to IIb according to the data from the PMR, IR, and UV spectra. PMR spectrum ($D_2SO_4$), δ: 2.7 ppm (s, $CH_3$).

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# RESEARCH IN THE 2,5-DIARYL-1,3,4-OXADIAZOLE SERIES.

## 1. ELECTRONIC STRUCTURES AND SPECTRAL-LUMINESCENCE PROPERTIES OF SUBSTITUTED 2,5-DIPHENYL-1,3,4-OXADIAZOLES

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UDC 547.793.4.07:535.37:540.14.5:543.42.52

The dependence of the spectral-luminescence properties of substituted 2,5-diphenyl-1,3,4-oxadiazoles on the electronic nature of the substituents was studied. Experimental and theoretical studies confirm the electron-acceptor character of the oxadiazole ring and constitute evidence for the rather effective transmission of the electronic effects through the heteroring.

The intense fluorescence and short lifetimes of the excited states of the 2,5-diaryl-substituted 1,3,4-oxadiazoles have been responsible for their use in scintillation technology and as the active media of lasers. The interest in the study of the dependence of the spectral-luminescence properties on the electronic structures of molecules in this series of compounds is linked with this fact. Despite the considerable number of studies devoted to the structures and properties of 2,5-diaryl-1,3,4-oxadiazoles (see reviews [1, 2]), the character of the electronic interactions between the heteroring and the π systems of the aromatic substituents has not been adequately studied. There is no unified opinion regarding the electronic nature of the oxadiazole ring itself in these molecular systems. Thus, in addition to data on the aromatic character of the heteroring [3, 4], there is information that makes it possible to regard it to a great extent as a conjugated diene [5, 6], whereas the results obtained in [7] lead to the conclusion that appreciable interaction of the π systems of the substituents through the heteroring is absent.

Studies of the effect of the substituents with identical and different electronic natures in the para positions of the phenyl rings of 2,5-diphenyl-1,3,4-oxadiazole (PPD) on the spectral-luminescence properties of the compounds (Table 1) are continued in the present paper. The dipole moments in the ground and first excited singlet states were measured for some of them, and quantum-chemical calculations of the absorption spectra and the electron-density distributions were made.

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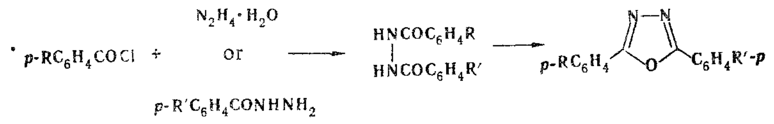
TABLE 1. Spectral Characteristics of 2-(p-R'-Phenyl)-5-(p-R-phenyl)-1,3,4-oxadiazoles

| Compound | R | R' | Absorption | | Luminescence | |
|---|---|---|---|---|---|---|
| | | | $\lambda_{max}$, nm | $\varepsilon \cdot 10^{-4}$ | $\lambda_{max}$, nm | η |
| 1 | H | $OCH_3$ | 298 | 2,88 | 360, 365 | 0,44 |
| 2 | H | $N(CH_3)_2$ | 335 | 3,60 | 395 | 0,64 |
| 3 | $OCH_3$ | $OCH_3$ | 300 | 3,27 | 355, 370 | 0,52 |
| 4 | $OCH_3$ | $N(CH_3)_2$ | 335 | 3,80 | 390 | 0,50 |
| 5 | $N(CH_3)_2$ | $N(CH_3)_2$ | 350 | 5,23 | 400 | 0,68 |
| 6 | H | $COOCH_3$ | 300 | 2,37 | 365 | 0,37 |
| 7 | $COOCH_3$ | $COOCH_3$ | 300 | — | 360 | 0,36 |
| 8 | H | COCl | 312 | 2,44 | 375 | 0,10 |
| 9 | COCl | COCl | 315 | — | 395 | 0,15 |
| 10 | H | $NO_2$ | 317 | 1,92 | Does not luminesce | |
| 11 | $NO_2$ | $NO_2$ | 320 | — | Does not luminesce | |
| 12 | H | $SO_2CHF_2$ | 300 | 2,40 | Weak luminescence | |
| 13 | $NO_2$ | $SO_2CHF_2$ | 310 | 3,10 | Does not luminesce | |
| 14 | $OCH_3$ | $COOCH_3$ | 310 | 3,27 | 380 | 0,54 |
| 15 | $OCH_3$ | COOH | 315 | — | 387 | 0,57 |
| 16 | $OCH_3$ | COCl | 330 | 2,31 | 415 | 0,54 |
| 17 | $OCH_3$ | 3,4-$(CO)_2O$ | 338 | 2,13 | 440 | 0,51 |
| 18 | $OCH_3$ | $NO_2$ | 337 | 1,84 | Does not luminesce | |
| 19 | $N(CH_3)_2$ | $COOCH_3$ | 297, 355 | 1,32; 2,28 | 450. | 0,69 |
| 20 | $N(CH_3)_2$ | 3,4-$(CO)_2O$ | 310, 360 | — | 500 | 0,40 |
| 21 | $N(CH_3)_2$ | $NO_2$ | 307, 390 | 3,08; 1,22 | 565 | 0,15 |

