# *N*-Substituted amides of 2-cyanopenta-2*E*,4-dienoic acid

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*N*-Substituted amides of 2-cyanopenta-2*E*,4-dienoic acid were synthesized by condensation of *N*-substituted cyanoacetamides with acrolein in a dioxane—DMSO solution in the presence of $Zn(OAc)_2 \cdot 2H_2O$ as a condensing agent. X-ray diffraction study of 3-methylanilide of 2-cyanopenta-2*E*,4-dienoic acid (**2d**) demonstrated that the crystal structure of this compound is similar to that of 2-cyanopenta-2*E*,4-dienoic acid studied previously. However, the presence of the *meta*-tolyl substituent in molecule **2d** apparently results in the fact that the β-structure, which is typical of 2-cyanopenta-2*E*,4-dienoic acid, does not exist in the crystalline phase of **2d**.

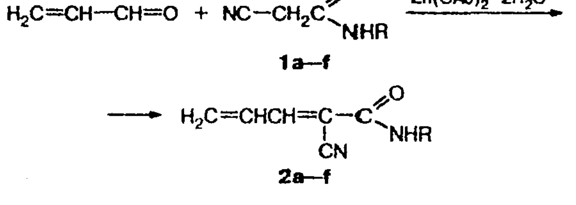
**Key words:** *N*-substituted cyanoacetamides, *N*-substituted amides of 2-cyanopenta-2*E*,4-dienoic acid, X-ray diffraction study, molecular structure, crystal structure, IR spectra, Raman spectra, hydrogen bonds.

Intermolecular hydrogen bonds with two competing basic centers, *viz.*, with carbonyl O atoms and N atoms of cyano groups, are among the important structure-forming factors governing the crystal systems of 2-cyanoacrylic[1] and 2-cyanopenta-2*E*,4-dienoic[2] acids and their esters.[2,3] Apparently, the type and strength of these bonds determine to a large extent the physical properties of these compounds and the possibility of the realization of conditions favorable for solid-phase topochemical conversions. Besides, these H-autocomplexes can be considered as models of intermediate states in studies of the reactivity of these compounds.[4]

As part of continuing studies of α-unsaturated 2-cyanocarboxylic acids, we used *N*-substituted cyanoacetamides (**1**) as starting compounds for preparing the previously unknown *N*-substituted amides of 2-cyanopenta-2*E*,4-dienoic acid (**2**). For compounds **2**, various types of H bonds would be expected to occur.

Monomeric amides **2** are of interest as cross-linking comonomers for preparing thermostable polymeric products (for example, in adhesive cyanoacrylate compositions). Previously, structures analogous to **2** were introduced as terminal and side groups into polymers with different structures with the aim of imparting thermoreactivity to these products.[5]

Amide **2a** was prepared by condensation of *N*-methylcyanoacetamide with acrolein in a phosphate buffer at pH 7. In other cases, the reactions were carried out in a dioxane—DMSO mixture with the use of $Zn(OAc)_2 \cdot 2H_2O$ as a condensing agent.



R = Me (**a**), Ph (**b**), 4-$MeC_6H_4$ (**c**), 3-$MeC_6H_4$ (**d**), 4-$ClC_6H_4$ (**e**), *cyclo*-$C_6H_{11}$ (**f**)

The compositions and structures of amides **2** were confirmed by the data of elemental analysis and IR spectroscopy (Table 1) as well as by the results of NMR spectroscopy. The spectral data for compounds **2a**—**f** are

