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Ferrocenyl-Substituted α,β-Unsaturated Ketones in Synthesis of Tetrahydropyrimidinones

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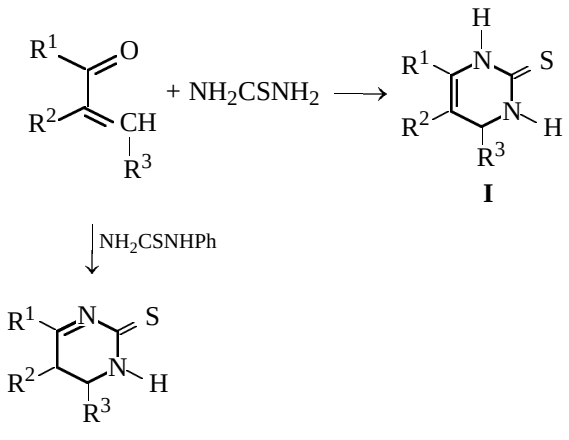
Abstract — Ferrocenyltetrahydropyrimidin-2-ones were prepared by reactions of linear and cyclic α,β-unsaturated ketones of the ferrocene series with urea in *i*-PrOH in the presence of *t*-BuOK. The structures of the compounds prepared were studied by ^{1}H and ^{13}C NMR and IR spectroscopy, and also by single crystal X-ray diffraction.

Urea, thiourea, and their derivatives are used in synthesis of pyrimidine derivatives, which are components of many biologically active substances and nucleic acids [1]. In the metallocene series, the reactions of thiourea [2–6] and phenylthiourea [6] with ferrocenyl-substituted chalcones have been reported, but there is no unambiguous opinion on the structure of the products. The reactions were performed in alcohol in the presence of sodium alcoholates with refluxing [3, 4, 6] or under ultrasonic treatment [5]. The products of condensation with thiourea were identified as tetrahydropyrimidinethiones **I** with the C=C double bond at the 4,5 position, and the products of condensation with phenylthiourea, as tetrahydropyrimidinethiones **II** with the C=C double bond at the 3,4 position (Scheme 1).

Scheme 1.



I **II**

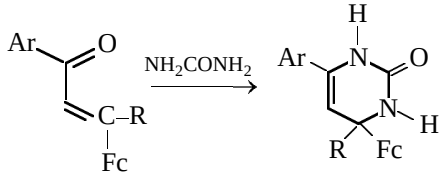
R^1 = Fc, R^2 = H, R^3 = Ar; R^1 = Ar, R^2 = H, R^3 = Fc; R^1, R^2 = FcCH=C$(CH_2)_3$; R^3 = Fc, Fc = $C_5H_5FeC_5H_4$.

With urea and its derivatives, the related condensations were not performed. The prospects for applying the expected products, e.g., as physiologically active substances make it interesting to prepare ferrocenyl-substituted tetrahydropyrimidinones and to study in detail their structure and chemical behavior.

In this work we studied the reactions of urea with α,β-unsaturated linear and cyclic ketones containing the ferrocenyl substituent.

We found that urea reacts with chalcones **IIIa**–**IIId** on refluxing in absolute isopropyl alcohol in the presence of *t*-BuOK to give 1,2,3,6-tetrahydropyrimidin-2-ones **IVa**–**IVd**, respectively, in ~70–80% yields (Scheme 2).

Scheme 2



IIIa–**IIId** **IVa**–**IVd**

R = H, A = 4-$CH_3OC_6H_4$ (**a**); R = H, Ar = 4-BrC_6H_4 (**b**); R = H, Ar = Fc (**c**); R = CH_3, Ar = Fc (**d**).

The isolated pyrimidin-2-ones **IVa**–**IVd** are the only reaction products. These are finely crystalline pale yellow substances stable in storage in the solid state. In common organic solvents ($CHCl_3$, CH_2Cl_2, CH_3COCH_3, CH_3COOEt, C_6H_6, etc.), they rapidly decompose, but in basic solvents (pyridine, morpholine, dimethylformamide, etc.) they are stable.

