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Alkylammonium and Alkylimidazolium Perhaloborates, Phosphates, and Aluminates as Catalysts in the Biginelli Reaction

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Abstract—Tri- and tetraalkylammonium, 1,3-dialkylimidazolium, and 1,2,3-trialkylimidazolium salts with BF_4^-, PF_6^-, $AlCl_4^-$, and $Al_2Cl_7^-$ as counterions catalyze the Biginelli reaction, ensuring preparation of various 3,4-dihydropyrimidin-2(1*H*)-one and 3,4-dihydropyrimidine-2(1*H*)-thione derivatives in high yields in the absence of a solvent.

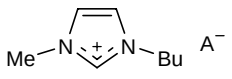
In the recent years, some 3,4-dihydropyrimidin-2(1*H*)-one derivatives were found to exhibit biological activity, in particular hypotensive [1] and antitumor [2], as well as to act as selective α_{1a} adrenoreceptor antagonists [3]. Some alkaloids isolated from marine algae contain a 3,4-dihydropyrimidin-2(1*H*)-one fragment [4]. One of the most efficient methods for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones is one-step three-component condensation of β-dicarbonyl compounds with aldehydes and urea or thiourea derivatives, which was discovered by P. Biginelli as early as 1893 [5]. This reaction is usually performed in an organic solvent (alcohol or THF) in the presence of a catalytic amount of a mineral [6] or Lewis acid [7].

Peng and Deng [8] recently described a procedure for the preparation of 3,4-dihydropyrimidin-2(1*H*)-ones from the same acyclic precursors without a solvent using ionic liquids as catalysts, specifically 1-butyl-3-methylimidazolium tetrafluoroborate (**I**) and hexafluorophosphate(V) (**II**). 1-Butyl-3-methylimidazolium chloride [BMIm][Cl] turned out to be less active, while tetrabutylammonium chloride $[Bu_4N][Cl]$ showed no catalytic activity [8].

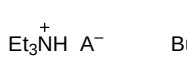
In order to elucidate the nature of the catalytic activity of quaternary ammonium salts and the scope of their application as catalysts in the Biginelli reaction under solvent-free conditions, we examined the catalytic activity of salts **III**–**X** consisting of alkylammonium or alkylimidazolium cation and the following anions: BF_4^-, PF_6^-, $AlCl_4^-$, and $Al_2Cl_7^-$. Salts **III**–**X** having bulky organic cations were selected due to their low melting points and good solubility in molten initial compounds. For the sake of comparison, we also reproduced the experimental conditions described in [8], i.e., catalysis of the Biginelli reaction by liquid salts **I** and **II**.

Triethylammonium salts **III** and **IV** were prepared by mixing equimolar amounts of triethylamine and tetrafluoroboric and hexafluorophosphoric acid, respectively, in methylene chloride. Tetrabutylammonium hexafluorophosphate(V) (**V**) was synthesized by anion exchange reaction between tetrabutylammonium bromide and hexafluorophosphoric acid in water; poorly soluble salt **V** separated from the solution. Benzyltriethylammonium chloroaluminates **VII** and **VIII** were obtained by fusion of benzyltriethylammonium chloride with 1 or 2 equiv of anhydrous aluminum chloride, respectively.

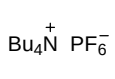
It should be noted that, although salts **III**–**V**, **VII**, and **VIII** have been widely used previously as catalysts and electrolytes (see, e.g., [9–13]), we have found no published data on their preparation and physical and spectral properties.



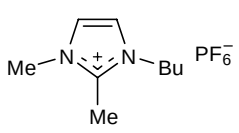
I, **II**, **IX**, **X**



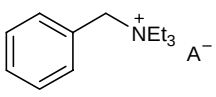
III, **IV**



V



VI



VII, **VIII**

I, **III**, A = BF_4; **II**, **IV**, A = PF_6; **VII**, **IX**, A = $AlCl_4$; **VIII**, **X**, A = Al_2Cl_7.

