Reactions of alkyl 2-benzylidene-2-polyfluoroacylacetates with *N*,*N*-dinucleophiles

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Alkyl 2-benzylidene-2-polyfluoroacylacetates react with urea and thiourea to yield 5-ethoxycarbonyl-4-fluoroalkyl-4-hydroxy-6-phenylhexahydropyrimidin-2-ones and -2-thiones and with guanidine sulfate to form 2-amino-5-ethoxycarbonyl-4-fluoroalkyl-6-phenyl-1,6-dihydropyrimidines and 3,6-diethoxycarbonyl-2,7-difluoroalkyl-4,5-diphenyl-4,5-dihydro-1*H*-pyrido[1,2-*a*]pyrimidines, and they react with phenylhydrazine to afford 4-alkoxycarbonyl-3-fluoroalkyl-3-hydroxy-1,5-diphenylpyrazolidines. Hydrazine hydrate catalyzes the formation of 3,5-diethoxycarbonyl-2,6-difluoroalkyl-2,6-dihydroxy-4-phenyltetrahydropyrans. When treated with anhydrous hydrazine and *o*-phenylenediamine, these esters cleave to form the products of condensation of fluoroacyl ester and benzaldehyde with diamines.

Key words: alkyl 2-benzylidene-2-polyfluoroacylacetates, dinucleophiles, urea, thiourea, guanidine, hydrazine, phenylhydrazine, *o*-phenylenediamine.

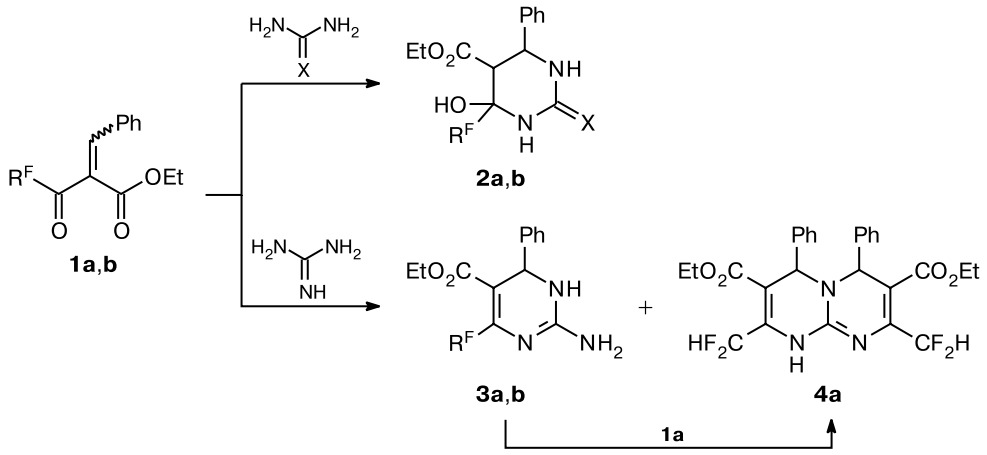
Recently synthesized fluoroalkyl-containing 2-benzylidene-3-oxoalkanoates[1,2] are promising reagents for syntheses of heterocycles, because they contain three nonequivalent reaction centers: keto group, ester fragment, and C=C bond. Therefore, 2-benzylidene-3-oxoalkanoates can possess reactivity of both 3-oxoalkanoates and vinyl ketones.

Nonfluorinated 2-arylidene-3-oxoalkanoates are known to be used for syntheses of heterocycles of the pyrimidine series.[3—6] Data on reactions of the fluorinated analogs with dinucleophiles are lacking.

In this work, we studied the reactions of alkyl 2-benzylidene-2-polyfluoroacylacetates **1** with various *N*,*N*-dinucleophiles, such as urea, thiourea, guanidine, hydrazine, phenylhydrazine, and *o*-phenylenediamine.

