

PYRIMIDOTETRATHIAFULVALENES

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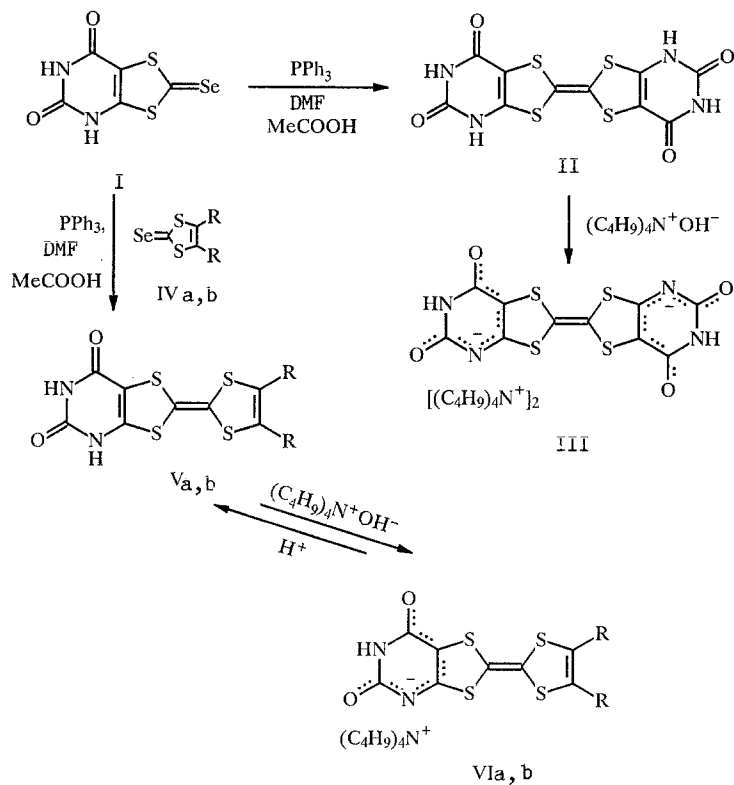
Bis[2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene has been synthesized by dimerization of 5,7-dioxo(4H,6H)-1,3-dithiolo[4,5-d]pyrimidine-2-selenone in the presence of triphenylphosphine. By dimerization of a mixture of 5,7-dioxo(4H,6H)-1,3-dithiolo[4,5-d]pyrimidine-2-selenone and 4,5-dimethyl- or 4,5-ethylenedithio-1,3-dithio-2-selenones, the corresponding asymmetric tetrathiafulvalenes have been obtained. By alkylation of the tetrabutylammonium salts of 2,4-dioxo(1H,3H)pyrimidotetrathiafulvalenes, their N-alkyl and N,N'-dialkyl derivatives have been obtained. It has been shown by means of cyclic voltammetry that the introduction of the 2,4-dioxo(1H,3H)-pyrimido system increases the oxidation potential of the tetrathiafulvalene derivative by 0.3-0.35 V.

Tetrathiafulvalenes are the principal starting materials in obtaining high-conductivity crystals, including those with superconducting properties [1, 2]. A large number of substituted tetrathiafulvalenes have now been synthesized, including compounds with condensed heterocycles; however, pyrimidotetrathiafulvalenes have not yet been described. It was mentioned in [3] that pyrimidotetrathiafulvalenes were obtained, but we have not been able to find, either in [3] or in subsequent publications, any description of the methods of synthesis or the properties of the compounds. We are particularly interested in 2,4-dioxo(1H,3H)-, 2-amino-4-oxo(3H)-, 4-amino-2-oxo(1H)-, and 4-aminopyrimidotetrathiafulvalenes, containing structural elements of uracil, guanine, cytosine, or adenine that are capable of forming specific intermolecular hydrogen bonds. The formation of such bonds may have a specific influence on the properties of crystals of ion-radical salts of such a type as the tetrathiafulvalenes, in the direction of a closer packing of molecules and increased conductivity.

