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2-PHENACYLBENZOTHIAZOLE IN THE BIGINELLI REACTION

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The cyclocondensation of 2-phenacylbenzothiazole with aromatic aldehydes and urea or thiourea in the Biginelli reaction gave previously unreported 4-aryl-5-(2-benzothiazolyl)-2-oxo-6-phenyl-1,2,3,4-tetrahydropyrimidines and their 2-thioxo analogs. An efficient procedure was developed for carrying out the three-component reaction at low temperatures. The structure peculiarities of the products were analyzed using IR, ^{1}H NMR, and X-ray diffraction data.

Keywords: aldehydes, urea, pyrimidines, thiourea, 2-phenacylbenzothiazoles, Biginelli reaction, cyclocondensation.

Recently, there has been renewed interest in the three-component cyclocondensation of ethyl acetoacetate with aldehydes and ureas (or thioureas) discovered by Biginelli in 1893 [1]. The reaction products are tetrahydropyrimidine derivatives, which display various types of biological activity and hold interest as modulators for the transport of calcium ions across the cell membrane [2]. The preparative scope of this reaction has been expanded by improving the procedure and variation of all three components. Thus, the standard procedure for this reaction involves heating the reagents in ethanol at reflux with hydrochloric acid catalysis and often gives product in low yield [2]. Better results are obtained in the presence of *p*-toluenesulfonic acid [3], boron trichloride etherate [4], and ethyl polyphosphorate [5] or by microwave irradiation [6]. Nitroacetone [12] and 2-acetonylpyrimidines [13] have recently begun to be used as the methylenecarbonyl component in addition to various esters, amides, and nitriles of β-keto carboxylic acids [2, 7-10] as well as phenylacetaldehyde and acetophenone [11]. The use of nitroacetone and 2-acetonylpyrimidines opens a pathway to obtain multifunctional pyrimidines.

We were the first to study the preparative scope of 2-phenacylbenzothiazole (**1**) in the Biginelli reaction and found simple conditions for good selectivity. Benzaldehydes **2a-f**, which contain electron-withdrawing or electron-donor substituents in the para, meta, or ortho positions, as well as heteroaromatic aldehydes **2i** and **2j**, which are π-electron-deficient or π-electron-rich heterocycles, were tried as the aldehyde component.

The cyclocondensation of compounds **1** and **2a-j** with urea leads to the formation of previously unreported 4-aryl-5-(2-benzothiazolyl)-2-oxo-6-phenyl-1,2,3,4-tetrahydropyrimidines **3a-j**. The reaction of compound **1** with *p*-nitrobenzaldehyde and thiourea gives the corresponding 2-thioxo derivative **4** (Scheme 1).

Carrying out the Biginelli reaction under the standard conditions (heating of the reagents in ethanol at reflux in the presence of hydrochloric acid) led to products containing impurities, which could not be eliminated even after multiple crystallization. A simple and efficient procedure was developed by varying the reaction conditions. Cyclocondensation in acetic acid with 1:1.1:3 mole ratio of the reagents at 40-50°C gave 72-97% yields. The reaction time was from 72 to 240 h. Longer reaction time was required with decreasing

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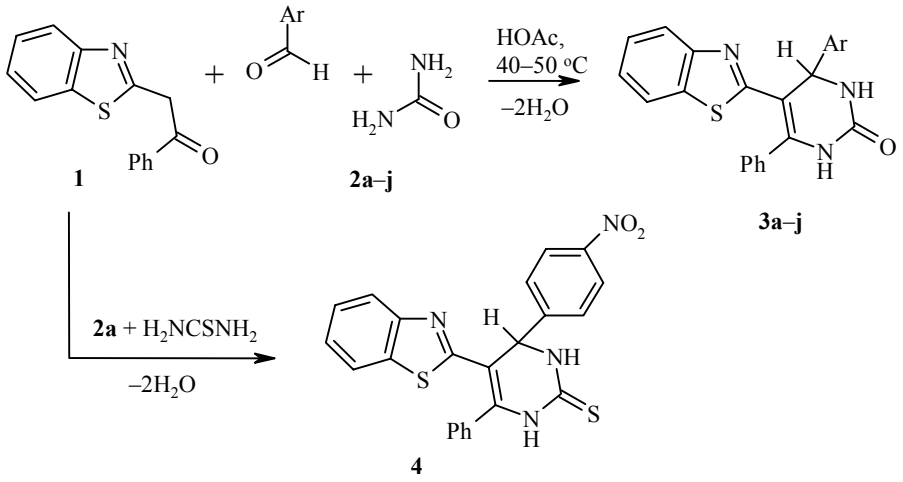
Scheme 1