It was found that refluxing of equimolar amounts of esters **1a**,**b** with urea or thiourea in DMF in the presence of sodium acetate for 6—8 h afforded substituted 4-hydr-

Scheme 1



R^F = HCF_2(**1a**, **3a**, **4a**), X = O (**2a**)
R^F = CF_3 (**1b**, **3b**), X = S (**2b**)

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Table 1. Main physicochemical characteristics of compounds **3a**,**b**, **4a**, **6b**, and **8a—c**

Compound	M.p. /°C	Yield (%)	Found / Calculated (%)				Molecular formula
			C	H	F	N	
3a	203—205	45	56.85	5.09	12.90	14.32	$C_{14}H_{15}F_2N_3O_2$
			56.94	5.12	12.87	14.23	
3b	166—168	59	53.67	4.45	18.13	13.46	$C_{14}H_{14}F_3N_3O_2$
			53.68	4.50	18.19	13.41	
4a	172—175	36* (59)	60.99	4.88	14.21	8.01	$C_{27}H_{25}F_4N_3O_4$
			61.01	4.74	14.29	7.91	
6b	160	48	26.85	0.83	61.39	7.01	$C_9H_3F_{13}N_2O$
			26.89	0.75	61.41	6.97	
8a	97—99	66	59.92	5.14	14.68	7.10	$C_{19}H_{19}F_3N_2O_3$
			60.00	5.03	14.98	7.36	
8b	95—97	52	58.24	4.83	18.59	6.75	$C_{20}H_{20}F_4N_2O_3$
			58.26	4.85	18.43	6.78	
8c	155—157	49	48.81	3.29	33.19	5.40	$C_{21}H_{17}F_9N_2O_3$
			48.85	3.32	33.11	5.42	

* The yield of the product synthesized by method ***A***, and that by method ***B*** is given in parentheses.

oxyhexahydropyrimidin-2-ones **2a** and 4-hydroxyhexahydropyrimidin-2-thiones **2b** (Scheme 1). The same heterocycles have been synthesized previously[7,8] by the three-component Biginelli reaction from 3-oxoalkanoates, urea or thiourea, and benzaldehyde.

Note that the reactions of nonfluorinated 2-arylidene-3-oxoalkanoates with urea or thiourea afford tetrahydropyrimidin-2-on(thion)es.[4] In the case of the fluorinated analogs, the formation of hexahydropyrimidines with the *gem*-aminoalcohol fragment is explained by the presence of electron-withdrawing fluoroalkyl substituents, which prevent water molecule elimination.

Unlike urea and thiourea, guanidine sulfate reacts with 2-benzylidene-3-oxoalkanoates **1a**,**b** in boiling DMF in the presence of the base (Na_2CO_3) to yield 2-aminodihydropyrimidines **3a**,**b** (Table 1). The formation of dihydropyrimidines **3** instead of hexahydropyrimidines **2** in the case of guanidine is caused, most likely, by the higher basicity of the latter (pK_{a_1} = 13.6, pK_{a_2} = 11) compared to those of urea (pK_a = 0.31) and thiourea (pK_a = −0.96).[9]

The reaction products of 2-benzylidene-3-oxoalkanoate **1a** containing the difluoromethyl substituent yielded pyrimidine **3a** and pyrimidopyrimidine **4a** (see Scheme 1). The latter was also obtained from 2-aminodihydropyrimidine **3a** and ester **1a**. It is most likely that bicycle **4b** is also formed in the reaction of ester **1b** with the guanidine salt. However, this compound cannot be isolated because of the formation of a large amount of by-products.

It has previously[5] been shown that similar binuclear heterocycles are formed from 2-aminodihydropyrimidines and also by the reactions of 2-arylidene-substituted benzoylacetates with guanidine sulfate. The three-component Biginelli condensation of benzoylacetate, aldehyde, and guanidine affords 2-aminodihydropyrimidines as the main products (yields up to 70%).[6] We succeeded to isolate only an insignificant amount of pyrimidine **3b** from the reaction of ester **1b** with benzaldehyde and guanidine, because the mixture of products that formed was difficult to separate.