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**Table 1.** Yields, melting points, data of elemental analysis, and IR spectra of amides **2**

| Compound | Yield (%) | M.p./°C | Found / Calculated (%) C | H | N | Molecular formula | IR, ν/cm$^{-1}$ C=C | Amide I | $C\equiv N$ | NH | Benzene ring |
|---|---|---|---|---|---|---|---|---|---|---|---|
| **2a** | 17.2 | 109—110 | 61.96<br>61.75 | 6.03<br>5.92 | 20.65<br>20.58 | $C_7H_8N_2O$ | 1574,<br>1613 | 1674 | 2216 | 3300 | — |
| **2b** | 43.4 | 108—110 | 72.87<br>72.71 | 5.09<br>5.08 | 14.13<br>14.13 | $C_{12}H_{10}N_2O$ | 1575,<br>1614 | 1673 | 2226 | 3313 | 1600 |
| **2c** | 28.6 | 103—104 | 73.51<br>73.56 | 5.85<br>5.70 | 12.83<br>13.20 | $C_{13}H_{12}N_2O$ | 1574,<br>1613 | 1684 | 2225 | 3330 | 1598 |
| **2d** | 32.3 | 86—87 | 73.43<br>73.56 | 5.79<br>5.70 | 13.32<br>13.20 | $C_{13}H_{12}N_2O$ | 1574,<br>1614 | 1683 | 2225 | 3345 | 1600 |
| **2e*** | 20.2 | 143—145 | 61.68<br>61.94 | 3.92<br>3.90 | 11.97<br>12.04 | $C_{12}H_9ClN_2O$ | 1573,<br>1612 | 1670 | 2223 | 3350 | 1597 |
| **2f** | 38.3 | 103—105 | 70.75<br>70.55 | 8.02<br>7.89 | 13.77<br>13.72 | $C_{12}H_{16}N_2O$ | 1575,<br>1607 | 1655 | 2218 | 3285 | — |

* Cl: found 15.01%; calculated 15.24%.

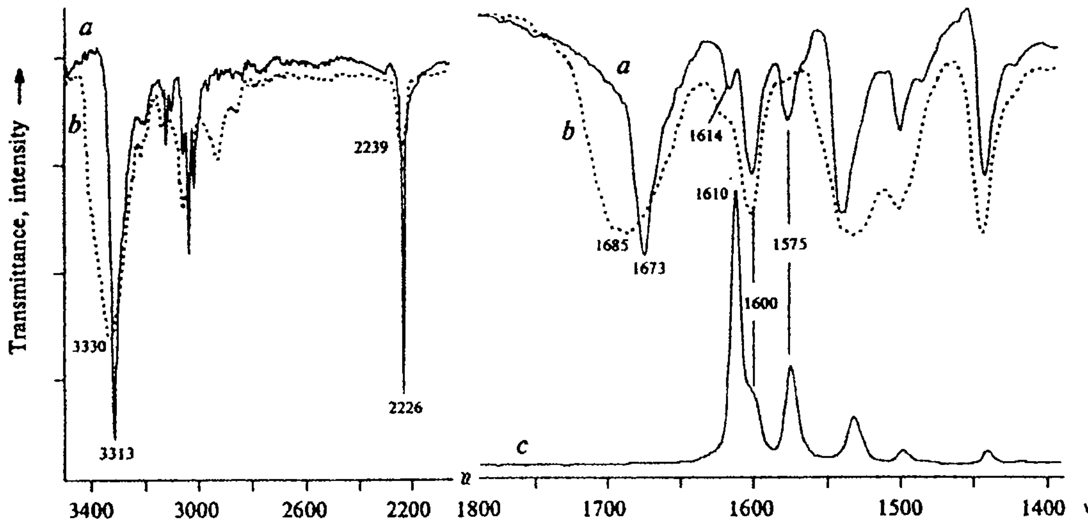

**Fig. 1.** IR (*a* and *b*) and Raman spectra (*c*) of compounds **2b**; *a* and *c*, the initial amide **2b**; *b*, amide **2b** after heating to 150 °C.

similar, which allows the conclusions that these compounds as a whole have similar structures and, in particular, the configurations about the $\Delta^2$-bonds are identical.