TABLE 2. Dipole Moments of Substituted 2,5-Diphenyl-1,3,4-oxadiazoles

| Compound | $\mu_g$, D | $\mu_e$, D | $\varphi^0$ |
|---|---|---|---|
| 1 | 3,78 | 6,60 | 30 |
| 2 | 5,15 | 11,60 | 30 |
| 3 | 4,00 | 6,70 | 0 |
| 5 | 5,00 | 9,8 | 0 |
| 6 | 3,56 | 4,6 | 53 |
| 7 | 3,70 | 2,5 | 0 |
| PPD | 3,55 | 3,55 | 0 |

The synthesis of mono- and disubstituted PPD was realized via a scheme that includes acylation of hydrazine hydrate or monoaroylhydrazines with substituted benzoyl chlorides and subsequent cyclodehydration of the resulting N,N'-diaroylhydrazines in the presence of dehydrating agents ($POCl_3$, $SOCl_2$).



Three bands usually appear in the absorption spectra of solutions of 2,5-diaryl-substituted 1,3,4-oxadiazoles in hexane. The low-intensity second (middle) band may be overlapped by the more intense short-wave band. The long-wave (first) band is due to a $S_0 \rightarrow S_1$ transition of the ππ* type delocalized in the π-electron system of the entire molecule [4, 8, 9]. This band, which is the most sensitive to structural changes in the molecule and is responsible for the luminescence properties, is the chief subject of discussion in our paper.

In toluene PPD has a long-wave absorption band with $\lambda_{max}$ 284 nm (ε $2.6 \cdot 10^4$) and luminescence in the UV region ($\lambda_{max}$ 365 nm). The experimental value of the dipole moment in the ground state ($\mu_g$) is 3.5 D and is in quite good agreement with the value (2.8 D) calculated by the CNDO/2 (complete neglect of differential overlap) method. The calculations show that the magnitude and direction of the dipole moment are determined chiefly by the charge distribution within the heteroring. The validity of this conclusion is also confirmed by the dipole moment of oxadiazole, which is 3.04 D [10]. The dipole moment measured by the method

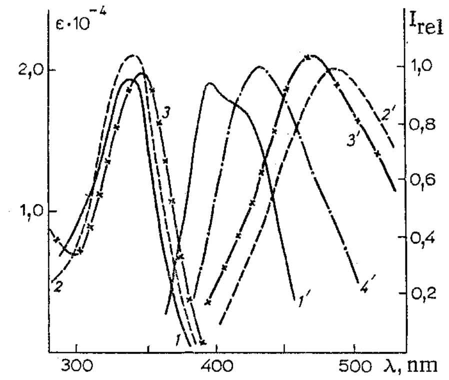


Fig. 1. Absorption (1-3) and luminescence (1'-4') spectra of compound No. 2 [R = H, R' = $N(CH_3)_2$] in dioxane (1), dimethyl sulfoxide (2), acetonitrile (3), and dichloroethane (4).

of spectral shifts remains virtually unchanged on passing to the first excited singlet state ($\mu_e$) (Table 2).

The introduction of an electron-donor substituent — a methoxy or a dimethylamino group — into the PPD molecule in the para position of the phenyl ring gives rise to a bathochromic shift of the long-wave absorption and luminescence bands vis-à-vis an increase in the molar extinction coefficient ($\varepsilon$) and retention of the high quantum yields ($\eta$). The introduction of a second electron-donor substituent in the same position of the other phenyl group leads to a further bathochromic shift of the bands and an increase in $\varepsilon$ (Table 1, compounds Nos. 1-5). The indicated effects are intensified as the donor properties of the substituent become more pronounced. It should be noted that a significant solvatochromic effect in the absorption and particularly in the luminescence is observed for compound No. 2 [R = H, R' = $N(CH_3)_2$] (Fig. 1).