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Table 1. ^{1}H NMR data for the compounds prepared, δ, ppm (*J*, Hz)

Comp. no.	C_5H_5 (s)	C_5H_4 (m)	CH_3, CH_2	CH	NH	Ar
IVa[a]	4.18 (5H)	4.14 (2H), 4.19 (1H), 4.23 (1H)	3.81 (3H)	4.94 d.d (1H, *J* 2.0, 4.4), 5.12 m (1H, *J* 4.4)	6.63 br.s (1H), 8.20 s (1H)	6.92 d (2H, *J* 8.7), 7.49 d (2H, *J* 8.7)
IVb[a]	4.19 (5H)	4.17 (3H), 4.23 (1H)	–	5.01 d (1H, *J* 0.9), 5.16 d (1H, *J* 0.9)	6.15 br.s (1H), 8.15 s (1H)	7.43 d (2H, *J* 8.3), 7.52 d (2H, *J* 8.3)
IVc[a]	4.18 (5H), 4.21 (5H)	4.14 (2H), 4.27 (2H), 4.71 (4H)	–	3.8 4.72 br.s (1H), 5.14 d (1H, *J* 0.9)	6.91 br.s (1H), 8.01 s (1H)	–
IVd[a]	4.17 (5H), 4.24 (5H)	4.13 (2H), 4.32 (2H), 4.69 (4H)	1.78 (3H)	4.80 br.s (1H), 5.17 s (1H)	6.99 br.s (1H) 8.20 s (1H)	–
VIa[a]	4.22 (5H)	4.12 (3H), 4.16 (1H)	2.24 t (2H, *J* 7.5), 2.72 d.t (2H, *J* 7.5, 15.0)	4.64 br.s (1H)	6.85 br.s (1H), 8.41 s (1H)	7.15 m (3H), 7.50 m (1H)
VIb[a]	4.21 (5H)	4.12 (3H), 4.16 (1H)	2.20 t (2H, *J* 8.1), 2.66 t (2H, *J* 8.1), 3.76 (3H)	4.62 d (1H, *J* 1.0)	6.82 br.s (1H), 8.41 s (1H)	6.91 m (1H), 7.15 m (2H)
VIc[b]	3.62 (5H)	3.50 (3H), 3.55 (1H)	2.08–2.25 m (4H), 3.14 (3H)	4.11 br.s (1H)	6.13 br.s (1H), 7.90 br.s (1H)	6.16 d (1H, *J* 8.8), 6.98 d (1H, *J* 8.8), 7.36 s (1H)
VId[c]	4.22 (5H)	4.12 (3H), 4.15 (1H)	2.21 t (2H, *J* 7.8), 2.63 q (2H, *J* 7.8), 3.73 (3H)	4.64 d (1H, *J* 1.2)	6.84 br.s (1H), 8.58 s (1H)	6.70 d.d (1H, *J* 2.1, 8.4), 7.05 d (1H, *J* 8.4), 7.12 d (1H, *J* 2.1)
VIe[a,b]	4.19 (5H)	4.07 (2H), 4.11 (1H), 4.14 (1H)	2.14 m (2H), 2.58 m (2H), 2.17 (3H), 2.81 (3H)	4.63 br.s (1H)	6.76 br.s (1H), 8.29 s (1H)	6.81 s (1H), 7.23 s (1H)
VIIIa[a,c]	4.20 (5H)	4.11 (1H), 4.12 (2H), 4.13 (1H)	1.25 m (1H), 1.45 m (3H), 1.63 m (2H), 1.93–2.22 m (4H)	4.42 d (1H, *J* 2.4)	5.87 d (1H, *J* 2.4), 7.79 s (1H)	–
VIIIb[a,c]	4.12 (5H), 4.22 (5H)	4.11 (1H), 4.14 (3H), 4.26 (1H), 4.29 (2H), 4.43 (1H)	1.24 m (2H), 1.69 m (3H), 2.24 m (3H)	4.39 d (1H, *J* 3.0), 6.36 s (1H)	6.68 d (1H, *J* 3.0), 7.72 s (1H)	–
VIIIc[c]	4.12 (5H), 4.25 (5H)	4.14 (1H), 4.23 (2H), 4.33 (2H), 4.41 (1H), 4.51 (1H), 4.56 (1H)	1.63–2.38 m (6H)	4.98 d (1H, *J* 0.9), 6.07 s (1H)	6.40 br.s (1H), 7.92 s (1H)	–
VIIId[d]	4.12 (5H), 4.28 (5H)	4.18 (2H), 4.34 (2H), 4.46 (4H)	2.40 m (1H), 2.54 m (1H), 2.74 m (2H)	5.11 s (1H), 6.70 s (1H)	6.97 s (1H), 9.92 s (1H)	–
VIIIe[a,b]	4.12 (5H), 4.23 (5H)	4.10 (2H), 4.16 (2H), 4.20 (2H), 4.25 (2H)	2.24 (3H), 2.71 d (2H, *J* 8.5), 2.86 d (2H, *J* 8.5)	4.56 d (1H, *J* 0.8), 6.59 s (1H)	6.69 d (1H, *J* 0.8), 8.29 s (1H)	–
X[a,b]	4.19 (5H)	4.07 (2H), 4.10 (1H), 4.17 (1H)	1.40–2.00 m (8H)	2.38 m (1H), 4.63 d (1H, *J* 1.8)	6.55 d (1H, *J* 1.8), 8.30 s (1H)	–
XII[a,b]	3.76 (5H), 3.97 (5H)	3.77 (2H), 3.87 (2H), 3.89 (2H), 3.93 (1H), 4.10 (1H)	1.80 m (2H), 2.10 m (2H), 2.16 (3H)	3.63 m (1H), 3.70 m (1H), 4.26 br.s (1H), 6.71 s (1H)	6.83 br.s (1H), 8.32 s (1H)	–