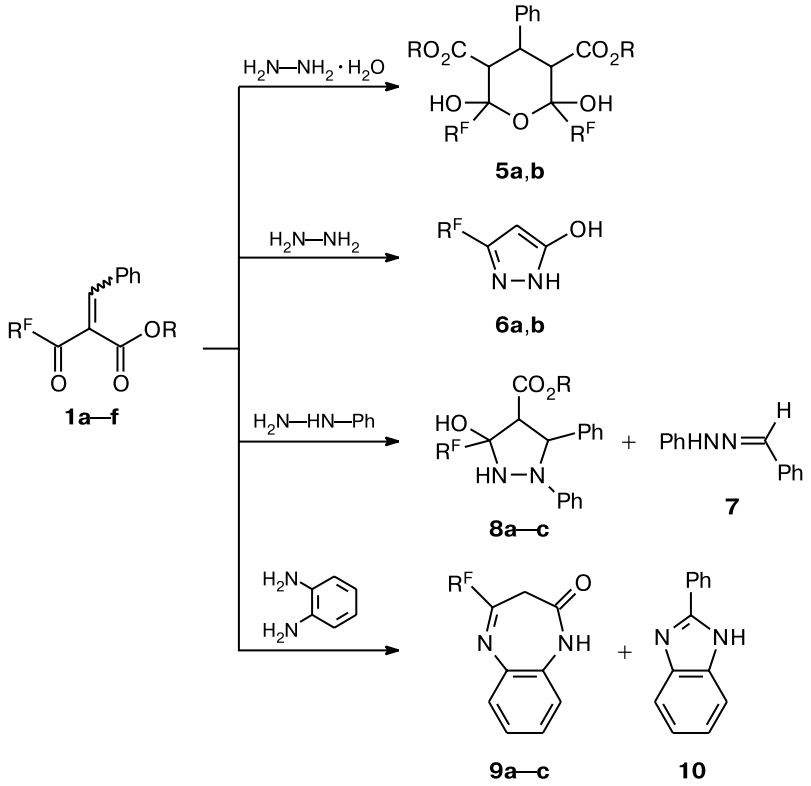
The reactions of 2-benzylidene-3-oxoalkanoates **1a**,**b** with hydrazine hydrate in diethyl ether or ethanol afford substituted tetrahydropyrans **5a**,**b** instead of expected pyrazoles (Scheme 2).

We have previously isolated heterocycles **5a**,**b** from the reactions of 2-polyfluoroacylacetates with aldehydes or 2-benzylidene-3-oxoalkanoates in ethanol in the presence of the base.[2] Probably, 2-benzylidene-3-oxoalkanoate **1** decomposes partially, under the reaction conditions, to benzaldehyde and 3-oxoalkanoate. The latter reacts with ester **1**, and hydrazine acts as a base.

Pyrazoles **6a**,**b** were isolated from the products of the reactions of 2-benzylidene-3-oxoalkanoates **1a,f** with anhydrous hydrazine in absolute ethanol (see Scheme 2). They are formed due to transformations of the products of cleavage of the starting ester **1**, 3-oxoalkanoate, and aldehyde. Attempts to obtain pyrazoles using hydrazine in the form of its hydrochloride were unsuccessful.

Phenylhydrazine reacts readily with 2-benzylidene-3-oxoalkanoates **1b**,**c**,**e** in anhydrous diethyl ether to form pyrazolidine derivatives **8a—c** (see Scheme 2) containing, as in the case of hexahydropyrimidines **2** and tetrahydropyrans **5**, the hydroxyl group in the position adjacent to the fluoroalkyl substituent. This reaction is accompanied by the partial decomposition of the starting 2-benzylidene-3-oxoalkanoate **1**, which is indicated

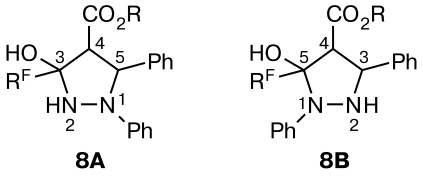
Scheme 2



R = Et, R^F = HCF_2 (**1a**, **5a**, **6a**, **9a**), CF_3 (**1b**, **5b**, **8a**), $H(CF_2)_2$ (**1c**, **4a**, **8b**, **9b**), $H(CF_2)_4$ (**1d**, **9c**), C_6F_{13} (**1f**, **6b**)
R = Me, R^F = C_4F_9 (**1e**, **4b**, **8c**)

by hydrazone **7** isolated from the reaction mixture (Scheme 2).

The formation of two regioisomeric pyrazolidines **8A** and **8B** is theoretically possible through the reactions of 2-benzylidene-3-oxoalkanoates **1** with phenylhydrazine.