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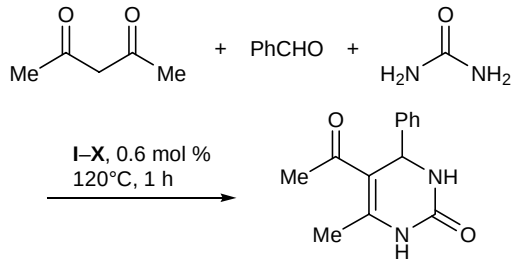
As model reaction we studied the condensation of acetylacetone with benzaldehyde and urea. The reactions were carried out by fusion equimolar amounts of the initial reactants and 0.6 mol % of salt **I**–**X** (or without catalyst) at 120°C over a period of 1 h. The yields of 5-acetyl-4-methyl-6-phenyl-3,4-dihydropyrimidin-2(1*H*)-one (**XIa**) (after recrystallization from 80% isopropyl alcohol) in the reactions of acetylacetone with benzaldehyde and urea in the presence of salts **I**–**X** are given below.



XIa

Catalyst	–	**I**	**II**	**III**	**IV**	**V**	**VI**	**VII**	**VIII**	**IX**	**X**
Yield, %	0	70[a]	85	80	88	50	86	67	66	75	78

[a] Yield 99% [8].

These data indicate that in all cases addition of salts **I**–**X** to the reaction mixture ensures formation of condensation product **XIa**. The catalytic activity of triethylammonium tetrafluoroborate (**III**) and hexafluorophosphate(V) (**IV**) somewhat exceeds the activity of 1,3-dialkylimidazolium salts **I** and **II** with the corresponding perfluorinated anions. A high yield of compound **XIa** (86%) was also obtained under catalysis by 1,2,3-trialkylimidazolium hexafluorophosphate(V) (**VI**). Benzyltriethylammonium chloroaluminates **VII** and **VIII** and 1-butyl-3-methylimidazolium chloroaluminates **IX** and **X** also turned out to be good catalysts in the Biginelli reaction. Using these salts, we succeeded in obtaining compound **XIa** containing no colored impurities typical of condensation products formed in the presence of fluorine-containing salts. The yield of **XIa** was slightly greater in the presence of imidazolium chloroaluminates **IX** and **X**, and it almost did not depend on the anion ($AlCl_4^-$ or $Al_2Cl_7^-$). Tetrabutylammonium hexafluorophosphate(V) (**V**) was the least active catalyst in the reaction under study: the yield of **XIa** was as low as 50%.

The yield of compound **XIa** obtained in the presence of salt **I** under the conditions reported in [8] was 70% against 99% given in [8]. Presumably, the reason for the observed difference is that our yield corresponds to an analytically pure sample (after recrystallization from isopropyl alcohol), while the yield given in [8], to the crude product.

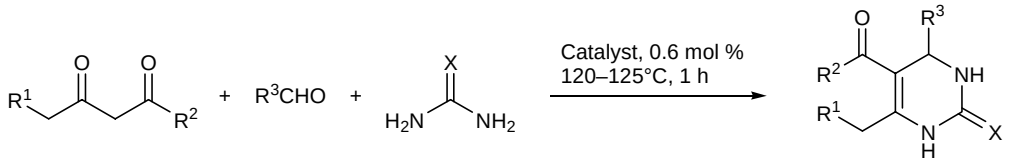
Ionic catalytic systems based on tri- and tetraalkylammonium cations and/or chloroaluminate anions are more advantageous than salts **I** and **II** due to their simple preparation and low costs of the initial compounds.

Salts **III**, **IV**, **IX**, and **X** were also used as catalysts in the syntheses of 3,4-dihydropyrimidin-2(1*H*)-ones **XIb**–**XIn** from various β-dicarbonyl compounds, aldehydes, and urea or thiourea in the absence of a solvent (see table). Both linear and cyclic β-dicarbonyl compounds and aromatic aldehydes with various substituents in the aromatic ring were involved. Higher yields of the condensation products from aromatic aldehydes were obtained in the presence of salts **III** and **IV** with perfluorinated anions, while the condensation with readily oxidizable aldehydes of the thiophene series was more effective in the presence of chloroaluminate salts **IX** and **X**. As a rule, the yields of compounds **XI** were somewhat lower than those obtained in an organic solvent [see, e.g., the data for **XIb**, **XIc**, **XIi**, and **XIj**). On the other hand, this loss in the yield is compensated by the simple experimental procedure and easy isolation of the products.