The dipole moments of compounds Nos. 1-5 in the ground state increase as compared with unsubstituted PPD (Table 2). Calculations show that these changes are due to an increase in the electron charge in the heteroring. Since the dipole moment of the oxadiazole ring is directed along the second-order axis toward the nitrogen atoms, the increase in the moment when electron-donor substituents are introduced constitutes evidence for the electron-acceptor character of the oxadiazole ring. The dipole moments of compounds Nos. 1-5 increase significantly in the excited state: calculations show transfer of electron density from the donor substituent and the phenyl ring bonded to it to the heteroring and the second phenyl group.

A bathochromic shift of the long-wave absorption band is also observed in the absorption spectra of compounds Nos. 6, 8, 10, and 12, which have electron-acceptor substituents; however, the effect of these substituents is not as strongly expressed as the effect of electron-donor substituents (Table 1). A further long-wave shift of the absorption maximum, which is displayed more distinctly in the case of the unsymmetrical disubstituted compound (No. 13) and is extremely weak in the case of symmetrical disubstituted compounds, is observed when a second electron-acceptor substituent is introduced (compounds Nos. 7, 9, 11, and 13). Compounds with electron-acceptor substituents luminesce. As in most cases, the introduction of a nitro group as a substituent quenches the luminescence.

The measurement of the dipole moments of most of the compounds with electron-acceptor substituents is difficult because of their low solubilities (for $\mu_g$) and relatively small spectral shifts (for $\mu_e$). With respect to their magnitude, the measured dipole moments of compounds Nos. 6 and 7 differ only slightly from the moment of unsubstituted PPD (Table 2).

The results of quantum-chemical calculations of the absorption spectra are in good agreement with the experimental data for all of the compounds except the dimethylamino derivatives, for which the calculated values of the energies of the transitions are shifted 0.3 eV hypsochromically relative to the experimental values; this is evidently associated with insufficient allowance for the $\sigma$-donor effect of the methyl groups in the standard parametrization of the Pariser–Parr–Pople (PPP) method.

An examination of the absorption spectra of compounds Nos. 14-21 with sutstituents with different electronic natures is most interesting. The maxima of their long-wave band are shifted bathochromically as compared with monosubstituted PPD and disubstituted (with the same substituents) PPD; this shift is greater, the greater the difference in the substituents with respect to their electronic effect (Table 1). An interesting feature of this series of compounds is the significant decrease in the extinction coefficient of the long-wave absorption

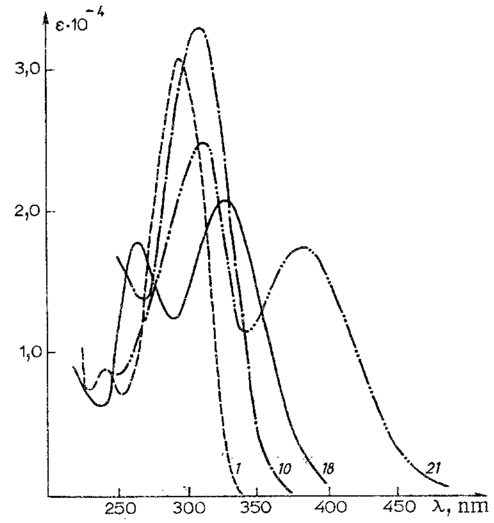
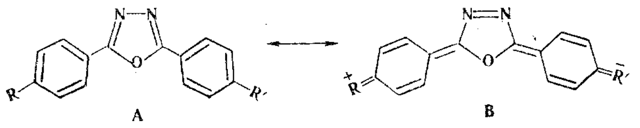


Fig. 2. Absorption spectra of compounds Nos. 1, 10, 18, and 21 in ethanol.

band vis-à-vis intensification of the acceptor properties of the substituents; this is particularly distinctly appreciable in the series of compounds Nos. 19-21, which contain a dimethylamino group as the fixed substituent. A second (middle) band, the intensity of which, on the contrary, increases as the acceptor properties of the substituents become more pronounced (Table 1), simultaneously appears distinctly in the absorption spectra of compounds Nos. 19-21. The maximum of this band is shifted bathochromically as the electron-acceptor or electron-donor properties of the substituents become more pronounced: whereas for the PPD molecule the second band shows up in the form of a shoulder (230-240 nm) on the descending part of the short-wave band, in the case of compound No. 21 [R = $N(CH_3)_2$, R' = $NO_2$] this band has a maximum at 310 nm with ε 30,000 (Fig. 2). Nurmukhametov and Nagornaya [8], who examined the nature of the short-wave bands of naphthyl- and biphenylyl-substituted 1,3,4-oxadiazoles, assigned the second band to electron transitions localized in the π systems of the naphthalene and biphenyl rings. The problem of the nature of the short-wave bands of disubstituted PPD requires further examination.