8A **8B**

The structures of pyrazolidines **8** were established by ^{1}H 2D NOESY, 2D ^{1}H-^{13}C HSQC, and 2D ^{1}H-^{13}C HMBC NMR spectra using compound **8a** as an example. The 2D NOESY NMR spectrum of **8a** contains cross peaks between the H(5) proton and *ortho*-protons of the *N*-phenyl group and between the *ortho*-protons of two phenyl substituents, which can take place in structure **8A**. This structure is favored by the presence of cross peaks of the H(5) proton with the C_{ipso} atom of the *N*-phenyl substituent and the proton of the amino group with the carbon atom bound to the trifluoromethyl group in the 2D ^{1}H-^{13}C HMBC NMR spectrum.

Based on the data obtained, we can conclude that pyrazolidines **8** are formed due to the primary addition of the free amino group of phenylhydrazine to the fluoroacyl fragment of ester **1** followed by ring closure due to the addition of the second amino group to the C=C double bond.

The structures of pyrazolidines **8a—c** are characterized by three chiral centers. Therefore, four possible diastereomers (and, in addition, four their enantiomeric forms) can be proposed for these compounds. However, analysis of the ^{1}H and ^{19}F NMR spectra shows that only one diastereoisomer is present in each case. The high conformational energy of the phenyl group (ΔG_{Ph} = 12.1 kJ mol^{-1}) makes it possible to fix preferentially the conformation in which this group is equatorial.[10] In addition, the experimentally found value of the spin-spin coupling constant (9.4—10.0 Hz) corresponding to the axial-axial interaction of protons at the C(4) and C(5) carbon atoms indicates the equatorial arrangement of the phenyl and alkoxycarbonyl groups.[11] The conformational energies[10] for the CF_3 (ΔG_{CF3} 8.8 kJ mol^{-1}) and OH (ΔG_{OH} 2.2 kJ mol^{-1}) groups in substituted cyclohexanones suggest the preferential equatorial arrangement of the trifluoromethyl residue. Moreover, according to X-ray

diffraction data, this arrangement of the trifluoromethyl and hydroxyl substituents is observed for ethyl 2-oxo-4-phenyl-6-trifluoromethyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate[12] and 2,6-dihydroxy-3,5-dimethoxycarbonyl-4-phenyl-2,6-di(1,1,2,2-tetrafluoroethyl)tetrahydropyran.[2]

2-Benzylidene-3-oxoalkanoates **1** in the reactions with *o*-phenylenediamine also undergo *retro*-decomposition similarly to transformations with anhydrous hydrazines. Regardless of the reaction conditions (refluxing in anhydrous benzene, diethyl ether, and ethanol), esters **1a**,**c**,**e** yield benzodiazepines **9a**—**c** and benzimidazole **10**, which

Table 2. Main spectral characteristics of compounds **3a**,**b**, **4a**, **6b**, and **8a**—**c**