Note: The ^{1}H NMR spectra were recorded in the following solvents: [a] $(CD_3)_2CO$, [b] $DCON(CD_3)_2$, [c] $CDCl_3$, and [d] C_5D_5N.

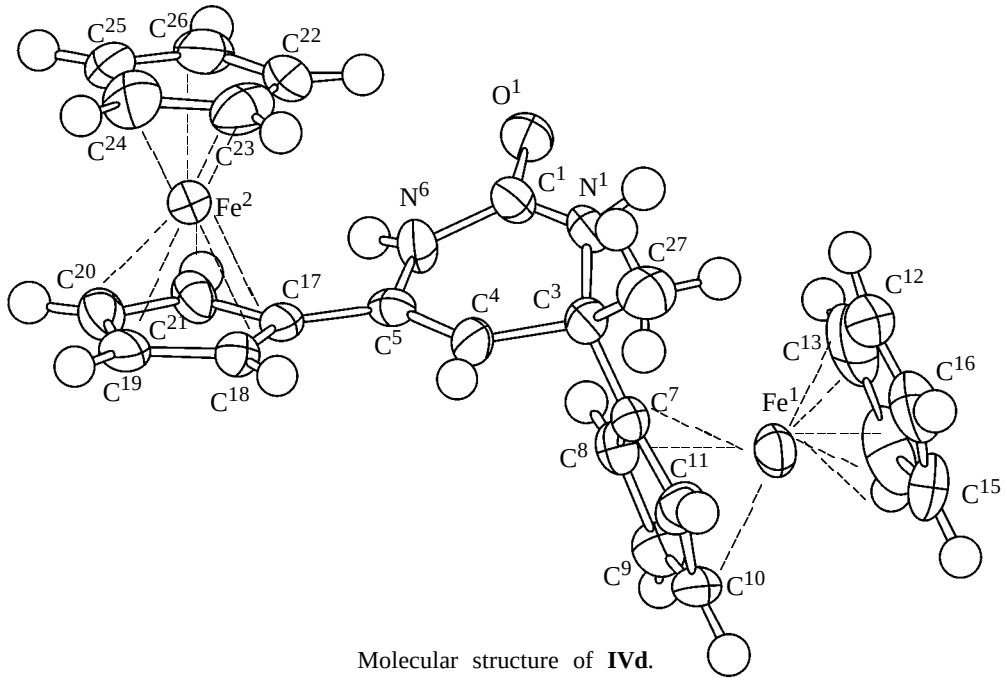
Table 2. ^{13}C NMR data for **IVb, IVc, VIa, VIc–VIe, VIIIa**, and **VIIIb** (75 MHz); **VIIIc, VIIIe, X**, and **XII** (9.0 kHz, solid state)], δ_C, ppm