Principles similar to those noted in the examination of the absorption spectra are observed in the luminescence spectra of the investigated compounds. The introduction of substituents with different electronic natures shifts the luminescence markedly to the long-wave region. Thus whereas PPD emits light in the UV region, compound No. 21 is a luminophore with orange-red luminescence. The quantum yields in the series of compounds with a dimethylamino group (19-21) decrease as the acceptor properties of the second substituent become more pronounced.

The described spectral properties, viz., the shifts of the absorption and luminescence bands in the case of PPD derivatives with substituents with identical and different electronic natures, constitute evidence that the oxadiazole ring is a rather effective conductor of electronic effects. An analysis of the quantum-chemical calculations of the distribution of the charges and the bond orders (Table 3) shows that upon excitation the electron density is transferred from the donor substituent to the acceptor substituent along the conjugation chain, which includes the —C=N—N=C— atoms of the oxadiazole ring (the C=N bond orders decrease, and the N—N bond orders increase; the C—O bonds do not participate in conjugation). Thus the



results obtained in this research constitute evidence for a substantial contribution of resonance structure B, i.e., for the diene character of the heteroring.

## EXPERIMENTAL

The absorption spectra of toluene solutions of the compounds were recorded with an SF-4 spectrophotometer. The luminescence spectra were recorded with a device consisting of a ZMR-3 mirror monochromator, an FÈU-18 optical emission detector, and an M-95 microammeter. The

TABLE 3. Charges and Bond Orders in 2-(p-R'-Phenyl)-5-(p-R-phenyl)-1,3,4-oxadiazoles

| R | R' | State | Bond orders | | | | | | | Charges on R and R' | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | N—N | $C_{(5)}$—N | $C_{(2)}$—N | $C_{(5)}$—O | $C_{(2)}$—O | $C_{(5)}$—Ph | $C_{(2)}$—Ph | R | R' |
| H | H | $S_0$ | 0,535 | 0,740 | 0,740 | 0,459 | 0,459 | 0,297 | 0,297 | — | — |
| | | $S_1$ | 0,682 | 0,540 | 0,540 | 0,354 | 0,354 | 0,481 | 0,481 | — | — |
| H | $NH_2$[a] | $S_0$ | 0,531 | | 0,734 | 0,454 | 0,461 | 0,298 | 0,310 | — | +0,246 |
| | | $S_1$ | 0,634 | | 0,580 | 0,374 | 0,348 | 0,419 | 0,493 | — | +0,401 |
| H | COOH[a] | $S_0$ | 0,537 | 0,739 | 0,740 | 0,462 | 0,457 | 0,297 | 0,299 | — | −0,045 |
| | | $S_1$ | 0,668 | 0,524 | 0,524 | 0,386 | 0,375 | 0,438 | 0,501 | — | −0,088 |
| $NH_2$ | $NH_2$ | $S_0$ | 0,527 | 0,738 | 0,738 | 0,456 | 0,456 | | | +0,243 | +0,243 |
| | | $S_1$ | 0,629 | 0,596 | 0,596 | 0,356 | 0,356 | | | +0,324 | +0,324 |
| $NH_2$ | COOH | $S_0$ | 0,534 | 0,732 | 0,747 | 0,463 | 0,452 | 0,312 | 0,301 | +0,248 | −0,047 |
| | | $S_1$ | 0,636 | 0,605 | 0,593 | 0,364 | 0,381 | 0,463 | 0,453 | +0,400 | −0,097 |

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[a] In the calculations the $N(CH_3)_2$ and $COOCH_3$ groups are modeled by the $NH_2$ and COOH groups, respectively.

photoluminescence was excited with an SVDSh-500 lamp, from the spectrum of which light with a wavelength of 365 nm was isolated by means of a DMR-4 quartz monochromator. The absolute luminescence quantum yields of toluene solutions ($\eta$) were determined by the equal absorption method.