2, **3 a** Ar = 4-$O_2NC_6H_4$; **b** Ar = 3-$O_2NC_6H_4$; **c** Ar = 2-$O_2NC_6H_4$; **d** Ar = 4-ClC_6H_4; **e** Ar = 2-ClC_6H_4; **f** Ar = Ph; **g** Ar = 4-$MeOC_6H_4$; **h** Ar = 4-$Me_2NC_6H_4$; **i** Ar = 4-pyridyl; **j** Ar = 2-thienyl

electrophilicity of the aldehyde used. A decrease in the urea fraction in the starting mixture and carrying out the reaction above 60°C lead to a significant decrease in the product yield. A three-fold excess of the dinucleophilic component, in our view, is necessary to make up the loss of this component due to side reactions such as hydrolytic decomposition. The use of mild conditions probably also prevents undesired transformations and provides the complete transformation of the reagents with simplified isolation of the products as pure compounds.

The products isolated are stable, high-melting crystalline compounds. Thioxo derivative **4** is yellow, nitro derivatives **3a-c** are pale yellow, while the other compounds are colorless. Some physicochemical indices of the pyrimidines synthesized are given in Table 1, while the IR and ^{1}H NMR spectral data are given in Table 2. The IR spectra of the products feature bands for the carbonyl group at 1690-1730 cm^{-1} and for the N–H groups at 3210-3220 and 3390-3450 cm^{-1}. Bands were found for the C=S group in compound **4** at 1540 and 1575 cm^{-1}.

More complete information on the structure of these products was provided by the ^{1}H NMR spectra. The doublet at 5.78-6.51 ppm corresponds to the vicinal proton at $C_{(4)}$ in the pyrimidine ring and permits us to assign the signals of the NH protons. The singlet for the proton at $N_{(1)}$ in compounds **3a-j** appears at 9.33-9.66 ppm, while the proton at $N_{(3)}$ appears as a doublet or broadened singlet (distortion of the doublet with small coupling constant) at 7.84-8.23 ppm. The corresponding protons in the compound **4** arise at 10.86 and 9.98 ppm. The signals for the protons attached to nitrogen atom disappear after the addition of D_2O, while the doublet for the proton at $C_{(4)}$ in the pyrimidine ring is converted to a singlet. The proton at $N_{(1)}$ appears at 0.88-1.65 ppm downfield relative to the proton at $N_{(3)}$, which probably indicates transfer of electron density from the $N_{(1)}$ atom through the vicinal $C_{(5)}$=$C_{(6)}$ fragment to the electron-withdrawing benzothiazole ring with formation of an energetically favorable conjugation system. The position of the signals for the protons at $N_{(1)}$, $N_{(3)}$, and $C_{(4)}$ atoms in the *m*- and *p*-phenyl derivatives **3a,b,d,f-h** shows a steady decrease in chemical shift in going from electron-withdrawing to electron-donor Ar substituents.