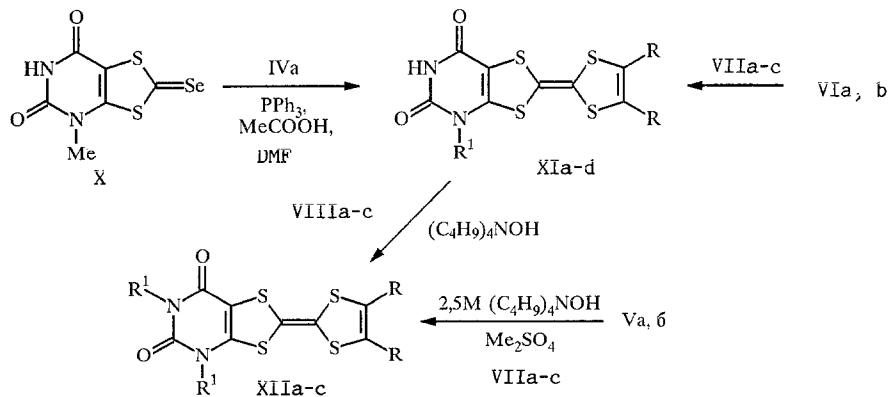
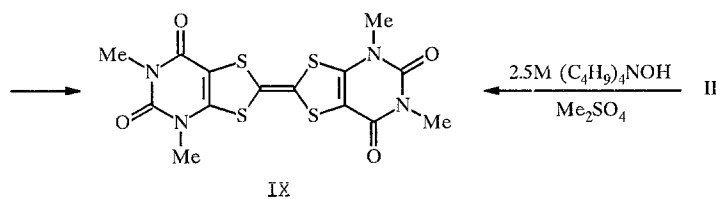
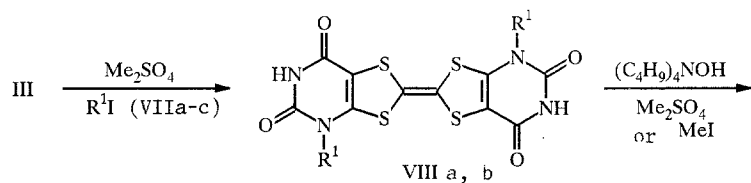
Here we report on the synthesis and investigation of properties of 2,4-dioxo(1H,3H)pyrimidotetrathiafulvalenes. The starting substance in obtaining tetrathiafulvalenes of this new type was 5,7-dioxo(4H,6H)-1,3-dithiolo[4,5-d]pyrimidine-2-selenone (I). The selenone I does not react with triethyl phosphite in benzene solution at temperatures up to 80°C. In a triethyl phosphite solution, I enters into reaction at a temperature above 100°C, but no individual products can be isolated; the reaction proceeds in a different direction, and no tetrathiafulvalene is formed. For dimerization of 1,3-dithio-2-selenones, triphenylphosphine is also used [4]. It has been established that the selenone I in DMF solution in the presence of triphenylphosphine and acetic acid forms bis[2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene (II) with a yield of about 25%. The reaction proceeds at room temperature. Raising the temperature to 80°C shortens the reaction time but does not increase the yield of the desired tetrathiafulvalene II. This method has also been applied in obtaining asymmetric 2,4-dioxopyrimidotetrathiafulvalenes, as has been shown in the example of compounds Va, b, obtained from the selenone I and the corresponding 4,5-dimethyl- and 4,5-ethylenedithio-1,3-dithio-2-selenones IVa, b. In order to suppress the formation of the symmetric tetrathiafulvalene II, the selenones IVa, b were taken in twofold excess. With such ratios of the original compounds, no formation of II was observed. The tetrathiafulvalenes Va, b were formed with yields of 25-35%.

Compound II is a difficultly soluble substance, dissolving hardly at all in boiling DMF. Compounds Va, b are more soluble; they can be crystallized from DMF, but the substance that is recovered from the reaction mixture contains impurities that are difficult to remove by crystallization. Therefore, the purification of Va, b is better performed through their tetrabutylammonium salts VIa, b by crystallization or column chromatography on silica gel [for VIb, $R_f = 0.24$, eluent $\text{CH}_3\text{CN}-\text{CH}_3\text{OH}-(\text{C}_4\text{H}_9)_4\text{NOH} = 3:1:0.05$]. The salts III and VIa, b are extremely stable compounds. They are difficultly soluble in water, readily soluble in ethanol, dioxane, or chloroform; in the presence of water they are partly hydrolyzed.