The regions of the IR and Raman spectra of amides **2b**, typical of compounds **2**, are shown in Fig. 1. The IR spectra of amides **2a—f**, which were measured according to a special procedure[6] for eliminating the effect of moisture, have characteristic absorption bands of NH groups (3300—3350 cm$^{-1}$), CN groups (2216—2226 cm$^{-1}$), and the butadiene fragment (1573—1575 and 1607—1615 cm$^{-1}$). Note that the νCN frequencies in the spectra of amides **2** are approximately 15—25 cm$^{-1}$ lower than those observed in the spectrum of compound **1**, which is apparently attributable to conjugation between the CN and C=C groups. The νNH bands in the spectra of amides **2** are narrowed and noticeably shifted to the long-wavelength region (by ~100 cm$^{-1}$) compared to the absorption bands of "free" NH groups.[7] Undoubtedly, this is indicative of the participation of the NH groups of compound **2** in hydrogen bonding. The $^1H$ and $^{13}C$ NMR spectra of amides **2a—f** are analogous to those of free 2-cyanopenta-2*E*,4-dienoic acid.[2,8]

An interesting characteristic feature of amides **2a—f** is their high solubility in organic solvents (including hexane). Using compound **2b** as an example, it was demonstrated by differential scanning calorimetry (Fig. 2) that amides **2** are stable up to their melting points, but they are, apparently, polymerized in melts. Actually, the IR spectrum of amide **2b** measured after heating of its specimen to 150 °C had no bands corresponding to

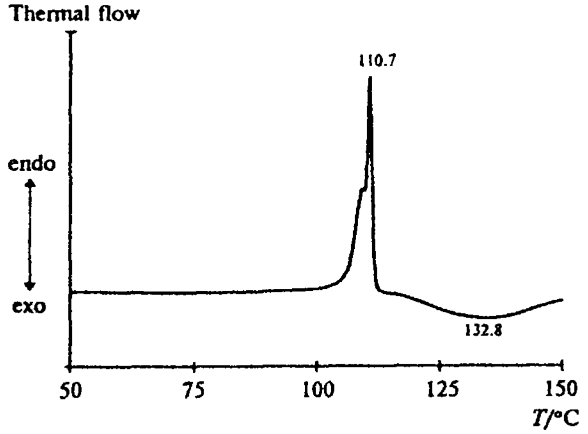

**Fig. 2.** Differential scanning calorimetry curve for amide **2b**.

**Table 2.** Coordinates (×10$^4$) and equivalent isotropic thermal parameters ($U_{eq}$×10$^3$) of nonhydrogen atoms in the structure of **2d**

| Atom | *x* | *y* | *z* | $U_{eq}$/Å$^2$ |
|---|---|---|---|---|
| N(1) | 1232(3) | 3700(2) | 8788(2) | 58(1) |
| N(2) | 512(3) | −472(3) | 11350(3) | 84(1) |
| O(1) | 3216(3) | 5034(2) | 9177(2) | 82(1) |
| C(1) | 2389(3) | 3737(3) | 9438(3) | 55(1) |
| C(2) | 2632(3) | 2081(3) | 10514(3) | 52(1) |
| C(3) | 3971(3) | 1881(4) | 11020(3) | 60(1) |
| C(4) | 4410(4) | 334(4) | 12014(3) | 68(1) |
| C(5) | 5837(4) | 194(5) | 12397(4) | 95(1) |
| C(6) | 1455(4) | 654(3) | 10985(3) | 60(1) |
| C(7) | 822(4) | 5050(3) | 7690(3) | 54(1) |
| C(8) | −695(4) | 4901(3) | 7412(3) | 61(1) |
| C(9) | −1147(4) | 6135(4) | 6314(3) | 69(1) |
| C(10) | −50(5) | 7558(4) | 5496(3) | 85(1) |
| C(11) | 1448(5) | 7707(4) | 5783(3) | 93(1) |
| C(12) | 1915(4) | 6486(4) | 6859(3) | 76(1) |
| C(13) | −2814(4) | 5949(4) | 6021(3) | 95(1) |

$\nu$C=C vibrations, while the bands corresponding to $\nu$C=O, $\nu$CN, and $\nu$NH vibrations were broadened and hypsochromically shifted by ~15 cm$^{-1}$ (see Fig. 1). These spectral patterns for amides **2** agree with the results of X-ray diffraction study of compound **2d** (Figs. 3 and 4; Tables 2—4).

