Intramolecular cyclization of *N*-(3-oxoalkenyl)phenylacetamides: synthesis of 3-phenyl-2(1*H*)-pyridones

Dmitry S. Goncharov,[a] Anna K. Garkushenko,[b] Alina P. Savelieva[b] and Alexander S. Fisyuk[b,c]*

[a] *Department of Pharmaceutical Chemistry, Omsk State Medical Academy, Lenin str., 12, 644043 Omsk, Russian Federation*
[b] *Department of Organic Chemistry, Omsk F.M. Dostoevsky State University, 55a Mira Ave, 644077 Omsk, Russian Federation*
[c] *Laboratory of New Organic Materials, Omsk State Technical University, 11 Mira Ave, 644050 Omsk, Russian Federation*
E-mail: fisyuk@chemomsu.ru

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Abstract
3-Phenyl-2(1*H*)-pyridinones were obtained by base-catalyzed intramolecular aldol-type cyclization of *N*-(3-oxoalkenyl)phenylacetamides. The effect of substituents in the starting compounds and the experimental conditions on the reaction course were established. It was shown that the transformations of *N*-(3-oxoalkenyl)amides in basic medium depend on structural and electronic factors as well as the reaction conditions.

Keywords: 2(1*H*)-Pyridinone, β-enaminone, *N*-(3-oxoalkenyl)amide, intramolecular cyclization

Introduction

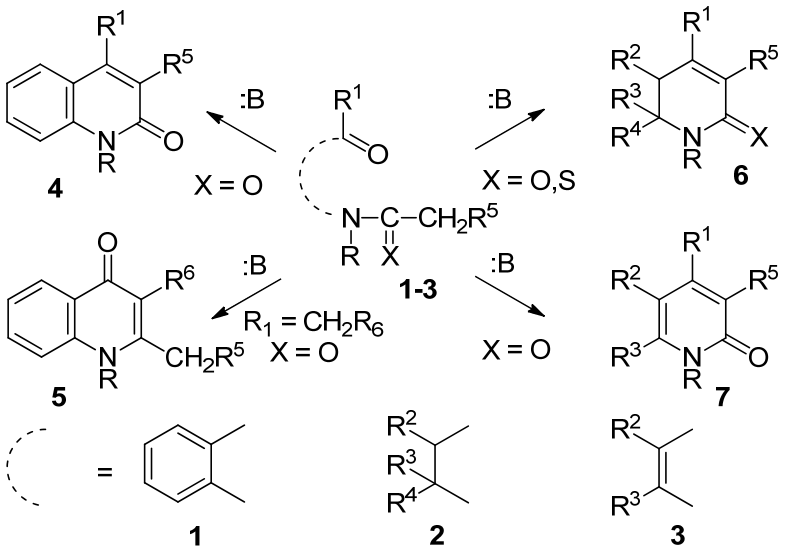
The 2(1*H*)-pyridinone ring is a structural fragment present within many alkaloids and other natural products.[1-7] Further, a significant number of compounds with a 2(1*H*)-pyridinone motif exhibit interesting activities against a number of biological targets, and are used as scaffolds in drug discovery.[8-11]

Methods for 2(1*H*)-pyridinone ring formation can be based on bi- and multicomponent reactions, intramolecular cyclizations and transformation of other hyterocycles.[12-16] While there are many ways of preparing 2-pyridinones, the study of new approaches to their synthesis is still desired due to the importance of this heterocycle.

Base-catalyzed intramolecular Knoevenagel condensation of bifunctional compounds **1-3**, which contain in the molecule both carbonyl and amide groups, is one of the ways to construct a

2-pyridinone ring. The cyclization of *o*-acylaminophenones **1** is well known as the Camps reaction and has been used for the synthesis of 2(1*H*)-quinolinones **4** and 4(1*H*)-quinolinones **5** since the 19th century.[17-22]

The regioselectivity of the reaction depends on the C-H acidity at the α-carbamoyl position of amides **1**. Increasing the α-carbamoyl acidity facilitates cyclization of amides **1** to 2(1*H*)-quinolinones **4**.



Scheme 1. Base-catalyzed intramolecular cyclization of *o*-acylaminophenones (1), *N*-(3-oxoalkyl)amides (**2**) and *N*-(3-oxoalkenyl)amides (**3**).