The dipole moments in the ground state ($\mu_g$) were determined in dilute benzene solutions by the dielectric method with the aid of the Hedestrand extrapolation formula [11]; the solutions were thermostatted during the measurements. The method of spectral shifts in the modification proposed by N. G. Bakhshiev [12, 13] was used for the determination of the dipole moments in the $S_1$ excited state ($\mu_e$).

For molecules that have luminescence in liquid solutions the computational formulas have the form: $\mu_e = \sqrt{\mu_g^2 - c_2 a^3}$

$$\cos\varphi = \frac{1}{2\mu_g\mu_e}\left[(\mu_g^2+\mu_e^2) - \frac{\Delta c^{a-f}}{2c_2}(\mu_g^2-\mu_e^2)\right];$$

for nonluminescing molecules

$$\mu_e \cos\varphi = \mu_g - \frac{c_1^a a^3}{2\mu_g},$$

where $\varphi$ is the angle between the $\mu_g$ and $\mu_e$ vectors, $a$ is the Onsager radius of the molecule, and $c_1^a$, $c_2$, and $c^{a-f}$ are the parameters of the theory of the universal intermolecular interactions determined from the shift of the absorption and luminescence spectra in different solvents [13]. The Onsager radius was determined from the structural formulas of the molecules. Calculations by the method of spectral shifts were made with a Nairi computer from specially composed programs. The error in the experimental determination of $\mu_e$ reaches 30%, as compared with 40-50% in the case of small spectral shifts.

The quantum-chemical calculations were made by means of the standard Pariser–Parr–Pople (PPP) method with allowance for the interactions of 20 singly excited configurations [14, 15]. Resonance integrals $\beta$ were calculated and varied over each interval of Mulliken iteration [16]. The two-center integrals of one-electron interactions were calculated from the Ono–Dewar formula [17]. The bond lengths were optimized by the method in [18]. The components of the dipole moment were calculated as

$$M_x = 4{,}80298\sum_\mu (D_\mu - P_{\mu\mu} - Z_{\text{av}})(x_\mu - x_{\text{av}});\ Z_{\text{av}} = \frac{\sum\limits_i g_i - \sum\limits_\mu D_\mu}{n};\ x_{\text{av}} = \frac{\sum\limits_\mu x_\mu}{n},$$

where $g_i$ is the number of electrons in the i-th MO, and n is the number of atoms in the molecule. Correspondingly, the magnitude of the dipole moment

$$|\vec{M}| = \sqrt{M_x^2 + M_y^2}.$$

TABLE 4. Substituted 2,5-Diphenyl-1,3,4-oxadiazoles

| Compound | mp, °C | Solvent for purification | Found, % | | | Empirical formula[a] | Calc., % | | | Yield, % |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | C | H | N | | C | H | N | |
| 6 | 178 | Ethanol | 68,4 | 4,1 | 10,3 | $C_{16}H_{12}N_2O_3$ | 68,6 | 4,3 | 10,0 | 74 |
| 8 | 159—160 | Benzene | 63,6 | 3,3 | 9,9 | $C_{15}H_9ClN_2O_2$ | 63,4 | 3,2 | 9,8 | 74 |
| 12 | 230—232 | Ethanol | 53,8 | 3,3 | 8,7 | $C_{15}H_{10}F_2N_2O_3S$ | 53,6 | 3,0 | 8,3 | 81 |
| 13 | 220—222 | Acetic acid | 47,4 | 2,2 | 10,8 | $C_{15}H_9F_2N_3O_5S$ | 47,2 | 2,4 | 11,0 | 80 |
| 14 | 185 | Ethanol | 65,6 | 4,7 | 9,3 | $C_{17}H_{14}N_2O_4$ | 65,8 | 4,5 | 9,0 | 79 |
| 15 | 255—260 | Ethanol | 64,7 | 4,3 | 9,5 | $C_{16}H_{12}N_2O_4$ | 64,9 | 4,0 | 9,5 | 68 |
| 16 | 190 | Ethanol | 61,4 | 3,3 | 8,7 | $C_{16}H_{11}ClN_2O_2$ | 61,1 | 3,5 | 8,9 | 82 |
| 17 | 241—243 | Chlorobenzene | 63,5 | 3,2 | 8,7 | $C_{17}H_{10}N_2O_5$ | 63,3 | 3,1 | 8,7 | 83 |
| 19 | 205 | Ethanol | 67,1 | 5,0 | 13,3 | $C_{18}H_{17}N_3O_3$ | 66,9 | 5,2 | 13,0 | 68 |
| 20[b] | 185 | Acetic acid | 61,5 | 4,3 | 11,8 | $C_{18}H_{11}N_3O_3$ | 61,2 | 4,2 | 11,9 | 54 |
| 21 | 257—258 | Acetic acid | 62,2 | 4,6 | 17,8 | $C_{16}H_{14}N_4O_3$ | 61,9 | 4,5 | 18,1 | 71 |

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[a] The compositions of compounds Nos. 8, 12, and 16 were also confirmed by determination of the percentage of halogen. [b] Acylation was carried out at 0-5°C to avoid reaction at the anhydride grouping.