The salts III and VIa, b can be alkylated in acetonitrile by dimethyl sulfate or alkyl iodides VII (methyl, octyl, or hexadecyl iodide). The respective N-alkyl derivatives VIIIa, b or XIa-d are obtained with yields of 70-90%. Further alkylation of VIIIa and XIa-c in the presence of tetrabutylammonium hydroxide leads to the tetraalkyl (IX) and dialkyl derivatives XIIa-c, which are also formed by alkylation of the corresponding II and Va, b in the presence of 2.5 moles of tetrabutylammonium hydroxide.



IV-VIa R = Me, b RR = S-(CH₂)₂-S



VII, VIII a R¹ = Me, b R¹ = C₈H₁₇, c R¹ = C₁₆H₃₃; XI, XII a R = R¹ = Me, b RR = S(CH₂)₂S
 R¹ = Me c. R = Me, R¹ = C₈H₇, d R = Me, R¹ = C₁₆H₃₃

TABLE 1. Spectral Characteristics of Derivatives of 2,4-Dioxo(1H,3H)pyrimidotetra-thiafulvalenes

Compound	IR spectrum, ν , cm^{-1} (1500-1800 and 2500-3600 cm^{-1})	PMR spectrum, δ , ppm (in DMSO- d_6)
II	1570, 1646, 1716, 1728, 3050, 3450	
III	1580...1626 br. i 2760, 2850, 2930, 3065, 3380	0,99 (24H, t, -CH ₃); 1,57 (32H, m, C-CH ₂); 3,10 (16H, m, N-CH ₂); 8,36 (2H, s, -NH)
Va	1575, 1640, 1687, 1700 sh 1715, 1724 sh 2815, 2984, 3134, 3450	1,89 (6H, s, -CH ₃); 11,31 & 11,90 (2H, s, -NH)
Vb	1578, 1641, 1685, 1698, 1730, 2790, 2900, 3045, 3450	3,33 (4H, s, -CH ₂ -); 11,4 and 12,0 (2H, s, -NH)
VIa	1600...1620 br. i 2800, 2915, 2935, 3010, 3100	0,98 (12H, t, CH ₂ -CH ₃); 1,57 (16H, m, -CH ₂ -); 1,9 (6H, s, S-C-CH ₃); 3,10 (8H, m, N-CH ₂); 8,13 (1H, s, -NH)
VIb	1582, 1609, 1627, 2800, 2930, 3080	0,98 (12H, t, CH ₂ -CH ₃); 1,55 (16H, m, -CH ₂ -); 3,10 (8H, m, N-CH ₂ -); 3,26 (4H, s, S-CH ₂ -); 7,92 (1H, s, -NH-)
VIIIa	1576, 1644, 1710, 2880, 3096, 3440	
VIIIb	1576, 1590, 1602, 1706, 2816, 2852, 2920, 3040, 4152	0,83 (6H, m, -CH ₃); 1,24 (24H, m, -CH ₂ -); 3,66 (4H, t, N-CH ₂ -); 11,74 (2H, s, -NH)
IX	1592, 1656, 1708, 2880, 2940, 2985	
XI a	1574, 1632, 1715, 1730, 2820, 2875, 3010, 3140	1,94 (6H, s, C-CH ₃); 3,27 (3H, s, -N-CH ₃); 11,9 (1H, s, -NH)
XI b	1576, 1638, 1655, 1712, 1725, 2820, 3024, 3105, 3160	3,27 (3H, s, -N-CH ₃); 3,37 (4H, s, S-CH ₂ -)
XI c	1572, 1644, 1656, 1680, 1708, 2810, 2860, 2940, 3010, 3150	0,85 (3H, m, -CH ₂ -CH ₃); 1,30 (12H, m, CH ₂ -); 11,95 (6H, s, >C-CH ₃); 3,74 (2H, t, N-CH ₂ -); 8,26 (1H, s, -NH)
XI d	1578, 1688, 1704, 2852, 2920, 3020, 3088, 3140	0,85 (3H, m, -CH ₂ -CH ₃); 1,30 (28H, m, CH ₂ -); 1,95 (6H, s, >C-CH ₃); 3,74 (2H, t, -N-CH ₂ -); 8,20 (1H, s, -NH)
XII a	1560, 1592, 1644, 1700, 2880, 2940, 2990	1,95 (6H, s, -C-CH ₃); 3,11 (3H, t, N ¹ -CH ₃); 3,32 (3H, s, N ³ -CH ₃)
XII b	1586, 1642, 1686, 1698, 2860, 2916	3,12 (3H, s, N ¹ -CH ₃); 3,32 (3H, s, N ³ -CH ₃); 3,36 (4H, s, S-CH ₂ -)
XII c	1592, 1654, 1694, 2852, 2920, 2956	0,85 (6H, m, -CH ₂ -CH ₃); 1,32 (24H, m, -CH ₂ -); 1,96 (6H, s, >C-CH ₃); 3,75 (4H, t, -N-CH ₂ -)