As in the case of 2-cyanopenta-2*E*,4-dienoic acid (**3**) and its ethyl ester (**4**) studied recently,[2] the $\Delta^2$-bond in molecule **2d** has an *E* configuration (see Fig. 3) and all lengths of the corresponding bonds in the molecules of these compounds are identical within the experimental error. The 2-cyanopentadiene fragment in the structure of **2d** is substantially flattened in spite of the presence of shortened intramolecular contacts: H(4)...C(6), 2.61 Å (2.87 Å; hereinafter, the sums of the van der Waals radii are given in parentheses[9]); and H(1)...C(6), 2.38 Å. The maximum deviation (0.07 Å) from the plane passing through all nonhydrogen atoms of the above-mentioned fragment is observed for the C(1) atom.

The benzene C(7)...C(12) ring in molecule **2d** is noticeably rotated with respect to the C(1)—N(1) bond (the C(1)—N(1)—C(7)—C(12) torsion angle is 17.0(4)°), apparently due to the presence of the following shortened intramolecular contacts: O(1)...C(12), 2.85 Å (3.00 Å); O(1)...H(12), 2.28 Å (2.45 Å); and C(1)...H(12), 2.77 Å.

In the crystal of **2d** (Fig. 4), the molecules are linked in centrosymmetric cyclic dimers through an N—H...N intermolecular hydrogen bond: N(1)—H(1)...N(2)′ (−*x*, −*y*, 2 − *z*) (H(1)...N(2)′, 2.42 Å; N(1)...N(2)′, 3.198(4) Å; the N(1)—H(1)...N(2)′ angle is 152°). This is typical of structures of substituted 2-cyanoacrylamides.[10,11] In the structure of **2d**, these dimers are linked in corrugated layers through C—H...O intermolecular hydrogen bonds: C(3)—H(3)...O(1)″ (1 − *x*, 1 − *y*, 2 − *z*) (H(3)...O(1)″, 2.63 Å; C(3)...O(1)″, 3.502(3) Å; the C(5)—H(5A)...O(1)″ angle is 157°). In

**Table 3.** Principal bond lengths (*d*) in the structure of **2d**

| Bond | *d*/Å | Bond | *d*/Å | Bond | *d*/Å | Bond | *d*/Å |
|---|---|---|---|---|---|---|---|
| N(1)—C(1) | 1.350(3) | C(1)—C(2) | 1.485(3) | C(4)—C(5) | 1.322(4) | C(9)—C(10) | 1.384(4) |
| N(1)—C(7) | 1.410(3) | C(2)—C(3) | 1.331(3) | C(7)—C(8) | 1.377(3) | C(9)—C(13) | 1.511(4) |
| N(2)—C(6) | 1.140(3) | C(2)—C(6) | 1.433(3) | C(7)—C(12) | 1.392(3) | C(10)—C(11) | 1.367(4) |
| O(1)—C(1) | 1.220(3) | C(3)—C(4) | 1.437(4) | C(8)—C(9) | 1.385(4) | C(11)—C(12) | 1.370(4) |

