^{11}\text{C}^{11}$  147(1)°] (the sum of the van der Waals radii of S and H is 3.00 Å [15]).



**Fig. 1.** Structure of the molecule of *N'''*-[(*E-s-cis*)-benzylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide (**IIIa**) with atom numbering according to the X-ray diffraction data.



**Fig. 2.** Packing of molecules of *N'''*-[(*E-s-cis*)-benzylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide (**IIIa**) in crystal.



**Fig. 3.** Associates in the crystalline structure of *N''*-[(*E-s-cis*)-benzylidene]-*N'''*-[(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide (**IIIa**). Intermolecular interactions are shown with dashed lines.

The IR spectrum of a crystalline sample of **IIIa** contains two bands in the region corresponding to N–H stretching vibrations, at 3265 and 3130  $\text{cm}^{-1}$ . Among these, the high-frequency band (3265  $\text{cm}^{-1}$ ) does not change its position in going to  $\text{CHCl}_3$  solution and is concentration-independent in the range from  $10^{-2}$  to  $10^{-4}$  M. These data indicate that this band belongs to stretching vibrations of the  $\text{N}^1\text{H}^1$  group involved in intramolecular hydrogen bonding  $\text{NH}\cdots\text{N}$  (in agreement with the X-ray diffraction data). The peak intensity of the low-frequency band (3130  $\text{cm}^{-1}$ ) in solution decreases in parallel with the concentration. Simultaneously, a new absorption band appears at 3335  $\text{cm}^{-1}$  whose intensity increases synchronously. We can conclude that dissolution of compound **IIIa** is accompanied by dissociation of associates linked through intermolecular hydrogen bonds  $=\text{S}\cdots\text{H}-\text{N}$  ( $\nu_{\text{NH}}$  3130  $\text{cm}^{-1}$ ), and the band appearing at 3335  $\text{cm}^{-1}$  in the IR spectrum of solution should be assigned to stretching vibrations of the free  $\text{N}^4\text{H}$  group. The higher frequency of  $\text{N}^4\text{H}$  stretching vibrations (3335  $\text{cm}^{-1}$ ) as compared to the  $\text{N}^1\text{H}^1$  group (3265  $\text{cm}^{-1}$ ) may be rationalized by participation of the latter in intramolecular hydrogen

bonding. On the other hand, the observed frequency is considerably lower than that typical of free NH groups in the IR spectra of aromatic thiourea derivatives (3390  $\text{cm}^{-1}$ ) [18].

The above assignment of  $\nu_{\text{NH}}$  bands in the IR spectra of **IIIa** was compared with the results of quantum-chemical calculations of the corresponding normal vibration frequencies of molecule **IIIa**, which were performed using the geometric parameters determined by X-ray analysis. The non-valence  $\text{N}\cdots\text{H}$  distance (2.209 Å) in the optimized structure was similar to that found for the crystalline state. The calculated  $\nu_{\text{NH}}$  values (with a scaling factor of 0.945) for the free NH group (3328  $\text{cm}^{-1}$ ) and NH group involved in intramolecular hydrogen bond (3283  $\text{cm}^{-1}$ ) agree well with the experimental frequencies measured for compound **IIIa** in solution (3335 and 3265  $\text{cm}^{-1}$ , respectively). The calculated frequencies for the free NH groups in aromatic derivatives of thiourea (3390–3400  $\text{cm}^{-1}$ ) were also consistent with the experimental values. Therefore, molecules **IIIa** in crystal and in solution have the same structure, and the low stretching vibration frequency of the free NH group in the IR spectra of **IIIa** in solution is determined by stereoelectronic factors.

Rupture of  $=\text{S}\cdots\text{H}-\text{N}$  intermolecular hydrogen bonds in going to solution is also confirmed by the  $^1\text{H}$  NMR data. Dilution of a solution of **IIIa** in  $\text{CDCl}_3$  is accompanied by upfield shift of the NH signal ( $\text{NHN}=\text{CHPh}$ ) from  $\delta$  11.01 to 10.12 ppm ( $^1J_{\text{NH}} = 98.3$  Hz). On the other hand, the NOESY spectrum of a dilute solution contained a cross peaks resulting from dipole–dipole interactions between the  $=\text{CH}$  and  $\text{HNN}^3$  protons, indicating *E* configuration of the benzylidene fragment in **IIIa**, as in crystal. The chemical shift of the NH proton that does not participate in intermolecular hydrogen bonding ( $\text{NHN}^2=\text{C}$  fragment) does not depend on the concentration ( $\delta$  11.20 ppm,  $^1J_{\text{NH}} = 93.58$  Hz).