The N-methyl derivatives of symmetric tetrathiafulvalene VIIIa and IX are difficultly soluble; when they are refluxed in DMF, they partially decompose. The introduction of octyl or cetyl groups at the nitrogen atom in the molecule of the tetrathiafulvalene greatly increases the solubility of the compounds. The N-substituted dioxopyrimido derivatives are dibasic NH-acids; and when they are alkylated, they can form two isomers. In order to confirm the structures of the alkyl derivatives, compounds VIIIa and XIa were also obtained by countersynthesis, starting with the selenone X. PMR and IR spectra indicated that the compounds were identical. The synthesis and properties of the selenones I and X will be reported in a future communication.

The selenone X in the presence of triphenylphosphine forms the tetrathiafulvalene VIIIa with an extremely low yield (~1%), whereas the reaction $X \rightarrow XIa$ proceeds with a higher yield (~65%) than that in the reaction $I \rightarrow V$ (Table 1).*

In order to evaluate the electron-donor properties of the synthesized 2,4-dioxo(1H,3H)pyrimidotetra-thiafulvalenes and their derivatives, their oxidation potentials were determined by cyclic voltammetry in acetonitrile. Compounds having E_{ox}^1 in the interval ~0.62-0.92 V can undergo two-stage one-electron reversible oxidation. Compounds II, Va, b, VIIIa, and XIa, b are difficultly soluble, and their oxidation potentials can be determined only in solution in DMF or hexamethylphosphoramide (HMPA), where shifts of oxidation potentials to higher values are observed [5]. Therefore, for the more soluble compounds XIc, d and XIIa, b, the values of E_{ox} were determined in three solvents. It should be noted that when the change is made from acetonitrile to DMF, the values of E_{ox}^1 and E_{ox}^2 approach each other; and in HMPA, sometimes only a single stage of oxidation and reduction is observed. In view of the data of [6], we believe that this maybe related to solvation effects. From our results we can evaluate the influence of the 2,4-dioxo(1H,3H)pyrimido system on E_{ox} of the tetrathiafulvalenes. To do this, first let us turn our attention to E_{ox} of the N-monosubstituted compounds XI in acetonitrile: in comparison with E_{ox} of the tetrathiafulvalene, an increase of 0.25-0.3 V is observed. It is more correct to compare with the value of E_{ox} of 2,3-dimethyltetrathiafulvalene

*As in Russian original; Table 1 does not include any yield data — Translator.