TABLE 1. Characteristics of Compounds **3a-j** and **4**

Compound	Empirical formula	Found, % / Calculated, %			mp, °C*	Reaction time, days	Yield, %
		C	H	N			
3a	$C_{23}H_{16}N_4O_3S$	64.39 / 64.47	3.85 / 3.76	12.94 / 13.08	288-291	3	91
3b	$C_{23}H_{16}N_4O_3S$	64.31 / 64.47	3.81 / 3.76	12.97 / 13.08	257-258	4	97
3c	$C_{23}H_{16}N_4O_3S$	64.62 / 64.47	4.91 / 3.76	13.11 / 13.08	306-308	5	83
3d	$C_{23}H_{16}ClN_3OS$	66.25 / 66.10	3.78 / 3.86	10.15 / 10.06	281-282.5	7	82
3e	$C_{23}H_{16}ClN_3OS$	66.22 / 66.10	3.73 / 3.86	10.21 / 10.06	296-297	10	96
3f	$C_{23}H_{17}N_3OS$	72.17 / 72.04	4.34 / 4.47	11.12 / 10.96	254-255.5	5	94
3g	$C_{24}H_{19}N_3O_2S$	69.59 / 69.71	4.72 / 4.63	10.22 / 10.16	247-248.5	7	77
3h	$C_{25}H_{22}N_4OS$	70.57 / 70.40	5.31 / 5.20	13.25 / 13.14	275-276	10	78
3i	$C_{22}H_{16}N_4OS$	68.88 / 68.73	4.15 / 4.20	14.43 / 14.58	314.5-316	3	77
3j	$C_{21}H_{15}N_3OS_2$	64.61 / 64.76	3.72 / 3.88	10.66 / 10.79	260-262	7	72
4	$C_{23}H_{16}N_4O_2S_2$	62.28 / 62.14	3.85 / 3.63	12.77 / 12.61	287-288	3	95

***** The water–DMF ratio was 1:2 for compound **3g**, 1:3 for **3c, 3e, 3h, 3i**, 1:4 for **3a**, and 1:5 for **3d**. The water–pyridine ratio was 1:2 for compound **4** and 1:4 for **3b, 3f,** and **3j**.

Let us examine the ^{1}H NMR signals for the benzothiazole protons. For most compounds, we uncoupled the signals corresponding to each ring proton, which appear mainly in narrow ranges at 7.74-7.80, 7.16-7.25, 7.32-7.37, and 7.69-7.75 ppm (in order from $C_{(4)}$ to $C_{(7)}$). In comparison, for example, with the corresponding signals of unsubstituted benzothiazole (8.23, 7.55, 7.55, and 8.12 ppm [14]), significant upfield shifts are noted for the protons at $C_{(5)}$ and $C_{(6)}$. These shifts may result from the electron-donor effect of $N_{(1)}$ atom or asymmetry in the shielding of the aryl substituents of the pyrimidine ring. However, we should note that the chemical shifts of the protons at $C_{(4)}$, $C_{(5)}$, $C_{(6)}$, or $C_{(7)}$ are virtually identical, that is, these shifts are quite independent on the nature of the substituents in the pyrimidine ring. These ^{1}H NMR spectral data for the benzothiazole protons would have been difficult to predict.

On the basis of previous studies of the mechanism of the Biginelli reaction [15], a different isomeric structure in agreement with the spectral data could not be eliminated. Thus, the structure of 5-(2-benzothiazolyl)-4-(*m*-nitrophenyl)-2-oxo-6-phenyl-1,2,3,4-tetrahydropyrimidine (**3b**) was finally established by X-ray diffraction analysis (Fig. 1, Tables 3 and 4).

It is interesting to note that the benzothiazole ring and the fragment of the pyrimidine ring comprising its vicinal bond and $N_{(1)}$ atom are found virtually in the same plane. This finding supports the conclusion that a conjugation system exists derived from analysis of the ^{1}H NMR spectra. The conjugation is energetically so favorable that the $C_{(6)}$-phenyl group is forced to rotate out of this plane due to steric hindrance. This rotation leads to loss of effective overlap of the π-orbitals of the phenyl substituent and the adjacent electron-withdrawing carbon atom of the double bond and the protons of the $C_{(6)}$–phenyl fragment appear in the ^{1}H NMR spectrum in a rather narrow range (7.45-7.66 ppm).