The  $^{13}\text{C}$  NMR spectrum of **IIIa** is characterized by a large difference in the chemical shifts of the  $\alpha$ - and  $\beta$ -acetylenic carbon atoms,  $\Delta\delta(\text{C}_{\text{sp}}) = 22.83$  ppm. This difference may be rationalized in terms of *anti* orientation of the acetylenic fragment with respect to the lone electron pair on the  $\text{N}^2$  atom and, correspondingly, *cis* orientation of the acetylenic fragment and NH group with respect to the  $\text{C}=\text{N}^2$  bond. An analogous  $\Delta\delta(\text{C}_{\text{sp}})$  value (20.29 ppm) was reported in [19] for (*Z*)-4-phenylbut-3-yn-2-one phenylhydrazone, while the  $\Delta\delta(\text{C}_{\text{sp}})$

value for the *E* isomer was 1.98 ppm. According to [19], signal from the methyl carbon atom oriented *syn* with respect to the lone electron pair on the hydrazone nitrogen atom (*Z* isomer) appears in a weaker field ( $\delta_C$  21.78 ppm) than the corresponding signal of the *E* isomer ( $\delta_C$  16.33 ppm). A similar pattern was also observed for the methyl carbon signals in the  $^{13}\text{C}$  NMR spectra of *Z* and *E* isomers of ketone oximes [20]. In the  $^{13}\text{C}$  NMR spectrum of **IIIa**, the methyl carbon nuclei resonated at  $\delta_C$  22.99 ppm. Taking into account that this value approaches that reported for the  $\text{HNN}=\text{C}(\text{CH}_3)\text{C}\equiv\text{CPh}$  fragment in the spectrum of (*Z*)-4-phenylbut-3-yn-2-one phenylhydrazone [19], the corresponding fragment in molecule **IIIa** was assigned *Z* configuration.

Thus the above data demonstrate that the *Z-s-trans* and *E-s-cis* configurations of the hydrazone fragments in molecule **IIIa**, determined by X-ray analysis, are conserved for monomeric molecules in dilute solution. This conclusion may be important, e.g., while planning syntheses of complexes on the basis of functionalized *N,S*-electron-donor ligands capable of coordinating metal cations in different modes.

The spectral parameters of compounds **IIIb–IIIId** both in crystal (IR) and in solution (IR, NMR) were similar to those found for compound **IIIa**, indicating their analogous structure. The nature of substituent in the aromatic ring of initial thiocarbonohydrazones **IIa–IIId** almost did not affect the yield of target products **IIIa–IIIId**.

Thus we have developed a simple procedure for the synthesis of previously unknown highly functionalized dihydrazones having an acetylenic bond as potential precursors of drugs, practically important materials, and intermediate products for organic synthesis.

## EXPERIMENTAL

The IR spectra were recorded on a Specord 75-IR spectrophotometer from samples prepared as KBr pellets or solutions in  $\text{CHCl}_3$ . The NMR spectra were measured from solutions in  $\text{CDCl}_3$  on Bruker DPX-400 and AV-400 spectrometers (400.13 MHz for  $^1\text{H}$ , 100.61 MHz for  $^{13}\text{C}$ , and 40.53 MHz for  $^{15}\text{N}$ ) using hexamethyldisiloxane ( $^1\text{H}$ ,  $^{13}\text{C}$ ) and nitromethane ( $^{15}\text{N}$ ) as internal references. The X-ray diffraction data were obtained on an Enraf Nonius CAD-4 diffractometer ( $\omega/2\theta$  scanning,  $\text{MoK}_\alpha$  irradiation, graphite monochromator) at room temperature. Quantum-chemical

calculations of isolated molecule **IIIa** were performed at the B3LYP/6-311G\* level using Gaussian-98 software package [21].