TABLE 2. Cyclic Voltammetric Data on Synthesized Derivatives of 2,4-Dioxo-(1H,3H)pyrimidotetrathiafulvalenes

Compound	E_{ox}^1 E_{ox}^2 (V, in relation to Ag/AgCl) E_{red}^1 E_{red}^2		
	in CH ₃ CN	in DMF	in HMPA
II			0,8; 0,65
Va		0,62; 0,86; 0,54; 0,69	0,7; 0,58
Vb		0,75; 0,8; 0,61; 0,75	
VIIIa			0,75; 0,6
XIa		0,79; 1,0; 0,62; 0,83	
XIb		0,92; 1,04; 0,75	
XIc	0,7; 0,99; 0,61; 0,87	0,72; 0,96; 0,63; 0,85	0,86; 0,62
XId	0,73; 0,99; 0,63; 0,86	0,77; 0,95; 0,64; 0,84	
XIIa	0,65; 0,97; 0,58; 0,88	0,7; 0,88; 0,6; 0,78	
XIIb		0,83; 0,97; 0,72	
Tetra- thiaful- valene	0,4; 0,79; 0,31; 0,71	0,55; 0,81; 0,41; 0,67	0,59; 0,78; 0,44

[5], which is about 0.05 V lower. Then, the 2,4-dioxo(1H,3H)-pyrimido system accounts for 0.3-0.35 V. The introduction of the ethylenedithiol group into the tetrathiafulvalene system in place of two methyl groups increases E_{ox} by ~ 0.15 V [5]. In the case of compounds Va, b and XIa, b we observe approximately the same difference. All that remains is to explain why, for the compounds with free NH groups Va, b in DMF solution, lower values of E_{ox} are observed in comparison with the monoalkylated and dialkylated compounds XI and XII. We believe that this difference is related to the relatively high NH-acidity of the compounds and the formation of intermolecular hydrogen bonds with the solvent. Thus, it has been ascertained that the 2,4-dioxo(1H,3H)pyrimido- system has a considerable electron-acceptor character and that it hinders the oxidation of the tetrathiafulvalene derivative by 0.3-0.35 V, similar to the effect of two alkoxy carbonyl groups [5] (Table 2).

Because of the high values of E_{ox} of the new tetrathiafulvalenes, particularly the ethylenedithiol derivatives Vb and XIb, together with low solubility, it is difficult to obtain their charge-transfer complexes or ion-radical salts. The salts III and VI are oxidized much more readily. The charge-transfer complexes and cation-radical salts of the 2,4-dioxo(1H,3H)pyrimidotetrathiafulvalenes will be described in a separate communication.

EXPERIMENTAL

IR spectra were taken in a Specord 75-IR instrument on suspensions in white mineral oil or hexachlorobutadiene; PMR spectra were taken in a Bruker WH-90/DS spectrometer (90 MHz) with TMS internal standard; the electronic absorption spectra were taken in a Specord UV-Vis spectrometer. The cyclic voltammetric curves were taken in a PI-50-1 potentiostat with a three-electrode circuit. The working electrode was glassy graphite (0.27 cm²), and the comparison electrode was Ag/AgCl; the potential sweep rate was 200 mV/sec.

Bis[2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene (II, C₁₀H₄N₄O₄S₄) and Its Tetrabutylammonium Salt (III, C₄₂H₇₄N₆O₄S₄). A 2.08-g quantity of I (7.8 mmoles) was dissolved in 70 ml of dimethylformamide (DMF); 1 ml of acetic acid and 4.2 g (16 mmoles) of triphenylphosphine were added, and the mixture was allowed to stand for 24 h at 20°C. The precipitate was filtered off and washed with DMF. Obtained 0.4 g (28%) of a yellow-orange powder II, which was suspended in 15 ml of ethanol, after which 3.5 ml of a 10% aqueous solution of tetrabutylammonium hydroxide was added. The mixture was heated to 80°C, and 10 ml of water was added to the hot solution. After cooling, the precipitate was crystallized from 40 ml of acetonitrile. Obtained 0.69 g (30%) of yellow crystals of III with mp 237-238°C. Compound III (0.69 g) was dissolved in 25 ml of ethanol, and 3 ml of acetic acid was added. Obtained 0.3 g (25%) of II in the form of a yellow-orange powder; mp > 350°C.