TABLE 2. Spectral Characteristics of Compounds **3a-j** and **4**

Compound	IR spectrum, ν, cm^{-1}		^1H NMR spectrum, δ, ppm (*J*, Hz)								
	C=X*	NH	$NH_{(1)}$, s	$NH_{(3)}$, br. s or d	$CH_{(4)}$, d	$CH_{(6)}$–Ph, m	$C_{(5)}$-benzothiazolyl				aromatic protons of substituent Ar, m
							$CH_{(4')}$, d	$CH_{(7')}$, d	$CH_{(6')}$, m	$CH_{(5')}$, m	
3a	1705	3210, 3420	9.66	8.23	6.04 (3.1)	7.54-7.65	7.77 (8.0)	7.72 (8.0)	7.34	7.22	7.83-8.29
3b	1700	3210, 3390	9.66	8.22	6.04 (3.0)	7.52-7.65	7.76 (8.6)	7.73 (8.6)	7.35	7.22	7.69-8.45
3c	1696	3210, 3430*[2]	9.64	8.09	6.51 (3.0)	—*[3]	—*[3]	—*[3]	7.32	7.16	7.45-7.95
3d	1690	3210, 3420	9.53	8.07	5.89 (2.8)	—*[3]	7.75 (8.0)	7.71 (8.0)	7.34	7.21	7.43-7.61
3e	1690	3220, 3415*[2]	9.53	7.88	6.29 (2.9)	7.58-7.69	—*[3]	—*[3]	7.36	7.18	7.26-.59
3f	1700	3220, 3408*[2]	9.46	8.03	5.92 (3.2)	7.54-7.62	7.75 (7.9)	7.69 (7.9)	7.35	7.20	7.24-7.51
3g	1690	3210, 3450	9.37	7.90	5.85 (3.4)	7.46-7.61	7.74 (8.2)	7.69 (8.2)	7.33	7.20	6.91-7.46*[4]
3h	1690	3220, 3390*[2]	9.33	7.84	5.78 (3.0)	7.46-7.61	7.74 (7.8)	7.69 (7.8)	7.32	7.20	6.69-7.34*[4]
3i	1730	3220, 3395	9.57	8.14	5.93 (3.0)	7.47-7.63	7.77 (8.2)	7.71 (8.2)	7.34	7.22	7.50-8.60
3j	1710	3220, 3390*[2]	9.60	8.15	6.16 (3.4)	7.45-7.63	7.80 (7.9)	7.72 (7.9)	7.36	7.22	6.98-7.40
4	1540, 1575	3220, 3395	10.86	9.98 (3.1)	6.09 (3.1)	7.53-7.66	7.80 (7.8)	7.75 (7.8)	7.37	7.25	7.78-8.30

* **3a-j** X = O, **4** X = S.

*[2] The high-frequency band is sharp and strong.

*[3] The signal is overlapped by the multiplet of the Ar substituent protons.

*[4] Additional signals: 3.72 (3H, s, OCH_3) for the compound **3g** and 2.85 ppm (6H, s, $N(CH_3)_2$) for **3h**.