Salts **III**, **IV**, **IX**, and **X** were ineffective in condensations involving aliphatic aldehydes or *N*-alkylureas. These reactions were not selective, and they resulted in formation of complex mixtures of products. Presumably, the catalytic activity of salts **I**–**X** originates from their ability to release the corresponding hydrogen halides. It is known that chloroaluminates **VII**–**X** are hydrolytically unstable: in the presence of atmospheric moisture they decompose with elimination of hydrogen chloride [12–14]. Alkylimidazolium tetrafluoroborates and hexafluorophosphates(V) are more resistant to hydrolysis, but they gradually release HF on heating in the presence of traces of water [14]. Probably, tetrabutylammonium hexafluorophosphate(V) (**V**) is also capable of undergoing hydrolysis at elevated temperature. Triethylammonium salts **III** and **IV** are comparable to chloroaluminates **VII**–**X** in hydrolytic stability: on exposure to air they release HF even at room temperature (a peace of litmus paper turns red when placed over the surface of solid salt).

Trace amounts of hydrogen halides present in the reaction mixture are likely to initiate the Biginelli reaction [6]. Water liberated during the process accelerates hydrolysis of the catalyst, and the reaction becomes

Condensation of β-dicarbonyl compounds with aldehydes and urea (thiourea) in the presence of salts **III**, **IV**, **IX**, and **X**



XIa–XIn

Comp. no.	R^1	R^2	R^3	X	Catalyst	Yield,[a] %	mp., °C
XIa	H	Me	Ph	O	**IV**	88	235–236 (233–236 [7])
XIb	H	Me	Ph	S	**X**	80 (85[15][b])	220–224 (220–221 [15])
XIc	H	Me	3-$O_2NC_6H_4$	O	**IV**	86 (92 [16][b])	267–269 (268–270 [16])
XId	H	Me	4-$O_2NC_6H_4$	S	**IV**	74	207–209
					IX	70	
XIe	H	Me	4-ClC_6H_4	S	**IV**	86	189–191
					X	74	
XIf	H	Me	4-FC_6H_4	S	**IV**	89	209–212
					IX	72	
XIg	H	Me	2-Thienyl	O	**X**	80	230–232
XIh	H	OMe	Ph	O	**IX**	78 (92 [17][b])	212–214 (209–212 [18])
XIi	H	OMe	Ph	S	**IV**	83 (91 [15][b])	202–204 (201–202 [15])
XIj	H	OMe	2-Thienyl	O	**IX**	80	239–242
XIk	H	OMe	2-Thienyl	S	**IV**	72	226–227
					IX	84	
XIl	–CMe_2CH_2–		Ph	S	**III**	70 (56 [18][b])	282–285 (281–282 [19][b])
XIm	–CMe_2CH_2–		4-$O_2NC_6H_4$	S	**III**	68	279–282
XIn	–CMe_2CH_2–		2-Thienyl	O	**IV**	65	285–287

[a] After recrystallization from 80% aqueous isopropyl alcohol.
[b] The reaction was carried out in ethanol.

autocatalytic. Gradual increase in the concentration of protons improves the selectivity of the condensation and the yield. When the reaction was performed without a solvent in the presence of a catalytic amount of concentrated hydrochloric acid, i.e., in a relatively strongly acidic medium at the initial stage, the process was accompanied by tarring.

We can conclude that alkylammonium and alkylimidazolium salts with BF_4^-, PF_6^-, $AlCl_4^-$, and $Al_2Cl_7^-$ anions may be regarded as general catalysts in the Biginelli reaction, which are characterized by a common mechanism of the catalytic action. The use of these catalysts ensures preparation of a wide series of 3,4-dihydropirimidin-2(1*H*)-ones and -thiones in high yields under simple experimental conditions requiring no organic solvent.

EXPERIMENTAL

The ^1H NMR spectra were recorded on a Bruker WM-250 instrument (250.3 MHz). The elemental composition were determined on a Perkin–Elmer 2400 analyzer. Alkylimidazolium salts **I** [20], **II** [21], and **VI** [22] were synthesized by known methods.

Triethylammonium tetrafluoroborate (III). To a solution of 13.9 ml (0.10 mol) of triethylamine in 50 ml of methylene chloride we added dropwise at 0°C under vigorous stirring 18 ml (0.10 mol) of a 50% aqueous solution of HBF_4. The mixture was stirred for 15–20 min at 20°C, a saturated aqueous solution of sodium chloride was added, and the organic phase was separated, dried over $MgSO_4$, and evaporated under reduced pressure. Yield 7.56 g (40%), mp 58–62°C.