Compound	IR, ν/cm^{-1}	NMR (DMSO-d$_6$) δ, *J*/Hz	
		^{1}H	^{19}F
3a	3470, 3380, 3320, 1600; 1580 (NH, NH$_2$); 3020 (C—H stret.); 1660 (CO$_2$Et); 1600, 1580, 1500 (C=C, C=N); 1220—1140 (C—F)	1.14 (t, 3 H, OCH$_2$C<u>H$_3$</u>, *J* = 7.1); 4.03 (q, 2 H, O<u>CH$_2$</u>Me, *J* = 7.1); 5.34 (s, 1 H, CH); 6.37 (br.s, 2 H, NH$_2$); 7.31 (m, 5 H, Ph); 7.42 (t, 1 H, HCF$_2$, *J* = 55.3); 7.96 (br.s, 1 H, NH)	39.16 (d, HCF$_2$, *J* = 55.3)
3b	3420, 3350, 3320, 1590, 1550 (NH, NH$_2$); 3110 (C—H stret.); 1650 (CO$_2$Et); 1590, 1575, 1500 (C=C, C=N); 1230—1130 (C—F)	1.13 (t, 3 H, OCH$_2$C<u>H$_3$</u>, *J* = 7.1); 4.03 (q, 2 H, O<u>CH$_2$</u>Me, *J* = 7.1); 5.44 (s, 1 H, CH); 5.99 (br.s, 2 H, NH$_2$); 7.09 (br.s, 1 H, NH); 7.25—7.36 (m, 5 H, Ph)	98.92 (s, CF$_3$)
4a	3370, 1620, 1550 (NH); 3010 (C—H stret.); 1700 (CO$_2$Et); 1620, 1550, 1500 (C=C, C=N); 1220—1020 (C—F)	1.05 (t, 6 H, 2 OCH$_2$C<u>H$_3$</u>, *J* = 7.1); 3.98 (q, 4 H, 2 O<u>CH$_2$</u>Me, *J* = 7.1); 5.38 (s, 2 H, 2 CH); 7.32 (t, 2 H, 2 HCF$_2$, *J* = 54.3); 7.35 (m, 10 H, 2 Ph); 11.13 (br.s, 1 H, NH)	41.92 (d, HCF$_2$, *J* = 54.3)
6b	3229, 1536 (NH); 2596, 2200 (OH); 1140—1242 (C—F)	5.66 (s, 1 H, CH); 11.26 and 12.96 (both c, 1 H each, NH and OH)	35.75 (m, 2 F, CF$_2$); 39.05 (m, 2 F, CF$_2$); 40.66 (m, 4 F, (CF$_2$)$_2$); 45.78 (m, 2 F, CF$_2$); 81.03 (s, 3 F, CF$_3$)
8a	3340, 3267, 1597 (NH, OH); 3015 (C—H stret.); 1694 (CO$_2$Et); 1175—1237 (C—F)	1.13 (t, 3 H, OCH$_2$C<u>H$_3$</u>, *J* = 7.1); 3.24 (dd, 1 H, CH(4), *J* = 9.4, *J* = 1.7); 4.11, 4.03 (two dq, both 1 H, O<u>CH$_2$</u>Me, *J* = 10.8, *J* = 7.1); 5.11 (d, 1 H, CH(5), *J* = 9.4); 6.71 (tt, 1 H, (N)CH$_p$, *J* = 7.3, *J* = 1.1); 6.85 (dd, 2 H, (N)CH$_o$, *J* = 8.8, *J* = 1.1); 6.96 (s, 1 H, NH); 7.01 (d, 1 H, OH, *J* = 1.7); 7.11(dd, 2 H, (N)CH$_m$, *J* = 8.8, *J*= 7.3); 7.31 (tt, 1 H, CH$_p$, *J* = 7.3, *J* = 1.3); 7.39 (dd, 2 H, CH$_m$, *J*= 8.2, *J* = 7.3); 7.54 (dd, 2 H, CH$_o$, *J* = 8.2, *J* = 1.3)	81.60 (s, CF$_3$)*
8b	3360, 3275 (NH, OH); 3020 (C—H stret.); 1680 (CO$_2$Et); 1585, 1480 (C=C, C=N); 1200—1090 (C—F)	1.23 (t, 3 H, OCH$_2$C<u>H$_3$</u>, *J* = 7.1); 3.45 (d, 1 H, CH(4), *J* = 10.0); 4.24, 4.27 (both dq, 1 H each, O<u>CH$_2$</u>Me, *J* = 10.8, *J* = 7.1); 4.84 (d, 1 H, CH(5), *J* = 10.0); 4.88 (dt, 1 H, OH, *J* = *J* = 1.4); 5.68 (d, 1 H, NH, *J* = 1.7); 6.29 (dddd, 1 H, HCF$_2$, *J* = 53.8, *J* = 52.0, *J* = 11.6, *J* = 2.0); 6.84 (tt, 1 H, (N)H$_p$, *J* = 7.3, *J* = 1.1); 6.98 (dd, 2 H, (N)H$_o$, *J* = 8.8, *J* = 1.1); 7.16 (dd, 2 H, (N)H$_m$, *J* = 8.8, *J* = 7.3); 7.4 (m, 5 H, Ph(5))*	—
8c	3400, 3320 (NH, OH); 3020 (C—H stret.); 1710 (CO$_2$Me); 1600, 1500 (C=C, C=N); 1230—1120 (C—F)	3.50 (d, 1 H, CH(4), *J* = 9.8); 3.79 (s, 3 H, OMe); 4.87 (d, 1 H, CH(5), *J* = 9.8); 4.91 (dt, 1 H, OH, *J* = *J* = 1.4); 5.34 (d, 1 H, NH, *J* = 1.7); 6.85 (tt, 1 H, (N)H$_p$, *J* = 7.3, *J* = 1.2); 6.99 (dd, 2 H, (N)H$_O$, *J* = 8.8, *J* = 1.2); 7.17 (dd, 2 H, (N)H$_m$, *J* = 8.8, *J* = 7.3); 7.4 (m, 5 H, Ph(5))*	35.96 (AB system, 2 F, γ-CF$_2$, Δ$_{AB}$= 0.67, *J*$_{AB}$ = 294.0); 40.84 (AB system, 2 F, β-CF$_2$, Δ$_{AB}$ = 1.53, *J*$_{AB}$ = 301.0); 42.27 (t.m, 2 F, α-CF$_2$, *J* = 15.0); 81.03 (tt, 3 F, CF$_3$, *J* = 10.2, *J* = 2.4)*