[2,4-Dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (Va, C₁₀H₈N₂O₂S₄) and Its Tetrabutylammonium Salt (VIa, C₂₆H₂₃N₃O₂S₄). In 320 ml of DMF, 4.25 g (16 mmoles) of I and 6.7 g (32 mmoles) of IVa were dissolved, after which 4 ml of acetic acid and 25.15 g (96 mmoles) of triphenylphosphine in 50 ml of dimethylformamide were added. The mixture was heated for 20 min at 80°C and then left at 20°C for 6 h. After adding 0.5 liter of water, the precipitate was filtered off and washed with ethanol and benzene. The residue (~3.5 g) was dissolved in 130 ml of methanol containing 12 mmoles of tetrabutylammonium hydroxide; the solution was filtered, and 800 ml of ether was added to the filtrate. The precipitate was crystallized from 500 ml of acetonitrile. Obtained 3.2 g (37%) of yellow crystals of VIa with mp 197-198°C. A 3.0-g quantity of VIa was dissolved in 40 ml of ethanol, 5 ml of acetic acid was added, and the resulting precipitate was crystallized from 200 ml of DMF. Obtained 2.0 g (30%) in the form of orange crystals with mp >250°C (decomp.).

[2,4-Dioxo(1H,3H)pyrimido]ethylenedithiotetrathiafulvalene (Vb, C₁₀H₆N₂O₂S₆) and Its Tetrabutylammonium salt (VIb, C₂₆H₄₁N₃O₂S₆). In 100 ml of DMF, 1.4 g (5 mmoles) of IVb and 0.66 g (2.5 mmoles) of I were dissolved; 2 ml of acetic acid was added, and also a solution of 4.0 g (15 mmoles) of triphenylphosphine in 20 ml of DMF. The reaction mixture was held for 2 days at 20°C. The precipitate of bis(ethylenedithio)tetrathiafulvalene was filtered off (0.34 g, 35%); the filtrate was diluted with 0.5 liter of water. The precipitate was filtered off and washed with 100 ml of ethanol and 100 ml of hot benzene. The residue (~0.4 g) was dissolved in 12 ml of methanol containing 1.5 mmoles of tetrabutylammonium hydroxide, and the solution was filtered. The filtrate was diluted with 100 ml of ether. The red resinous substance that was obtained was crystallized by mulling with a small quantity of acetone. This mixture was filtered, and the residue was crystallized from 20 ml of acetone, obtaining 0.35 g (30%) of VIb in the form of yellow crystals with mp 83-85°C. A 0.24-g quantity of VIb was dissolved in 5 ml of ethanol, and 0.5 ml of acetic acid was added. The resulting precipitate was crystallized from 10 ml of DMF, obtaining 0.1 g (20%) of Vb in the form of fine orange crystals with mp >250°C (decomp.).

Bis[1-methyl-2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene (VIIIa, C₁₂H₈N₄O₄S₄). A 0.33-g quantity (0.38 mmole) of III was dissolved in 15 ml of absolute ethanol, and 0.12 ml (1.2 mmoles) of dimethyl sulfate was added. The mixture was held for 6 h at room temperature; the precipitate was filtered off and washed with DMF. Obtained 0.14 g (90%) of yellow-orange crystals of VIIIa with mp >300°C (decomp.).

Bis[1-octyl-2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene (VIIIb, C₂₆H₃₆N₄O₄S₄). A solution of 0.17 g (0.2 mmole) of III and 0.2 ml (1 mmole) of octyl iodide in 20 ml of acetonitrile was refluxed for 30 min. After cooling, the precipitate was filtered off, washed with acetonitrile, and crystallized from 4 ml of DMF. Obtained 0.1 g (93%) of yellow-orange crystals of VIIIb with mp >300°C.

Bis[1,3-dimethyl-2,4-dioxo(1H,3H)pyrimido]tetrathiafulvalene (IX, C₁₄H₁₂N₄O₄S₄). To 0.17 g (0.5 mmole) of II, 30 ml of methanol was added, the methanol containing 3 mmoles of tetrabutylammonium hydroxide and 0.4 ml (6 mmoles) of methyl iodide; the mixture was refluxed for 15 min and then left at 20°C for 6 h. The resulting precipitate was crystallized from DMF, obtaining 0.2 g (95%) of IX with mp >300°C.