The total moment was determined as the sum of the π and σ components $\vec{M} = \vec{M}_\pi + \vec{M}_\sigma$, and the σ moments were calculated by the method in [19].

It is known that such systems with a large number of heteroatoms in the ring present the greatest complexity for semiempirical calculations of the dipole moments and electron-density distribution [20]; in particular, allowance for the penetration integrals leads to overstated π moments in furans [19]. The results of our calculations of the dipole moments in the ground state by the PPP method also proved to be overstated as compared with the experimentally obtained values. The calculation of unsubstituted PPD by the CNDO/2 method made it possible to obtain a correction of –2.2 D for the dipole moments ($\mu_g$) of the given series.

Symmetrical 1,2-Diaroylhydrazines. Equimolar amounts (0.02 mole) of the substituted benzoyl chloride and hydrazine hydrate in pyridine was heated gradually in the course of 3 h with stirring to 90°C, after which the mixture was cooled and poured into a mixture of ice and water, and the resulting precipitate was removed by filtration.

Unsymmetrical 1,2-Diaroylhydrazines. A saturated solution of sodium carbonate was added with vigorous stirring to a mixture of solutions of 0.02 mole of the substituted benzoic acid hydrazide in 5% hydrochloric acid and 0.02 mole of the substituted benzoyl chloride in benzene until the mixture had pH ∿ 9, after which it was stirred for 1 h. It was then neutralized with dilute hydrochloric acid, and the precipitate was removed by filtration and dried.

1,3,4-Oxadiazole Derivatives. The 1,2-diaroylhydrazines were refluxed in phosphorus oxychloride (1:10) until they dissolved completely, after which the solution was refluxed for another 30 min. It was then cooled and poured over ice, and the resulting precipitate (if a solution was obtained, it was first neutralized with ammonium hydroxide) was removed by filtration, washed with water until the washings were neutral, and dried. The product was purified by crystallization from the appropriate solvents. Compounds Nos. 7-9 were obtained by the method in [21]; compound No. 16 was obtained from corresponding acid 15 by refluxing in thionyl chloride.

4-[2-(p-Methoxyphenyl)-1,3,4-oxadiazol-5-yl]phthalic Anhydride (17). A mixture of 1.7 g (0.01 mole) of p-methoxybenzhydrazide and 2.1 g (0.01 mole) of 4-chloroformylphthalic anhydride in 50 ml of chlorobenzene was refluxed until hydrogen chloride evolution ceased, after which the precipitate was removed by filtration and dried. The solid was refluxed in 30 ml of thionyl chloride until it had dissolved completely, after which the solution was refluxed for another hour. The thionyl chloride was evaporated to dryness *in vacuo* (with a water aspirator), 30 ml of benzene was added, and the solvent was removed by distillation until the volume was two thirds of the original value. The residue was poured into petroleum ether, and the precipitate was removed by filtration, washed with benzene, and dried to give 2.7 g

(83%) of product. Successive crystallization from acetic acid and chlorobenzene gave a product with mp 241-243°C.

The constants of the compounds described above were in agreement with the literature data: Compound No. 1 had mp 146-147°C (mp 146°C [22]), No. 2 had mp 142-143°C (mp 143°C [23]), No. 3 had mp 162°C (mp 161-162°C [24]), No. 4 had mp 187°C (mp 187°C [25]), No. 5 had mp 225-227°C (mp 225-227°C [26]), No. 7 had mp 264-265°C (mp 265°C [21]), No. 9 had mp 196°C (mp 195-196°C [21]), No. 10 had mp 209°C (mp 207-209°C [22]), No. 11 had mp 314-315°C (mp 315°C [27]), and No. 18 had mp 228°C (mp 227-228°C [27]).

The melting points, results of elementary analysis, yields, and solvents for purification of the compounds obtained for the first time are presented in Table 4.

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