**Table 4.** Principal bond angles (ω) in the structure of **2d**

| Angle | ω/deg | Angle | ω/deg | Angle | ω/deg | Angle | ω/deg |
|---|---|---|---|---|---|---|---|
| C(1)—N(1)—C(7) | 127.9(2) | C(3)—C(2)—C(1) | 120.7(2) | C(8)—C(7)—C(12) | 119.2(3) | C(10)—C(9)—C(13) | 120.5(3) |
| O(1)—C(1)—N(1) | 123.4(3) | C(6)—C(2)—C(1) | 120.1(2) | C(8)—C(7)—N(1) | 118.1(2) | C(8)—C(9)—C(13) | 120.9(3) |
| O(1)—C(1)—C(2) | 120.3(2) | C(2)—C(3)—C(4) | 126.6(3) | C(12)—C(7)—N(1) | 122.6(3) | C(11)—C(10)—C(9) | 119.6(3) |
| N(1)—C(1)—C(2) | 116.3(2) | C(5)—C(4)—C(3) | 123.2(3) | C(7)—C(8)—C(9) | 121.6(3) | C(10)—C(11)—C(12) | 122.2(3) |
| C(3)—C(2)—C(6) | 119.2(3) | N(2)—C(6)—C(2) | 179.4(3) | C(10)—C(9)—C(8) | 118.6(3) | C(11)—C(12)—C(7) | 118.8(3) |

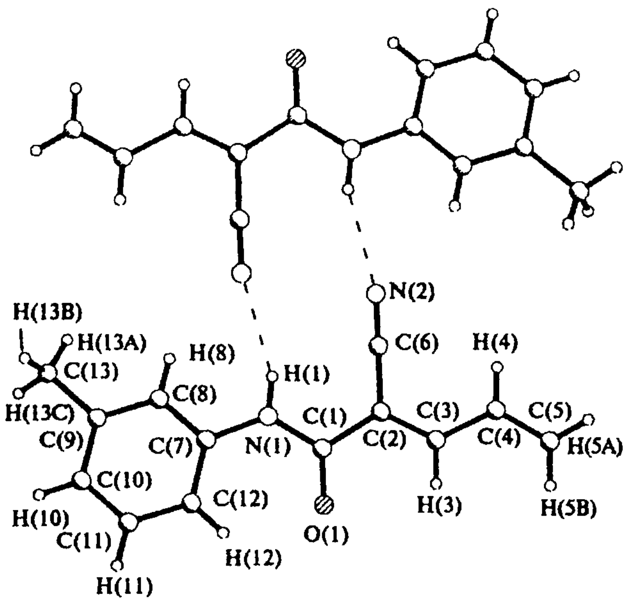

**Fig. 3.** Centrosymmetric dimer **2d**.

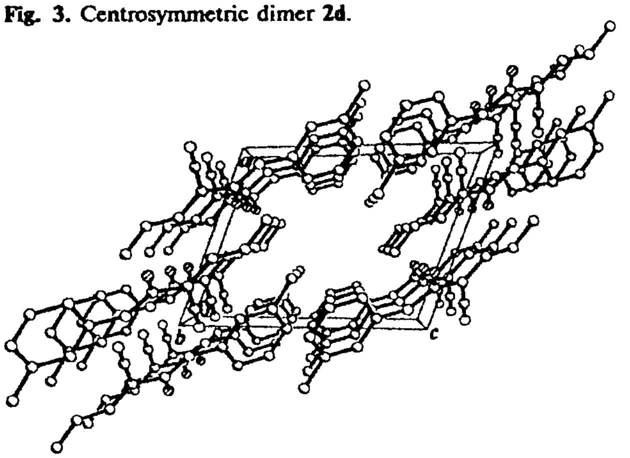

**Fig. 4.** Crystal structure of compound **2d**.

addition, the molecules in the crystal of **2d** form translation stacks along the direction (010) (see Fig. 4).

On the whole, the crystal structure of compound **2d** is very similar to that of compound **3**.[2] However, the presence of the relatively bulky *meta*-tolyl substituent in molecule **2d** apparently leads to a substantial increase in the distance between stacks and, consequently, the β structure is not formed in the crystal of **2d**, unlike the crystals of compounds **3** and **4**.[2,12] As a result, conditions for topochemical solid-phase reactions are not realized in compound **2d**.