[1-Methyl-2,4-dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (XIa, C₁₁H₁₀N₂O₂S₄). A. A 0.14-g quantity (0.25 mmole) of VIa was dissolved in 20 ml of acetonitrile with heating; 0.2 ml (3 mmoles) of methyl iodide was added, and the mixture was refluxed for 5 min. The precipitate that formed upon cooling was separated and then crystallized from 10 ml of dimethylformamide. Obtained 0.07 g (85%) of XIa with mp 256-260°C.

B. A 0.42-g quantity (1.5 mmoles) of X and 0.63 g (3 mmoles) of IVa were dissolved in 20 ml of DMF; 0.2 ml of acetic acid and 2.4 g (3 mmoles) of triphenylphosphine in 5 ml of DMF were added. The mixture was heated for 15 min at 70-80°C and then left at 20°C for 12 h. The precipitate was filtered off, washed with small quantities of DMF and benzene, and crystallized from 40 ml of DMF. Obtained 0.32 g (65%) of XIa with mp 256-260°C.

[1-Methyl-2,4-dioxo(1H,3H)pyrimido]ethylenedithiotetrathiafulvalene (XIb, C₁₁H₈N₂O₂S₆). A 0.12-g quantity (0.19 mmole) of VIb was dissolved in 25 ml of acetonitrile, 0.12 ml (12 mmoles) of dimethyl sulfate was added, and the mixture was refluxed for 5 min. The precipitate that formed upon cooling was separated and washed with acetonitrile. Obtained 0.06 g (80%) of XIb with mp 278-279°C.

[1-Octyl-2,4-dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (XIc, C₁₈H₂₄N₂O₂S₄). A 0.3-g quantity (0.5 mmole) of VIa and 0.3 ml (1.5 mmoles) of octyl iodide were dissolved in 20 ml of acetonitrile and refluxed for 10 min, then left for 12 h at 20°C. The resulting precipitate was crystallized from 50 ml of acetonitrile. Obtained 0.16 g (70%) of yellow-orange crystals of XIc with mp 236-239°C.

[1-Cetyl-2,4-dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (XId, C₂₆H₄₀N₂O₂S₄). A 0.84-g quantity (1.5 mmoles) of VIa and 0.53 g (3 mmoles) of cetyl iodide were dissolved in 50 ml of acetonitrile and refluxed for 30 min. The precipitate

that formed upon cooling was separated off and crystallized from 5 ml of DMF. Obtained 0.53 g (65%) of XIId in the form of orange crystals with mp 206-210°C.

[1,3-Dimethyl-2,4-dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (XIIa, C₁₂H₁₂N₂O₂S₄). A mixture of 0.09 g (0.28 mole) of Va, 0.2 ml (3 mmoles) of methyl iodide, and 10 ml of methanol, containing 0.8 mmole of tetrabutylammonium hydroxide, was refluxed for 30 min in 10 ml of absolute ethanol. After cooling the solution, the precipitate was separated and crystallized from acetone. Obtained 0.09 g (92%) of yellow-orange crystals of XIIa with mp 241-243°C.

[1,3-Dimethyl-2,4-dioxo(1H,3H)pyrimido]ethylenedithiotetrathiafulvalene (XIIb, C₁₂H₁₀N₂O₂S₆). To 0.19 g (0.5 mmole) of Vb in 20 ml of acetonitrile, 0.2 ml (2 mmoles) of dimethyl sulfate was added, along with 5 ml of methanol containing 1.5 mmoles of tetrabutylammonium hydroxide; the mixture was refluxed for 15 min. The precipitate was crystallized from 10 ml of acetone. Obtained 0.17 g (85%) of orange crystals of XIIb with mp 244-245°C.

[1,3-Dioctyl-2,4-dioxo(1H,3H)pyrimido]dimethyltetrathiafulvalene (XIIc, C₂₆H₄₀N₂O₂S₆). To 0.2 g (0.5 mmole) of Xc in 6 ml of acetonitrile, 0.2 ml (1 mmole) of octyl iodide was added, along with 3 ml of methanol containing 1 mmole of tetrabutylammonium hydroxide; the mixture was refluxed for 15 min. Upon cooling the solution, obtained 0.14 g (52%) of XIIc in the form of lustrous orange crystals with mp 114-116°C.

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