^{1}H NMR spectrum (acetone-d_6), δ, ppm: 8.80 br.s (1H), 3.35 m (6H), 1.35 t (9H, J = 7.5 Hz). Found, %: C 38.1; H 8.5; F 40.2; N 7.4. $C_6H_{16}BF_4N$. Calculated, %: C 38.12; H 8.55; F 40.2; N 7.41.

Triethylammonium hexafluorophosphate(V) (IV) was synthesized in a similar way using 24 ml (0.10 mol) of a 60% aqueous solution of HPF_6. Yield 16.2 g (70%), mp 72–74°C. ^{1}H NMR spectrum (acetone-d_6), δ, ppm: 8.00 br.s (1H), 3.40 m (6H), 1.40 t (9H, J = 7.5 Hz). Found, %: C 29.2; H 6.5; F 46.1; N 5.7. $C_6H_{16}F_6NP$. Calculated, %: C 29.16; H 6.52; F 46.12; N 5.67.

Tetrabutylammonium hexafluorophosphate(V) (V). To a solution of 6.44 g (0.02 mol) of tetrabutylammonium bromide in 30 ml of water we added dropwise at 0°C under vigorous stirring 4.8 ml (0.02 mol) of a 60% aqueous solution of HPF_6. The mixture was stirred for 15–20 min at 20°C, 25 ml of chloroform was added, and the organic phase was separated, washed with water (2×10 ml), dried over $MgSO_4$, and evaporated under reduced pressure. Yield 7.0 g (90%), mp 243–246°C. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.15 m (8H), 1.60 m (8H), 1.35 m (8H), 1.00 t (12H). Found, %: C 49.6; H 9.4; F 29.4; N 3.6. $C_{16}H_{36}F_6NP$. Calculated, %: C 49.6; H 9.37; F 29.42; N 3.62.

Benzyltriethylammonium tetrachloroaluminate (VII). A flask was dried, purged with argon, and charged with 1.14 g (5.0 mmol) of benzyltriethylammonium chloride, and 0.67 g (5.0 mmol) of $AlCl_3$ was slowly added at 20°C under stirring in a weak stream of argon. The mixture was then stirred for 2–3 h at 80–90°C (until it became homogeneous) and cooled to room temperature. Yield 1.60 g (90%), yellowish hygroscopic crystals, mp 72.5–75°C. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 7.50 m (5H), 4.55 s (2H), 3.25 d (6H, J = 7.6 Hz), 1.40 m (9H). Found, %: C 43.2; H 6.1; Cl 39.3; N 3.9. $C_{13}H_{22}AlCl_4N$. Calculated, %: C 43.23; H 6.15; Cl 39.26; N 3.88.

Benzyltriethylammonium heptachlorodialuminate (VIII) was synthesized in a similar way using excess $AlCl_3$ (1.30 g, 10.0 mmol). Yield 2.30 g (98%), yellowish hygroscopic crystals, mp 210–214°C. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 7.50 m (5H), 4.55 s (2H), 3.25 (6H, J = 7.7 Hz,), 1.40 m (9H). Found, %: C 31.6; H 4.5; Cl 50.2; N 2.8. $C_{13}H_{22}Al_2Cl_7N$. Calculated, %: C 31.57; H 4.49; Cl 50.18; N 2.83.

Condensation of β-dicarbonyl compounds with aldehydes and urea or thiourea (*general procedure*). A mixture of 3.0 mmol of β-dicarbonyl compound, 3.0 mmol of aldehyde, 3.0 mmol of urea or thiourea, and 0.018 mmol of salt **I**–**X** was thoroughly stirred and was heated for 1 h at 120–125°C. It was then cooled to room temperature, and the resulting crystalline product was washed in succession with hot water (2×4 ml), methylene chloride (2×4 ml), and diethyl ether (3 ml) and recrystallized from 2-propanol–water (4:1). The yields and melting points of compounds **XIa**–**XIn** are given in table.

5-Acetyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidine-2(1*H*)-thione (XId). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 10.35 s (NH, 1H), 9.75 s (NH, 1H), 8.20 d (2H, J = 7.5 Hz), 7.55 d (2H, J = 7.5 Hz), 5.40 d (1H, J = 7.4 Hz), 2.35 s (3H), 2.25 s (3H). Found, %: C 53.6; H 4.5; N 14.4; S 11.1. $C_{13}H_{13}N_3O_3S$. Calculated, %: C 53.64; H 4.47; N 14.42; S 11.01.