Comp. no.	C_5H_5	C_5H_4	$C_{ipso}Fc$	CH_2, CH_3	C	CH=	CH	C=O	CN	Ar
IVb[a]	68.6	65.3, 65.4, 67.15, 67.2	91.3	–	121.5, 127.6	98.1	50.3	134.2	132.7	126.3, 130.7
IVc[a]	68.4, 69.2	65.3, 65.4, 65.7, 66.8, 67.4, 67.5, 68.7, 68.8	79.8, 92.7	–	–	94.2	49.8	153.3	134.2	–
VIa[a]	68.6	65.2, 66.1, 67.1, 67.5	92.9	24.4, 27.8, 54.5	108.9, 127.5, 128.9	–	53.4	154.2	135.5	121.1, 126.3, 127.4, 127.5
VIc[b]	68.2	64.8, 66.1, 66.8, 67.2	93.1	24.1, 28.1, 55.3	121.8, 121.9, 121.95, 153.5	–	53.8	158.7	127.7	106.2, 110.3, 113.6
VId[a]	68.5	65.2, 66.1, 67.1, 67.5	92.8	24.8, 26.9, 55.3	127.2, 127.6, 128.2, 154.2	–	53.5	158.2	130.0	107.1, 109.3, 112.9
VIe[a,b]	68.5	65.2, 66.2, 67.1, 67.4	93.1	19.0, 20.5, 23.4, 24.2	108.2, 127.8, 128.8, 130.5, 134.2	–	53.7	154.3	134.5	119.9, 129.9
VIIIa[a,c]	68.6	66.9, 67.0, 69.2, 69.6	78.5	24.1, 25.4, 28.4, 29.9, 30.1	107.5	–	54.2	152.7	131.6	–
VIIIb[a,c]	66.5, 67.0	63.3, 64.2, 64.9, 65.5, 66.2, 66.4, 66.8, 68.2	78.8, 91.3	23.8, 24.1, 27.6, 28.2	123.7, 129.5	107.9	53.9	152.0	131.9	–
VIIIc[d]	68.1, 69.2	63.0–75.0	82.8, 93.1	23.7, 25.0, 26.5	107.6, 127.5	113.0	55.9	156.9	128.3	–
VIIId[d]	70.1, 70.4	65.2–75.0	83.3, 92.9	47.4, 52.2, 54.0	118.4, 126.1	114.5	55.1	156.7	127.4	–
X[d]	69.8	64.2, 67.8	95.4	25.0–32.0	–	–	52.2, 53.9	157.1	122.7, 135.9	– –
XII[d]	68.5, 69.9	65.0–76.0	82.8, 92.6	32.5, 33.4, 36.2	121.5, 125.6	112.0	52.9, 58.9, 69.5	157.5	127.9	

The ^{13}C NMR spectra were recorded in the following solvents: [a] $(CD_3)_2SO$, [b] $DCON(CD_3)_2$, [c] $CDCl_3$, and [d] neat sample, solid phase.