* In CDCl$_3$.

are the products of the reaction of *o*-phenylenediamine with 3-oxoalkanoate and benzaldehyde, respectively (Scheme 2). The formation of benzodiazepine from 2-benzylidene-3-oxoalkanoate **1** is likely thermodynamically unfavorable.

Thus, 2-benzylidene-2-polyfluoroacylacetates in the reactions with dinucleophiles (urea, thiourea, guanidine, phenylhydrazine) behave as typical vinyl ketones adding dinucleophiles to the carbon—carbon double bond and keto group. The reaction of guanidine manifests an additional possibility of formation of the binuclear heterocycle, *viz.*, pyrimidopyrimidine. However, it is difficult to use 2-benzylidene-3-oxoalkanoates because of their easy cleavage to 3-oxoalkanoate and benzaldehyde. This process becomes the main reaction in the case of the reactions with *o*-phenylenediamine and hydrazine.

Experimental

IR spectra were recorded on a Specord 75IR spectrometer in an interval of 400—4000 cm^{-1} in Nujol. NMR spectra were obtained on Tesla BS-587A (^1H, 80 MHz, relatively to $SiMe_4$; ^{19}F, 75.3 MHz, relatively to C_6F_6) and Bruker DRX-400 (^1H, 400 MHz; ^{13}C, 100 MHz relatively to $SiMe_4$; ^{19}F, 376 MHz, relatively to C_6F_6) spectrometers. Elemental analysis was carried out on an Carlo Erba CHNS-O EA 1108 analyzer.

Physicochemical constants and elemental analysis data are presented in Tables 1 and 2. The data of the ^{13}C NMR spectrum for compound **8a** are given in Table 3.

The starting alkyl 2-benzylidene-2-polyfluoroacylacetates **1a—f** were synthesized according to previously described procedures.[1,2]

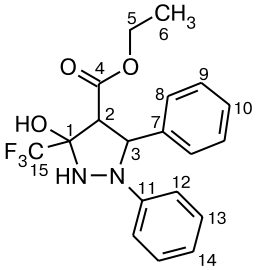


Table 3. ^{13}C NMR spectrum (δ, $J_{^{13}C-^{19}F}$/Hz) of 4-ethoxycarbonyl-3-hydroxy-1,5-diphenyl-3-trifluoromethylpyrazolidine (**8a**) in DMSO-d_6

Atom	δ ($J_{C,F}$/Hz)	Atom	δ ($J_{C,F}$/Hz)
C(1)	89.88 (q, J = 31.0)	C(9)	128.95
C(2)	62.50	C(10)	127.81
C(3)	69.07	C(11)	152.20
C(4)	166.15	C(12)	113.38
C(5)	60.86	C(13)	128.26
C(6)	13.79	C(14)	118.40
C(7)	141.96	C(15)	122.27 (q, J = 286.0)
C(8)	126.21		

5-Ethoxycarbonyl-4-fluoroalkyl-4-hydroxy-6-phenylhexahydropyrimidin-2-on(thion)es (2a,b) (general procedure). A mixture of 2-benzylidene-3-oxoalkanoate **1a,b** (0.01 mol), urea (thiourea) (0.01 mol), and sodium acetate (0.16 g, 0.003 mol) in DMF (10 mL) was heated for 6—8 h at 70 °C. Then the reaction mixture was poured into water, and the precipitate that formed was filtered off and recrystallized from ethanol. Compounds **2a** (1.91 g, 64%) and **2b** (1.81 g, 52%) were obtained, whose physicochemical characteristics correspond to published data.[8]

2-Amino-5-ethoxycarbonyl-4-fluoroalkyl-6-phenyl-1,6-dihydropyrimidines (3a,b). A mixture of 2-benzylidene-3-oxoalkanoate **1a**,**b** (0.01 mol), guanidine sulfate (1.89 g, 0.01 mol), and sodium bicarbonate (1.10 g, 0.013 mol) in DMF (10 mL) was heated for 8—10 h at 70 °C. Then the reaction mixture was poured into water, and the precipitate that formed was filtered off, washed with hot chloroform, and recrystallized from ethanol. Compounds **3a** (1.33 g) and **3b** (1.84 g) were obtained (see Tables 1 and 2).