TABLE 3. Bond Lengths (*l*) and Bond Angles (ω) in Compound **3b**

Bond	*l*, Å	Angle	ω, deg.
$C_{(5)}$–$C_{(7)}$	1.527(3)	$N_{(2)}$–$C_{(7)}$–$C_{(10)}$	110.0(2)
$C_{(7)}$–$N_{(2)}$	1.462(3)	$N_{(2)}$–$C_{(7)}$–$C_{(5)}$	110.3(2)
$C_{(7)}$–$C_{(10)}$	1.520(2)	$C_{(10)}$–$C_{(7)}$–$C_{(5)}$	112.4(2)
$C_{(8)}$–$O_{(3)}$	1.247(3)	$O_{(3)}$–$C_{(8)}$–$N_{(2)}$	122.1(2)
$C_{(8)}$–$N_{(2)}$	1.329(3)	$O_{(3)}$–$C_{(8)}$–$N_{(3)}$	120.9(2)
$C_{(8)}$–$N_{(3)}$	1.369(3)	$N_{(2)}$–$C_{(8)}$–$N_{(3)}$	116.9(2)
$C_{(9)}$–$C_{(10)}$	1.342(3)	$C_{(10)}$–$C_{(9)}$–$N_{(3)}$	120.5(2)
$C_{(9)}$–$N_{(3)}$	1.396(3)	$C_{(10)}$–$C_{(9)}$–$C_{(18)}$	125.1(2)
$C_{(9)}$–$C_{(18)}$	1.493(2)	$N_{(3)}$–$C_{(9)}$–$C_{(18)}$	114.4(2)
$C_{(10)}$–$C_{(11)}$	1.465(3)	$C_{(9)}$–$C_{(10)}$–$C_{(11)}$	126.5(2)
$C_{(11)}$–$N_{(4)}$	1.300(3)	$C_{(9)}$–$C_{(10)}$–$C_{(7)}$	120.7(2)
$C_{(11)}$–$S_{(1)}$	1.764(2)	$C_{(11)}$–$C_{(10)}$–$C_{(7)}$	112.9(2)
$N_{(4)}$–$C_{(12)}$	1.380(3)	$C_{(8)}$–$N_{(2)}$–$C_{(7)}$	126.8(2)
$S_{(1)}$–$C_{(17)}$	1.735(2)	$C_{(8)}$–$N_{(3)}$–$C_{(9)}$	122.6(2)

The phenyl ring in this orientation apparently has a shielding effect on the benzothiazole ring protons, leading to an upfield shift of signals from the corresponding protons.

We should also note the appearance of the protons at $C_{(4)}$ and $C_{(7)}$ of the benzothiazole fragment in the ^{1}H NMR spectra for compounds **3c** and **3e**. The signals for these protons do not appear in the ranges noted for the other compounds. It is difficult to determine their precise location but the signals for compounds **3c** and **3e** certainly appear at 7.45-7.69 ppm and are overlapped by the multiplet for the aromatic protons of $C_{(6)}$–Ph and Ar as indicated by the corresponding integral intensities. The reason for this sharp upfield shift is probably related to the structure of the Ar fragments in compounds **3c** and **3e**. These fragments have substituents in the *ortho* position relative to the site of attachment to the pyrimidine ring. The resultant steric hindrance provides for conformationally rigid attachment of the Ar fragment to the heterocycle. The spatial orientation of the aromatic ring of the Ar fragment is strengthened such that the zone of its shielding effect strongly and relatively constantly acts at $C_{(4)}$ and $C_{(7)}$ of the benzothiazole ring. In contrast, the protons at $C_{(4)}$ of the pyrimidine ring are in the zone of a strong deshielding effect of the Ar substituent and their signals are shifted downfield by 0.20-0.42 ppm relative to the range typical for the other 4-phenyl derivatives. Thus, the conclusion that the cyclocondensation studied proceeds through the usual pathway of the Biginelli reaction may be extended to all the present examples.

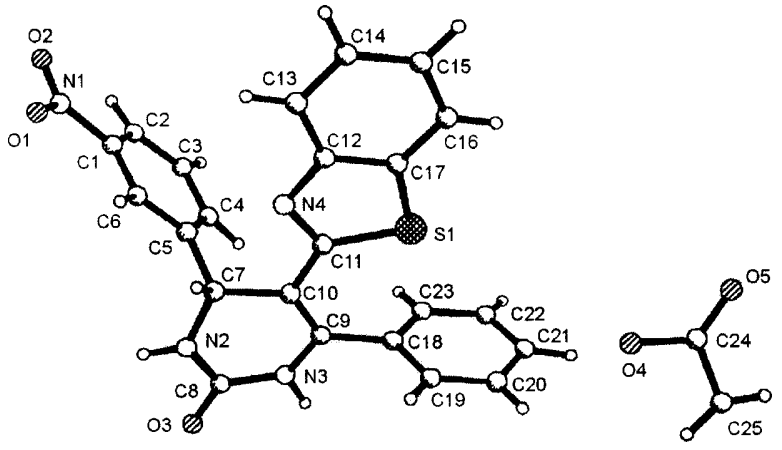


Fig. 1. General view of compound **3b** molecule with solvate acetic acid molecule.