Table 3. IR data (KBr) for the compounds prepared

Comp. no.	ν, cm^{-1}
IVa	769, 800, 1027, 1180, 1246, 1295 1459, 1513, 1613, 1681, 2836, 2962, 3096, 3220, 3330, 3462
IVb	748, 816, 1005, 1149, 1297, 1467, 1561, 1620, 1684, 2923, 3091, 3230, 3423
IVc	701, 749, 813, 999, 1103, 1160, 1288, 1436, 1482, 1584, 1693, 2843, 2942, 3093, 3224, 3429
IVd	689, 759, 817, 1002, 1105, 1241,1290, 1437, 1573, 1673, 2828, 2920, 2976, 3090, 3255, 3367
VIa	723, 768, 808, 820, 1028, 1108, 1269, 1294, 1390, 1450, 1481, 1560, 1681, 2832, 2934, 3098, 3223, 3331, 3426
VIb	750, 789, 823, 1035, 1111,1218, 1263, 1295, 1344, 1432, 1468, 1577, 1680, 2835, 2934, 3090, 3230, 3428
VIc	769, 814, 1001, 1037, 1125, 1156, 1252, 1276,1378, 1426, 1467, 1592, 1647, 1680, 2835, 2933, 3091, 3223, 3433
VId	772, 819, 1033, 1120, 1220, 1295, 1473, 1484, 1578, 1607, 1679, 2831, 2936, 3093, 3231, 3430
VIe	767, 811, 1008, 1027, 1106, 1228, 1298, 1395, 1462, 1480, 1654, 1678, 2830, 2923, 3101, 3226, 3429
VIIIa	763, 775, 820, 1027, 1075, 1214, 1301, 1393, 1443, 1481, 1647, 1684, 2846, 2921, 3097, 3254, 3342
VIIIb	672, 750, 812, 997, 1104, 1220, 1298, 1325, 1458, 1470, 1656, 1682, 2851, 2925, 3091, 3234, 3428
VIIIc	670, 769, 819, 997, 1105, 1220, 1296, 1330, 1423, 1467, 1637, 1681, 2830, 2933, 3093, 3240, 3433
VIIId	679, 754, 816, 1001, 1104, 1293, 1351, 1414, 1458, 1630, 1677, 2847, 2923, 3092, 3214, 3438
VIIIe	689, 756, 818, 893, 1001, 1067, 1129, 1293, 1370, 1465, 1623, 1681, 2767, 2938, 3091, 3252, 3430
X	762, 814, 957, 1027, 1106, 1197, 1236, 1369, 1451, 1475, 1624, 1681, 2868, 2939, 3099, 3257, 3327
XI	695, 745, 830, 1002, 1157, 1220, 1262, 1287, 1324, 1402, 1463, 1627, 1680, 2180, 2866, 2932, 3092, 3344, 3440



Molecular structure of **IVd**.

The compounds were characterized by IR and ^{1}H and ^{13}C NMR spectroscopy and by elemental analysis (Tables 1–4); the data obtained confirm the suggested structures. In particular, the ^{1}H NMR spectrum of each of compounds **IVa**–**IVd** contains two NH proton signals, characteristic signals of olefinic protons at δ 5.12, 5.16, and 5.14 ppm with $^3J_{H^5H^6}$ 4.4, 1.2, and 0.9 Hz for **IVa**–**IVc**, respectively (for **IVd**, a singlet at 5.14 ppm), H^6 signal (compounds **IVa**–**IVc**), and cyclopentadienyl proton signals.

The structure of **IVd** was determined by single crystal X-ray diffraction. The required crystal was obtained by crystallization from morpholine. It was found that compound **IVd**, indeed, has the structure of 6-methyl-4,6-diferrocenyl-1,2,3,6-tetrahydropyrimi-

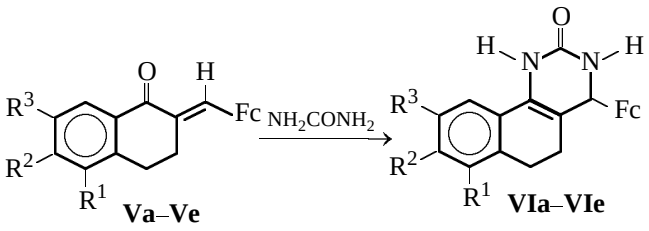
Table 4. Yields, melting points, and elemental analyses of **IVa**–**IVd, VIa**–**VIe, VIIIa**–**VIIIe, X**, and **XII**