5-Acetyl-4-(4-chlorophenyl)-6-methyl-3,4-dihydropyrimidine-2(1*H*)-thione (XIe). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 10.25 s (NH, 1H), 9.65 s (NH, 1H), 7.45 d (2H, J = 7.5 Hz), 7.20 d (2H, J = 7.5 Hz), 5.25 d (1H, J = 7.6 Hz), 2.35 s (3H), 2.25 s (3H). Found, %: C 55.6; H 4.7; N 10.0; S 12.6. $C_{13}H_{13}ClN_2OS$. Calculated, %: C 55.63; H 4.71; N 10.02; S 12.58.

5-Acetyl-4-(4-fluorophenyl)-6-methyl-3,4-dihydropyrimidine-2(1*H*)-thione (XIf). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 10.20 s (NH, 1H), 9.65 s (NH, 1H), 7.20–7.45 m (4H), 5.31 d (1H, J = 6.5 Hz), 2.35 s (3H), 2.25 s (3H). Found, %: C 59.1; H 5.0; N 10.6; S 12.1. $C_{13}H_{13}FN_2OS$. Calculated, %: C 59.09; H 5.01; N 10.62; S 12.11.

5-Acetyl-6-methyl-4-(2-thienyl)-3,4-dihydropyrimidin-2(1*H*)-one (XIg). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 9.03 s (NH, 1H), 7.75 s (NH, 1H), 7.20–6.85 m (3H), 5.42 d (1H, J = 6.4 Hz), 2.35 s (3H), 2.25 s (3H). Found, %: C 55.9; H 5.1; N 11.9; S 13.6. $C_{11}H_{12}N_2O_2S$. Calculated, %: C 55.91; H 5.09; N 11.88; S 13.64.

Methyl 6-methyl-2-oxo-4-(2-thienyl)-1,2,3,4-tetrahydropyrimidine-5-carboxylate (XIj). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 9.03 s (NH, 1H), 7.75 s (NH, 1H), 7.20–6.85 m (3H), 5.42 d (1H, J = 6.4 Hz), 3.60 s (3H), 2.25 s (3H). Found, %: C 52.4; H 4.8; N 11.1; S 12.7. $C_{11}H_{12}N_2O_3S$. Calculated, %: C 52.39; H 4.82; N 11.09; S 12.72.

Methyl 6-methyl-4-(2-thienyl)-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (XIk). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm:

10.25 s (NH, 1H), 9.55 s (NH, 1H), 7.20–6.85 m (3H), 5.42 d (1H, *J* = 6.4 Hz), 3.60 s (3H), 2.25 s (3H). Found, %: C 49.2; H 4.5; N 10.4; S 23.9. $C_{11}H_{12}N_2O_2S_2$. Calculated, %: C 49.23; H 4.52; N 0.41; S 23.88.

7,7-Dimethyl-4-(4-nitrophenyl)-2-thioxo-2,3,4,6,7,8-hexahydro-1*H*-quinazolin-5-one (XIm). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 10.40 s (NH, 1H), 9.55 s (NH, 1H), 8.12 d (2H, *J* = 7.3 Hz), 7.65 d (2H, *J* = 7.4 Hz), 5.35 d (1H, *J* = 7.4 Hz), 2.35 d (1H, *J* = 16 Hz), 2.40 d (1H, *J* = 16 Hz), 2.10 d (1H, *J* = 15 Hz), 2.20 d (1H, *J* = 15 Hz), 1.15 s (3H), 0.9 s (3H). Found, %: C 58.0; H 5.2; N 12.7; S 9.7. $C_{16}H_{17}N_3O_3S$. Calculated, %: C 58.04; H 5.23; N 12.68; S 9.69.

7,7-Dimethyl-4-(2-thienyl)-2-thioxo-2,3,4,6,7,8-hexahydro-1*H*-quinazolin-5-one (XIn). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 9.03 s (NH, 1H), 7.75 s (NH, 1H), 7.20 s (1H,), 6.85 s (2H), 5.35 d (1H, *J* = 7.4 Hz), 2.35 d (1H, *J* = 16 Hz), 2.40 d (1H, *J* = 16 Hz), 2.10 d (1H, *J* = 15 Hz), 2.20 d (1H, *J* = 15 Hz), 1.15 s (3H), 0.90 s (3H). Found, %: C 60.8; H 5.8; N 10.1; S 11.6. $C_{14}H_{18}N_2O_2S$. Calculated, %: C 60.78; H 5.77; N 10.09; S 11.64.

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