***N''*-(*E-c-cis*)-Arylmethylidene]-*N''*'-(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazides **IIIa–IIIId** (general procedure). Thiocarbonohydrazone **IIa–IIId**, 5 mmol, was added under stirring to a solution of 0.72 g (5 mmol) of 4-phenylbut-3-yn-2-one (**I**) in 20 ml of acetic acid, the mixture was stirred for 1 h at 20°C, 20 ml of water was added, and the mixture was stirred for 1 h more. The precipitate was filtered off, washed with 20 ml of water, dried in a vacuum desiccator over  $\text{CaCl}_2$ , and recrystallized from acetonitrile.**

***N''*-(*E-s-cis*)-Benzylidene]-*N''*'-(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide (**IIIa**). Yield 1.31 g (82%), mp 154–156°C. IR spectrum (KBr),  $\nu$ ,  $\text{cm}^{-1}$ : 2179 w ( $\text{C}\equiv\text{C}$ ); 3130, 3265 (NH).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 2.41 s (3H, Me), 7.07–7.59 m (10H, Ph), 8.07 s (1H,  $\text{CH}=\text{N}$ ), 10.12 s (1H,  $\text{HNN}=\text{CH}$ ,  $^1J_{\text{NH}} = 98.3$  Hz), 11.20 s (1H,  $\text{HNN}=\text{CMe}$ ,  $^1J_{\text{NH}} = 93.58$  Hz).  $^{13}\text{C}$  NMR spectrum,  $\delta_C$ , ppm: 22.99 (Me); 80.05 ( $\text{MeCC}\equiv$ ); 102.88 ( $\text{PhC}\equiv$ ); 120.33, 127.41, 128.65, 128.74, 130.21, 130.51, 132.06, 132.86 ( $\text{C}_{\text{arom}}$ ); 137.10 ( $\text{C}=\text{N}$ ); 144.11 ( $\text{CH}=\text{N}$ ); 173.89 ( $\text{C}=\text{S}$ ).  $^{15}\text{N}$  NMR spectrum,  $\delta_N$ , ppm: –208.30 ( $\text{HNN}=\text{CH}$ ,  $^1J_{\text{NH}} = 98.3$  Hz), –207.04 ( $\text{HNN}=\text{CMe}$ ,  $^1J_{\text{NH}} = 93.58$  Hz), –70.3 ( $\text{N}=\text{CH}$ ), –55.3 ( $\text{N}=\text{CMe}$ ). Found, %: C 67.23; H 5.21; N 17.38; S 10.34.  $\text{C}_{18}\text{H}_{16}\text{N}_4\text{S}$ . Calculated, %: C 67.48; H 5.03; N 17.49; S 10.01.**

***N''*-(*E-s-cis*)-4-Dimethylaminobenzylidene]-*N''*'-(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonohydrazide (**IIIb**). Yield 1.44 g (79%), mp 170–173°C. IR spectrum (KBr),  $\nu$ ,  $\text{cm}^{-1}$ : 2179 w ( $\text{C}\equiv\text{C}$ ); 3117, 3256 (NH).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 2.39 s (3H, Me), 2.95 s (6H,  $\text{Me}_2\text{N}$ ), 6.28–7.61 m (9H,  $\text{H}_{\text{arom}}$ ), 7.90 s (1H,  $\text{CH}=\text{N}$ ), 10.67 (1H,  $\text{HNN}=\text{CH}$ ,  $^1J_{\text{NH}} = 98.3$  Hz), 11.21 s (1H,  $\text{HNN}=\text{CMe}$ ,  $^1J_{\text{NH}} = 94.02$  Hz).  $^{13}\text{C}$  NMR spectrum,  $\delta_C$ , ppm: 22.97 (Me); 40.06 ( $\text{NMe}_2$ ); 80.36 ( $\text{MeCC}\equiv$ ); 102.36 ( $\text{PhC}\equiv$ ); 111.59, 120.44, 120.74, 128.72, 128.92, 129.81, 132.26, 151.78 ( $\text{C}_{\text{arom}}$ ); 136.02 ( $\text{C}=\text{N}$ ); 144.73 ( $\text{CH}=\text{N}$ ); 173.12 ( $\text{C}=\text{S}$ ).  $^{15}\text{N}$  NMR spectrum,  $\delta_N$ , ppm: –210.6 ( $\text{HNN}=\text{CH}$ ,  $^1J_{\text{NH}} = 98.3$  Hz), –207.6 ( $\text{HNN}=\text{CMe}$ ,  $^1J_{\text{NH}} = 94.02$  Hz), –87.6 ( $\text{N}=\text{CH}$ ), –59.81 ( $\text{N}=\text{CMe}$ ). Found, %: C 66.19; H 5.83; N 19.02; S 8.80.  $\text{C}_{20}\text{H}_{21}\text{N}_5\text{S}$ . Calculated, %: C 66.09; H 5.82; N 19.27; S 8.82.**