N-(3-oxoalkyl)amides **2** and *N*-(3-oxoalkenyl)amides **3** are analogues of *o*-acylaminoacetophenones **1**. A few years ago we developed the synthesis of 5,6-dihydro-2(1*H*)-pyridinones and -thiones **6** by intramolecular cyclization of *N*-(3-oxoalkyl)amides and -thioamides **2**[23-30] (Scheme 1). However, only a few examples of cyclization of *N*-(3-oxoalkenyl)amides **3** to the 2(1*H*)-pyridinones **7** are known.[31-36] Accordingly, the full synthetic potential of this reaction has not been studied.

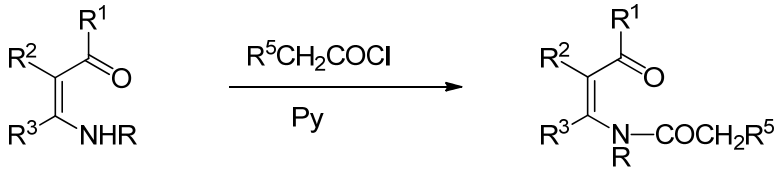
In this paper we have widened the range of starting materials in order to elucidate the influence of both structural and electronic effects on this cyclization and, as a result, have determined its limitations.

Results and Discussion

Previously, we have reported that *N*-(3-oxoalkenyl) phenylacetamide **3b** undergoes an aldol-type intramolecular ring closure to give 3-phenyl-2(1*H*)-pyridinone **7b**.[34] In order to establish the possibilities of this method we studied a similar cyclization for substituted *N*-(3-

oxoalkenyl)amides **3a-k**. Compounds **3a-k** were synthesized by acylation of enaminones **8a-k** with phenylacetyl chloride in the presence of pyridine in 39-85% yields (Table 1).

Table 1. Preparation of *N*-(3-oxoalkenyl)amides **3a-k** by N-acylation of enaminones **8a-k**



8a-k **3a-k**

Entry	Compound	R^1	R^2	R^3	R^5	R	Yield (%)
1	*Z*-**3a**	Me	H	Me	Ph	Me	45[a]
2	*Z*-**3b**	Me	H	Me	Ph	H	56
3	*Z*-**3c**	Me	$(CH_2)_4$		Ph	H	78
4	*Z*-**3d**	$(CH_2)_3$		Me	Ph	H	67
5	*Z*-**3e**	Ph	H	Me	Ph	H	85
6	*Z*-**3f**	C_6H_4-4-Me	H	Me	Ph	H	70
7	*Z*-**3g**	C_6H_4-4-Cl	H	Me	Ph	H	65
8	*Z*-**3h**	2-Naphthyl	H	Me	Ph	H	70
9	*Z*-**3i**	1-Naphthyl	H	Me	Ph	H	63
10	*Z*-**3j**	Me	H	Me	H	H	74
11	*E*-, *Z*-**3k**	$(CH_2)_4$		H	Ph	H	39