Comp. no.	Yield, %	mp, °C	Found, %				Formula	Calculated, %			
			C	H	Fe	N		C	H	Fe	N
IVa	71	233–234	64.81	5.43	14.17	7.05	$C_{21}H_{20}FeN_2O_2$	64.97	5.20	14.38	7.21
IVb	68	191–193	55.10	3.84	12.91	6.23	$C_{20}H_{17}BrFeN_2O$	54.94	3.92	12.78	6.40
IVc	75	181–182	64.78	4.92	24.12	5.83	$C_{24}H_{22}Fe_2N_2O$	64.84	4.76	23.97	6.00
IVd	80	255–256	62.61	5.09	23.20	5.71	$C_{25}H_{24}Fe_2N_2O$	62.53	5.04	23.26	5.83
VIa	92	264–265	68.54	5.39	14.68	7.41	$C_{22}H_{20}FeN_2O$	68.77	5.25	14.53	7.23
VIb	87	282–284	66.84	5.23	13.24	6.54	$C_{23}H_{22}FeN_2O_2$	66.68	5.35	13.49	6.76
VIc	91	266–267	66.73	5.49	13.70	6.44	$C_{23}H_{22}FeN_2O_2$	66.68	5.35	13.49	6.76
VId	86	313–314	66.51	5.52	13.58	6.53	$C_{23}H_{22}FeN_2O_2$	66.68	5.35	13.49	6.76
VIe	92	293–295	69.98	6.02	13.72	6.93	$C_{24}H_{25}FeN_2O$	69.74	6.10	13.51	6.78
VIIIa	91	291–293	64.97	6.45	16.08	8.12	$C_{19}H_{22}FeN_2O$	65.16	6.33	15.95	8.00
VIIIb	94	295–297	66.08	5.37	20.28	5.26	$C_{30}H_{30}Fe_2N_2O$	65.96	5.53	20.45	5.13
VIIIc	90	263–265	65.67	5.21	21.04	5.11	$C_{29}H_{28}Fe_2N_2O$	65.44	5.30	20.99	5.26
VIIId	87	278–280	64.76	4.87	21.69	5.23	$C_{28}H_{26}Fe_2N_2O$	64.90	5.06	21.55	5.40
VIIIe	88	257–259	63.79	5.23	20.27	7.80	$C_{29}H_{29}Fe_2N_3O$	63.65	5.34	20.41	7.67
X	89	284–286	62.97	5.74	15.28	11.72	$C_{19}H_{21}FeN_3O$	62.83	5.83	15.37	11.56
XII	85	295–297	63.83	5.94	14.71	10.92	$C_{20}H_{23}FeN_3O$	63.68	6.14	14.80	11.13

din-2-one. The general view of the molecule of **IVd** is shown in the figure; the main geometric parameters are given in Table 5 and do not require special comments.

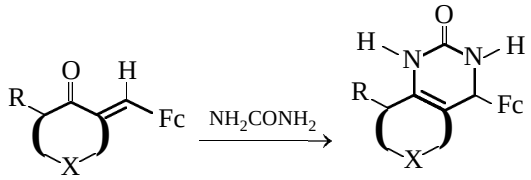
We studied the behavior in this reaction of a series of mono- and bisferrocenalacetones of the carbo- and heterocyclic series (compounds **Va**–**Ve**, **VIIa**–**VIIe**, **IX**, **XI**) and found that, in all the cases, the reactions are fast and give polycyclic ferrocenyltetrahydropyrimidin-2-ones **VIa**–**VIe**, **VIIIa**–**VIIIe**, **X**, and **XII**, respectively, in high yields (Scheme 3).

Scheme 3.



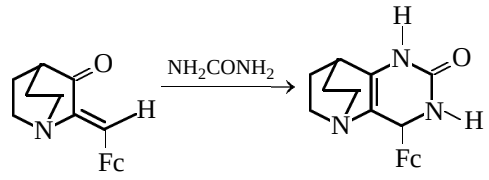
Va–**Ve** **VIa**–**VIe**

$R^1 = R^2 = R^3 = H$ (**a**); $R^1 = CH_3O$, $R^2 = R^3 = H$ (**b**); $R^1 = R^3 = H$, $R^2 = CH_3O$ (**c**); $R^1 = R^2 = H$, $R^3 = CH_3O$ (**d**); $R^1 = R^3 = CH_3$, $R^2 = H$ (**e**).