***N''*-(*E-s-cis*)-4-Chlorobenzylidene]-*N'''*-(*Z-s-trans*)-1-methyl-3-phenylprop-2-yn-1-ylidene]thiocarbonylhydrazide (IIIc). Yield 1.43 g (81%), mp 169–171°C. IR spectrum (KBr),  $\nu$ ,  $\text{cm}^{-1}$ : 2180 w (C≡C); 3119, 3276 (NH).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 2.40 s (3H, Me), 6.98–7.56 m (9H,  $\text{H}_{\text{arom}}$ ), 7.98 s (1H, CH=N), 10.76 s (1H, HNN=CH,  $^1J_{\text{NH}} = 98.1$  Hz), 11.17 s (1H, HNN=CMe,  $^1J_{\text{NH}} = 93.9$  Hz).  $^{13}\text{C}$  NMR spectrum,  $\delta_{\text{C}}$ , ppm: 22.96 (Me); 80.03 (MeCC≡); 102.93 (PhC≡); 120.28, 128.47, 128.85, 129.00, 130.38, 131.36, 131.99, 136.46 ( $\text{C}_{\text{arom}}$ ); 137.42 (C=N); 142.49 (CH=N); 173.91 (C=S).  $^{15}\text{N}$  NMR spectrum,  $\delta_{\text{N}}$ , ppm: –210.8 (HNN=CH,  $^1J_{\text{NH}} = 98.1$  Hz), –207.2 (HNN=CMe,  $^1J_{\text{NH}} = 93.9$  Hz), –74.0 (N=CH), –61.8 (N=CMe). Found, %: C 60.68; H 4.04; Cl 10.12; N 15.98; S 9.13.  $\text{C}_{18}\text{H}_{15}\text{ClN}_4\text{S}$ . Calculated, %: C 60.92; H 4.26; Cl 9.99; N 15.79; S 9.04.**

***N''*-(*Z-s-trans*)-1-Methyl-3-phenylprop-2-yn-1-ylidene]-*N'''*-(*E-s-cis*)-4-nitrobenzylidene]thiocarbonylhydrazide (IIIId). Yield 1.43 g (78%), mp 174–176°C. IR spectrum (KBr),  $\nu$ ,  $\text{cm}^{-1}$ : 2177 w (C≡C); 3100, 3275 (NH).  $^1\text{H}$  NMR spectrum,  $\delta$ , ppm: 2.40 s (3H, Me), 7.42–7.78 (9H,  $\text{H}_{\text{arom}}$ ), 8.09 s (1H, CH=N), 10.71 s (1H, HNN=CH), 11.23 s (1H, HNN=CMe).  $^{13}\text{C}$  NMR spectrum,  $\delta_{\text{C}}$ , ppm: 22.94 (Me); 79.82 (MeCC≡); 103.20 (PhC≡); 120.08, 123.95, 127.72, 129.03, 130.72, 131.88, 138.82, 148.42 ( $\text{C}_{\text{arom}}$ ); 138.21 (C=N); 140.66 (CH=N); 174.02 (C=S).  $^{15}\text{N}$  NMR spectrum,  $\delta_{\text{N}}$ , ppm: –208.7 (HNN=CH), –205.7 (HNN=CMe), –74.6 (N=CH), –61.9 (N=CMe). Found, %: C 59.31; H 4.07; N 19.38; S 8.60.  $\text{C}_{18}\text{H}_{15}\text{N}_5\text{O}_2\text{S}$ . Calculated, %: C 59.16; H 4.14; N 19.17; S 8.78.**

**X-Ray analysis of compound IIIa.** Single crystals suitable for X-ray analysis were obtained by slow evaporation of a solution of compound IIIa in acetonitrile at a constant temperature. Rhombic crystals with the following unit cell parameters:  $a = 20.911(4)$ ,  $b = 6.841(1)$ ,  $c = 24.241(5)$  Å;  $V = 3467.7(1)$  Å<sup>3</sup>; space group *Pbcn*;  $Z = 8$ ;  $d_{\text{calc}} = 1.23$  g/cm<sup>3</sup>. The structure was solved by the direct methods with subsequent Fourier syntheses using SHELXS-97 program [22] and was refined by the least-squares procedure in full-matrix anisotropic approximation for all non-hydrogen atoms using SHELXL-97 program [23]. The coordinates of hydrogen atoms were determined experimentally and were refined in isotropic approximation. The complete set of crystallographic data was deposited to the Cambridge Crystallographic Data Center (entry no. CCDC 648092).

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