## Experimental

The IR spectra of solid samples were recorded on a Perkin—Elmer 1725-FTIR instrument equipped with a Perkin—Elmer PEDR diffuse-reflection attachment and a modified sample holder.[6] The Raman Fourier spectra excited with a Nd:YAG laser (1064 nm) were obtained on a Perkin—Elmer 1725-X FTIR Raman Attachment instrument equipped with an InGaAs detector (operating at 3500—200 cm$^{-1}$) at 4 cm$^{-1}$ resolution.

The $^1$H (400.26 MHz) and $^{13}$C (100.68 MHz) NMR spectra were recorded on a Bruker AMX-400 instrument in acetone-d$_6$.

Amide **2b** was analyzed by differential scanning calorimetry on a Perkin—Elmer-7 instrument in the temperature programming mode at a rate of 10 K min$^{-1}$.

**Amide 2a.** A 1 *M* KOH aqueous solution was added to a solution of *N*-methylcyanoacetamide (4.95 g, 50 mmol) in a 1 *M* $K_2HPO_4$ aqueous solution (35 mL) to pH 7. Then freshly distilled acrolein (5 mL, 75 mmol) was added at 2—5 °C over 20 min. The mixture was stirred at 10 °C for 1 h and then at ~20 °C for 4 h. The precipitate was separated, washed with cold water, dried in a vacuum desiccator over $P_2O_5$, and crystallized from heptane.

**Amides 2b—f.** A solution of freshly distilled acrolein (5 mL, 75 mmol) in dioxane (5 mL) was added dropwise to a mixture of the corresponding cyanoacetamide **1** (50 mmol) and $Zn(OAc)_2 \cdot 2H_2O$ (5 g) in dioxane (40 mL) containing DMSO (4 mL) at 5 °C over 20 min. The mixture was stirred at 10 °C for 1 h, at 20 °C for 3 h, and at 30 °C for 2 h and then cooled to 10 °C. Then concentrated HCOOH (1 mL) and a 1 : 1 toluene—ether mixture (50 mL) were added. The reaction mixture was poured into 5 *M* HCl (50 mL). The precipitate (unconsumed cyanoacetamide and oligomeric products) and the organic layer were separated. The aqueous layer was extracted with ether (2×50 mL). The extracts were combined with the organic layer and dried over $MgSO_4$. The solvent was removed *in vacuo*. The residue was crystallized from heptane.

**Methylamide of 2-cyanopenta-2*E*,4-dienoic acid (2a).** $^1$H NMR, δ: 3.12 (d, 3 H, CH$_3$, $J$ = 6.3 Hz); 6.20 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 9.84 Hz); 6.36 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.6 Hz); 7.08 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 11.4 Hz, $J_{H(4),H(5A)}$ = 9.84 Hz, $J_{H(4),H(5B)}$ = 16.6 Hz); 7.70 (br.s, 1 H, NH); 8.07 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 11.4 Hz). $^{13}$C NMR, δ: 26.99 (CH$_3$); 110.12 (C(6)); 115.26 (C(2)); 132.84 (2 C, C(3), C(5)); 152.14 (C(4)); 161.07 (C(1)).

**Anilide of 2-cyanopenta-2*E*,4-dienoic acid (2b).** $^1$H NMR, δ: 5.99 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 10.0 Hz); 6.15 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.6 Hz); 6.89 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 11.3 Hz, $J_{H(4),H(5A)}$ = 10.0 Hz, $J_{H(4),H(5B)}$ = 16.6 Hz); 7.15 (m, 1 H, H arom.); 7.35 (m, 2 H, H arom.); 7.73 (d, 2 H, H arom., $J$ = 7.8 Hz); 7.95 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 11.3 Hz); 9.32 (br.s, 1 H, NH). $^{13}$C NMR, δ: 110.41 (C(6)); 114.31 (C(2)); 132.33 (C(3)); 132.86 (C(5)); 152.13 (C(4)); 159.14 (C(1)); 120.81, 124.78, 128.92, 138.40 (C arom.).