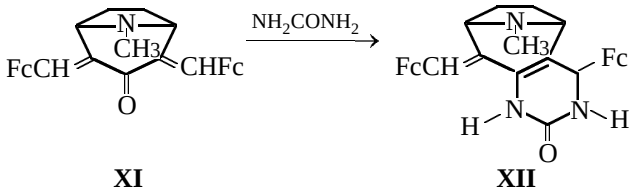


VIIa–**VIIe** **VIIIa**–**VIIIe**

R = H, X = $(CH_2)_4$ (**a**); R = FcCH=, X = $(CH_2)_4$ (**b**); R = FcCH=, X = $(CH_2)_3$ (**c**); R = FcCH=, X = $(CH_2)_2$ (**d**); R = FcCH=, X = $CH_2N(CH_2)CH_2$ (**e**).



IX **X**



XI **XII**

The structures of **IVa**–**IVe**, **VIIIa**–**VIIIe**, **X**, and **XII** were unambiguously proved by IR and ^1H and ^{13}C NMR spectra, and also by elemental analysis (Tables 1–4). The position of the C=C double bond in the tetrahydropyrimidin-2-one ring is confirmed by the multiplicity and position of the proton signals of the substituted cyclopentadienyl ring of the Fc moiety [7]; all or some of them are shifted upfield relative to the C_5H_5 singlet, as in products **IVa**–**IVd**. The signals of the quaternary carbon atoms in the ^{13}C NMR spectra additionally confirm the suggested structures.

Table 5. Selected bond lengths *d* and bond angles ω in the molecule of **IVd**

Bond	*d*, Å	Bond angle	ω, deg
O^1–C^1	1.230(4)	$O^1C^1N^1$	123.5(4)
C^1–N^1	1.354(5)	$O^1C^1N^6$	114.5(4)
C^1–N^6	1.360(4)	$C^1N^1C^3$	126.4(3)
N^1–C^3	1.474(5)	$N^1C^3C^4$	106.8(3)
C^{11}–C^{14}	1.519(16)	$N^1C^3C^7$	110.3(3)
C^3–C^4	1.512(5)	$C^4C^3C^7$	109.0(3)
C^3–C^7	1.524(5)	$C^5C^4C^3$	123.2(4)
C^4–C^5	1.333(5)	$C^4C^5N^6$	117.6(4)
C^5–N^6	1.407(4)	$C^1N^6C^5$	124.5(4)
C^5–C^{17}	1.469(5)	$N^1C^3C^{27}$	108.5(3)
C^7–C^8	1.410(5)	$C^4C^3C^{27}$	110.0(3)
C^{17}–C^{18}	1.414(5)	$C^8C^7C^{11}$	106.3(4)
C^{17}–C^{21}	1.422(5)	$C^8C^7C^3$	126.7(4)
C^7–C^{11}	1.412(5)	$C^{11}C^7C^3$	126.4(4)
C^3–C^{27}	1.522(5)	$C^8C^7Fe^1$	69.7(2)
C^7–Fe^1	2.046(4)	$C^5C^{17}Fe^{34}$	122.1(3)

The regioselectivity of formation of **VIa**–**VIe**, **VIIIa**–**VIIIe**, **X**, and **XII** is very high. All these compounds were isolated as single isomers. The content of possible alternative isomers was too low for their reliable detection and identification.