3,6-Diethoxycarbonyl-2,7-di(difluoromethyl)-4,5-diphenyl-4,5-dihydro-1*H*-pyrimido[1,2-*a*]pyrimidine (4a). ***A.*** Compound **4a** was isolated from the filtrate after purification of product **3a**. The filtrate was concentrated, and the residue was washed with Et_2O. Compound **4a** was obtained (1.91 g) (see Tables 1 and 2).

B. A mixture of ester **1a** (2.72 g, 0.01 mol), pyrimidine **3a** (2.95 g, 0.01 mol), and sodium bicarbonate (1.10 g, 0.003 mol) in DMF (10 mL) was heated for 8—10 h at 70 °C. Then the reaction mixture was poured into water, and the precipitate that formed was filtered off and washed with Et_2O. Compound **4a** (3.13 g) was obtained (see Tables 1 and 2).

3,5-Diethoxycarbonyl-2,6-di(fluoroalkyl)-2,6-dihydroxy-4-phenyltetrahydropyrans (5a,b). A mixture of 2-benzylidene-3-oxoalkanoate **1a,b** (0.01 mol) and hydrazine hydrate (0.50 g, 0.01 mol) in EtOH (10 mL) was refluxed for 6—8 ч. Then the solvent was evaporated, and the residue was recrystallized from hexane. Compounds **5a** (1.97 g, 45%) and **5b** (1.80 g, 38%) were obtained, and their physicochemical characteristics correspond to published data.[1]

3-Fluoroalkyl-5-hydroxypyrazoles (6a,b). A mixture of 2-benzylidene-3-oxoalkanoate **1a,f** (0.01 mol) and hydrazine (0.32 g, 0.01 mol) was refluxed in anhydrous EtOH for 2 h. Then the solvent was evaporated, and the residue was recrystallized from hexane. Compound **6a** (0.48 g, 36%), whose physicochemical characteristics correspond to published data,[13] and compound **6b** (1.93 g, 48%) were obtained (see Tables 1 and 2).

4-Alkoxycarbonyl-3-fluoroalkyl-3-hydroxy-1,5-diphenylpyrazolidines (8a—c). A mixture of 2-benzylidene-3-oxoalkanoate **1b,c,e** (0.01 mol) and phenylhydrazine (1.08 g, 0.01 mol) in anhydrous Et_2O (10 mL) was kept at ~20 °C for a week. Then the solvent was evaporated, and the residue was washed with hot hexane. Compounds **8a** (4.01 g), **8b** (2.14 g), and **8c** (2.55 g) were obtained (see Tables 1—3). After the filtrate was concentrated, **benzaldehyde phenylhydrazone (7)** was obtained in 15% (0.29 g), 22% (0.43 g), and 29% (0.56 g) yields, respectively, and its physicochemical characteristics correspond to those presented in the literature.[14]

Reaction of 2-benzylidene-3-oxoalkanoates 1 with *o*-phenylenediamine. A mixture of 2-benzylidene-3-oxoalkanoate **1a**,**c**,**d** (0.01 g) and *o*-phenylenediamine (1.08 g, 0.01 mol) in anhydrous Et_2O (benzene or EtOH) (20 mL) was refluxed for 4—6 h. The solvent was evaporated, and the residue was recrystallized from ethanol. **4-Fluoroalkyl-1*H*-1,5-benzodiazepin-2-**

ones 9a—c were obtained in 59% (1.24 g) (**9a**), 50% (1.30 g) (**9b**) and 58% (2.08 g) (**9c**) yields, and their physicochemical characteristics correspond to published data.[15] The filtrate was concentrated, and the residue was recrystallized from hexane. **2-Phenylbenzimidazole (10)** was obtained in 19% (0.34 g), 22% (0.40 g), and 20% (0.36 g) yields, respectively, and its physicochemical characteristics correspond to published data.[16]

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