***p*-Methylanilide of 2-cyanopenta-2*E*,4-dienoic acid (2c).** $^1$H NMR, δ: 2.29 (s, 3 H, CH$_3$); 6.05 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 10.0 Hz); 6.18 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.7 Hz); 6.99 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 11.3 Hz, $J_{H(4),H(5A)}$ = 10.0 Hz, $J_{H(4),H(5B)}$ = 16.7 Hz); 7.27 (d, 2 H, H arom., $J$ = 8.1 Hz); 7.58 (d, 2 H, H arom., $J$ = 8.1 Hz); 7.94 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 11.3 Hz); 8.55 (br.s, 1 H, NH). $^{13}$C NMR, δ: 20.24 (CH$_3$); 110.42 (C(6)); 114.36 (C(2)); 132.34 (C(3)); 132.71 (C(5)); 151.99 (C(4)); 158.93 (C(1)); 120.87, 129.36, 134.28, 135.86 (C arom.).

***m*-Methylanilide of 2-cyanopenta-2*E*,4-dienoic acid (2d).** $^1$H NMR, δ: 2.31 (s, 3 H, CH$_3$); 5.98 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 10.0 Hz); 6.13 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.7 Hz); 6.88 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 11.3 Hz, $J_{H(4),H(5A)}$ = 10.0 Hz, $J_{H(4),H(5B)}$ = 16.7 Hz); 6.96 (d, 1 H, H arom., $J$ = 7.5 Hz); 7.22 (m, 1 H, H arom.); 7.53 (d, 2 H, H arom., $J$ = 8.9 Hz); 7.93 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 11.3 Hz); 9.21 (br.s, 1 H, NH).

***p*-Chloroanilide of 2-cyanopenta-2*E*,4-dienoic acid (2e).** $^1$H NMR, δ: 6.01 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 10.0 Hz); 6.16 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.7 Hz); 6.89 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 11.3 Hz, $J_{H(4),H(5A)}$ = 10.0 Hz, $J_{H(4),H(5B)}$ = 16.7 Hz); 7.37 (d, 2 H, H arom., $J$ = 8.9 Hz); 7.76 (d, 2 H, H arom., $J$ = 8.9 Hz); 7.94 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 11.3 Hz); 9.43 (br.s, 1 H, NH).

**Cyclohexylamide of 2-cyanopenta-2*E*,4-dienoic acid (2f).** $^1$H NMR, δ: 1.98—2.28 (m, 10 H, *cyclo*-Hex); 4.00 (br.s, 1 H, *cyclo*-Hex); 6.16 (d, 1 H, H(5A), $J_{H(5A),H(4)}$ = 10.0 Hz); 6.31 (d, 1 H, H(5B), $J_{H(5B),H(4)}$ = 16.7 Hz); 7.06 (ddd, 1 H, H(4), $J_{H(4),H(3)}$ = 12.2 Hz, $J_{H(4),H(5A)}$ = 10.0 Hz, $J_{H(4),H(5B)}$ = 16.7 Hz); 7.55 (br.s, 1 H, NH); 8.03 (d, 1 H, H(3), $J_{H(3),H(4)}$ = 12.2 Hz).

**X-ray diffraction study of compound 2d.** Crystals suitable for X-ray diffraction study were grown by isothermal evaporation of a solution of compound **2d** in hexane. The crystals of $C_{13}H_{12}N_2O$ are triclinic at 293 K, $a$ = 7.959(2) Å, $b$ = 8.055(4) Å, $c$ = 10.578(4) Å, $\alpha$ = 70.92(3)°, $\beta$ = 67.76(3)°, $\gamma$ = 81.07(3)°, $V$ = 592.9(4) Å$^3$, crystal dimensions 0.4×0.3×0.2 mm, space group $P\bar{1}$, $Z$ = 2, $d_{calc}$ = 1.189 g cm$^{-3}$, $F(000)$ = 224, $\mu$ = 0.077 mm$^{-1}$.