TABLE. 4. Atomic Coordinates (Å×10^4) and Equivalent Isotropic Temperature Parameters (Å^2×10^3) in the Structure of Compound **3b**

Atom	*x*	*y*	*z*	U_{eq}
$S_{(1)}$	2922(1)	426(1)	3895(1)	41(1)
$C_{(1)}$	1927(3)	-3261(2)	7354(3)	54(1)
$C_{(2)}$	3468(4)	-2989(2)	7879(3)	62(1)
$C_{(3)}$	4290(3)	-1938(2)	8137(3)	60(1)
$C_{(4)}$	3547(3)	-1177(2)	7894(3)	46(1)
$C_{(5)}$	1976(2)	-1466(2)	7385(2)	34(1)
$C_{(6)}$	1154(3)	-2526(2)	7104(2)	43(1)
$C_{(7)}$	1135(2)	-632(1)	7180(2)	33(1)
$C_{(8)}$	1672(2)	875(2)	9013(2)	37(1)
$C_{(9)}$	2668(2)	1187(2)	6987(2)	34(1)
$C_{(10)}$	1986(2)	183(2)	6412(2)	33(1)
$C_{(11)}$	1959(2)	-258(2)	5049(2)	33(1)
$C_{(12)}$	1350(2)	-1530(2)	3334(2)	36(1)
$C_{(13)}$	700(3)	-2548(2)	2612(3)	47(1)
$C_{(14)}$	957(3)	-2723(2)	1337(3)	54(1)
$C_{(15)}$	1824(3)	-1913(2)	768(3)	54(1)
$C_{(16)}$	2469(3)	-906(2)	1461(2)	48(1)
$C_{(17)}$	2235(2)	-722(2)	2753(2)	37(1)
$C_{(18)}$	3522(2)	2050(1)	6326(2)	34(1)
$C_{(19)}$	2703(3)	2602(2)	5619(3)	45(1)
$C_{(20)}$	3489(3)	3380(2)	4979(3)	51(1)
$C_{(21)}$	5093(3)	3613(2)	5025(3)	52(1)
$C_{(22)}$	5903(3)	3073(2)	5728(3)	60(1)
$C_{(23)}$	5128(3)	2297(2)	6388(3)	50(1)
$C_{(24)}$	2998(3)	3871(2)	692(3)	66(1)
$C_{(25)}$	3266(8)	4998(3)	1286(7)	142(3)
$N_{(1)}$	1046(5)	-4392(2)	7030(4)	91(1)
$N_{(2)}$	924(2)	-107(1)	8436(2)	39(1)
$N_{(3)}$	2620(2)	1500(1)	8306(2)	41(1)
$N_{(4)}$	1208(2)	-1245(1)	4622(2)	38(1)
$O_{(1)}$	-320(4)	-4642(2)	6543(4)	108(1)
$O_{(2)}$	1742(6)	-5024(2)	7239(6)	191(2)
$O_{(3)}$	1526(2)	1222(1)	10145(2)	48(1)
$O_{(4)}$	2064(3)	3196(2)	1263(2)	82(1)
$O_{(5)}$	3552(3)	3597(2)	-221(3)	95(1)
$H_{(1)}$	3894(45)	-3520(31)	8143(39)	102(11)
$H_{(2)}$	5392(43)	-1683(27)	8509(35)	92(10)
$H_{(3)}$	4046(34)	-414(24)	8113(29)	58(8)
$H_{(4)}$	86(38)	-2776(25)	6710(32)	65(9)
$H_{(5)}$	44(27)	-1030(18)	6682(23)	31(6)
$H_{(6)}$	382(39)	-450(26)	8808(33)	63(9)
$H_{(7)}$	3010(32)	2114(23)	8627(28)	47(7)
$H_{(8)}$	1523(38)	2407(25)	5563(31)	67(8)
$H_{(9)}$	2821(39)	3730(27)	4518(34)	77(10)
$H_{(10)}$	5663(37)	4111(25)	4530(32)	62(9)
$H_{(11)}$	7052(39)	3269(25)	5800(32)	70(9)
$H_{(12)}$	5772(36)	1997(24)	6897(31)	61(8)
$H_{(13)}$	173(34)	-3093(24)	3010(30)	59(8)
$H_{(14)}$	552(38)	-3475(26)	840(33)	70(9)
$H_{(15)}$	1890(39)	-2028(27)	-92(40)	75(10)
$H_{(16)}$	3082(34)	-377(24)	1081(30)	65(8)
$H_{(17)}$	2892(54)	5070(36)	1888(49)	153(15)
$H_{(18)}$	3828(51)	5371(35)	1096(42)	119(13)