EXPERIMENTAL

The IR spectra were taken on a Specord IR-75 spectrometer in KBr. The ^1H and ^{13}C NMR spectra of solutions were measured on a Varian Unity-Inova spectrometer [300 and 75 MHz, respectively; solvents $CDCl_3$, $(CD_3)_2SO$, $DCON(CD_3)_2$, and C_5D_5N; internal reference Me_4Si]. The ^{13}C NMR spectra of solid **VIIIc**, **VIIIe**, **X**, and **XII** were taken on a Bruker DMX-500 spectrometer (9.3 kHz). The ^1H and ^{13}C NMR and IR data are given in Tables 1–3. For the chromatography of the starting chalcones, we used columns packed with Al_2O_3 (Brockmann grade III); the eluent was hexane–ether, 1 : 2 (by volume).

The following chemicals (Aldrich) were used: ferrocenecarbaldehyde, 99%; cyclohexanone, 99%; cycloheptanone, 99%; 1-methyl-4-piperidone, 97%;

Table 6. Crystallographic data and parameters of X-ray diffraction experiment for **IVd**

Parameter	Value
Formula	$C_{22}H_{24}Fe_2N_2O$
Molecular weight, g mol^{-1}	480.16
Temperature, K	291(2)
Crystal system	Monoclinic
Space group	*C*2/*c*
a, Å	22.3342(15)
b, Å	14.7506(10)
c, Å	14.6421(10)
α, deg	90.0
β, deg	123.21
γ, deg	90.0
V, Å^3	4035.9(5)
Z	8
d_{calc}, g cm^{-3}	1.580
Absorption coefficient, mm^{-1}	1.458
F(000)	1984
Radiation	MoK_α
λ, Å	0.71073
Monochromator	Graphite
θ, deg	1.76–24.99
Total number of reflections	16354
Number of unique reflections with $I > 2\sigma(I)$	3552
R_1	0.0479
WR_2	0.0591
R_{int}	0.0755
Number of refined parameters	272
Weight scheme	$w = 1/[\sigma^2(Fo^2) + (0.1693P)^2 + 10.77P]$, where $P = (Fo^2 + 2Fc)/3$
Goodness of fit (full-matrix least-squares method on F^2)	0.908
Residual electron density, e Å^{-3}, ρ_{min}/ρ_{max}	–0.386/0.733

4-methoxybenzaldehyde, 98%; acetylferrocene, 95%; 4-bromobenzaldehyde, 99%; 3-quinuclidinone hydrochloride, 97%; tropinone, 99%; cyclopentanone, 99%; α-tetralone, 98%; 5-methoxy-1-tetralone, 97%; urea, 99%; 6-methoxy-1-tetralone, 99%; 7-methoxy-1-tetralone, 99%; 5,7-dimethyl-1-tetralone, 97%; potassium *tert*-butylate, 95%.

The unit cell parameters and reflection intensities were measured on a Bruker-Smart APEX-CCD diffractometer. The crystallographic data and parameters of the X-ray diffraction experiment and refinement are given in Table 6. The structure of **IVd** was solved by the direct method and refined by the least-squares method in the full-matrix anisotropic approximation for nonhydrogen atoms.

α,β-Unsaturated ketones IIIa–IIId, Va–Ve, VIIc–VIIe, IX, and XI were prepared from ferrocenecarbaldehyde and the corresponding ketones in aqueous-alcoholic alkali [8]. Compounds **VIIa** and **VIIb** were prepared by condensation of cycloheptanone with ferrocenecarbaldehyde in benzene in the presence of *t*-BuOK [9].

Synthesis of 6-ferrocenyl-1,2,3,6-tetrahydropyrimidin-2-ones IVa–IVd, VIa–VIe, VIIIa–VIIIe, X, and XII. A mixture of 10 mmol of chalcone **IIIa–IIId**, **Va–Ve**, **VIIc–VIIe**, **IX**, or **XI**, 25 mmol of urea, 20 mmol of *t*-BuOK, and 150 ml of *i*-PrOH was refluxed with stirring for 3–5 h until the bright red color of the chalcone disappeared and a yellow solution formed. This solution was quickly poured into 200 ml of water. The precipitate was filtered off, washed with water on the filter, and vacuum-dried. The finely crystalline products obtained were virtually pure ferrocenyltetrahydropyrimidinones, yields 70–90%. For additional purification, they were recrystallized from morpholine.

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