The intensities of 3054 reflections (2852 independent reflections, $R_{int}$ = 0.049) were measured on an automated four-circle Enraf-Nonius CAD-4 diffractometer (graphite monochromator, Mo-Kα radiation, θ/2θ scanning technique, $2\theta_{max}$ = 56°).

The structure was solved by the direct method with the use of the SHELXTL PLUS program package.[13] The positions of the hydrogen atoms were located from the difference electron density synthesis and refined using the riding model with fixed values of $U_{iso} = nU_{eq}$ of the nonhydrogen atoms to which the hydrogen atoms are attached ($n$ = 1.5 for the methyl group and 1.2 for the remaining hydrogen atoms). The full-matrix least-squares refinement based on $F^2$ with anisotropic thermal parameters (146 parameters) for nonhydrogen atoms using 2793 reflections converged to $R_1$ = 0.046 (for 877 reflections with $F > 4\sigma(F)$), $wR_2$ = 0.141, $S$ = 0.92. The coordinates of nonhydrogen atoms in the structure of **2d** are given in Table 2. The bond lengths and bond angles are listed in Tables 3 and 4, respectively.

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## References

1. V. E. Shklover, Yu. T. Struchkov, I. I. Kandror, I. R. Gol'ding, and Yu. G. Gololobov, *Zh. Strukt. Khim.*, 1991, **32**, 139 [*J. Struct. Chem. (USSR)*, 1991, **32** (Engl. Transl.)].
2. O. Ya. Borbulevych, O. V. Shishkin, I. R. Gol'ding, V. N. Khrustalev, and Yu. G. Gololobov, *Izv. Akad. Nauk, Ser. Khim.*, 1998, 1991 [*Russ. Chem. Bull.*, **47**, 1935 (Engl. Transl.)].
3. V. N. Khrustalev, O. V. Shishkin, S. V. Lindeman, Yu. T. Struchkov, M. A. Galkina, and Yu. G. Gololobov, *Izv. Akad. Nauk, Ser. Khim.*, 1996, 2288 [*Russ. Chem. Bull.*, 1996, **45**, 2172 (Engl. Transl.)].
4. Yu. G. Gololobov and W. Gruber, *Usp. Khim.*, 1997, **66**, 1054 [*Russ. Chem. Rev.*, 1997, **66** (Engl. Transl.)].
5. UK Pat. 1374464, 1974.
6. A. N. Shchegolikhin and O. L. Lazareva, *Int. J. Vibr. Spectrosc.*, 1997, **1**, 98.
7. J. S. Nowick, V. Antonovich, G. Noronha, and J. W. Ziller, *J. Org. Chem.*, 1995, **60**, 1888.
8. I. R. Gol'ding, P. V. Petrovskii, and Yu. G. Gololobov, *Izv. Akad. Nauk, Ser. Khim.*, 1998, 364 [*Russ. Chem. Bull.*, 1998, **47**, 356 (Engl. Transl.)].
9. Yu. V. Zefirov and P. M. Zorkii, *Usp. Khim.*, 1989, **58**, 713 [*Russ. Chem. Rev.*, 1989, **58** (Engl. Transl.)].
10. H. D. Block, H. D. Lockenhoff, and R. Allmann, *Cryst. Struct. Commun.*, 1975, **4**, 77.
11. O. Durruthy, R. Pomes, F. Fajardo, and A. Dago, *Acta Crystallogr.*, 1993, **C49**, 558.
12. G. M. J. Schmidt, *J. Chem. Soc.*, 1964, **B**, 2014.
13. G. M. Sheldrick, *SHELXTL PLUS. PC Version, A System of Computer Programs for the Determination of Crystal Structure from X-ray Diffraction Data, Revision 5.02*, Siemens Analytical X-ray Instruments Inc., Göttingen (Germany), 1994.

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