Therefore, 2-phenacylbenzothiazole is a convenient component in the Biginelli reaction for obtaining 2-oxo-6-phenyl-1,2,3,4-tetrahydropyrimidines and their 2-thioxo analogs containing 2-benzothiazole and various aromatic and heterocyclic substituents at $C_{(5)}$ and $C_{(4)}$, respectively.

EXPERIMENTAL

The course of the reactions and purity of the products were monitored by thin-layer chromatography on Silufol UV-254 plates with 9:1 benzene–ethanol as the eluent. The compounds were dried for 5 h in vacuum created by a water pump at 150°C prior to determination of the melting point, elemental analysis, and spectral investigation. The IR spectra were taken on a UR-40 spectrometer in KBr pellets. The ^{1}H NMR spectra were taken on a Varian VXR-300 spectrometer at 300 MHz in DMSO-d_6 with TMS as the internal standard. A sample of 2-phenacylbenzothiazole (**1**) was prepared according to Ciurdaru [16].

A monocrystal of compound **3b** was grown by slow crystallization from acetic acid and was obtained as crystal solvate **3b**·H_3CCO_2H with 1:1 mole ratio of the components. The X-ray diffraction analysis of the 0.5×0.35×0.15-mm monocrystal was carried out on a KM-4 diffractometer at the Institute of Chemical Physics Problems of the Russian Academy of Sciences in Chernogolovka, Russia.

The unit cell parameters were determined and refined using 25 reflections at $9° < \theta < 80°$. The number of independent reflections with intensity greater than twice the error margin ($I > 2\sigma(I)$), $N = 3686$. The reflections were measured with $\omega/2\theta$ scanning with Cu$K\alpha$ radiation and a graphite monochromator.

The structure was solved by the direct method. The positions of the hydrogen atoms were found from the difference maps. The positions of all the atoms with the exception of hydrogen were refined anisotropically and the positions of the hydrogen atoms were refined isotropically. The final $R_f = 0.0578$. The Shell 97 program package was used in the solution and refinement.

4-Aryl-5-(2-benzothiazolyl)-2-oxo-6-phenyl-1,2,3,4-tetrahydropyrimidines (3a-j) and 5-(2-Benzothiazolyl)-4-(*p*-nitrophenyl)-6-phenyl-2-thioxo-1,2,3,4-tetrahydropyrimidine (4). Mixture of compound **1** (1 mmol), aldehyde **2a-j** (1.1 mmol), and urea (3 mmol) in glacial acetic acid (1 ml) was stirred at 40-50°C until homogeneous and then maintained for a prolonged period without external interference. The analogous reaction with thiourea (3 mmol) was carried out in a mixture of glacial acetic acid (1 ml) and DMF (1 ml). The reaction times are given in Table 1. Water (1 ml) was added to the resultant reaction mixture and heated to reflux with stirring. After cooling to 20-25°C, the precipitate formed was filtered off and washed with water, 2-propanol, and ether.

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