[a] Yield of the isolated product after column chromatography

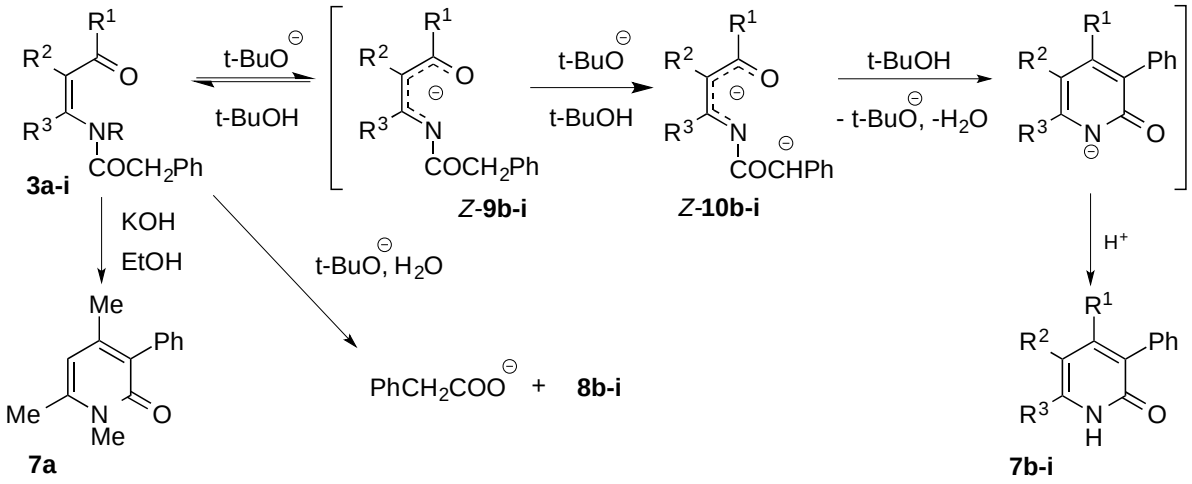
It was found that cyclization of compound *Z*-**3a** occurs at room temperature over 15 min by the action of potassium hydroxide in ethanol to give the 2-pyridinone **7a** in 94% yield (Table 2). On the other hand, the cyclization of secondary amide **3b** did not occur under the action of an alcoholic solution of alkali. According to TLC analysis, enaminone **8b**, the product of hydrolysis of *N*-(3-oxoalkenyl)amide **3b**, was identified in the reaction mixture. 3-Phenyl-2(1*H*)-pyridinones **7b-i** were obtained only in dry THF using 1.5 equivalents of potassium *tert*-butoxide and a cyclization time of 5-18 hours with 40-73% yields. However, in this case too, according to chromato-mass spectrometry data, enaminones **8a-i** were present in the reaction media. The appearance of enaminones **8a-i** as by-products was a result of hydrolysis of compounds **3a-i** by the water which was formed in the cyclization process.

Attempts to carry out cyclization of *N*-(3-oxoalkyl)acetamide **3j** which has low C-H acidity at the α-carbamoyl position, in basic conditions were unsuccessful. Secondary amide **3j** remained unchanged even on heating with potassium *tert*-butoxide. Apparently only *N*-(3-

oxoalkenyl)amides having significant C-H acidity at the α-carbamoyl position, are capable of cyclisation via an aldol condensation pathway in basic conditions. Increasing the α-carbamoyl acidity facilitates cyclization, while its low acidity makes cyclization impossible.

Thus the cyclization of the tertiary amide *Z*-**3a** proceeds under milder conditions and with better yield in comparison with secondary amides **3b-i**. In contrast to the tertiary amide *Z*-**3a**, the cyclization of secondary amides **3 b-i** to 2(1*H*)-pyridinones **7b-i** occurs via formation of dianions *Z*-**10b-i**. Deprotonation of N-hydrogen in the first stage of interaction of secondary amides **3b-i** with potassium *tert*-butoxide leads to formation of mesomeric anions **9b-i**, which have a delocalized negative charge in an β-enaminoketone fragment. As a result of the charge delocalization, the carbonyl group activity of anions **9b-i** decreases. This leads to a decrease of the cyclization rate of secondary amides **3b-i** in comparison with the tertiary example **3a**.

Table 2. Scope of the cyclization *N*-(3-oxoalkenyl)phenylacetamide **3** to synthesize 3-phenyl-2-(1*H*)-pyridinones **7**



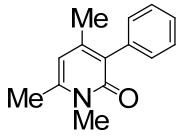
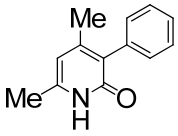
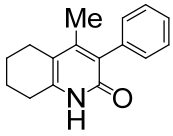
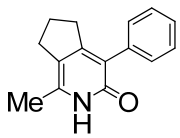
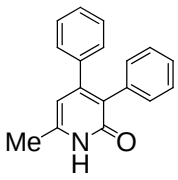
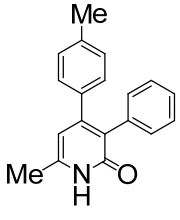
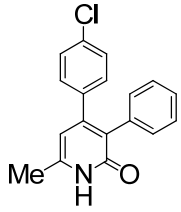
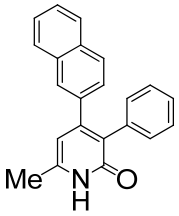
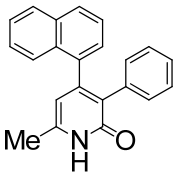
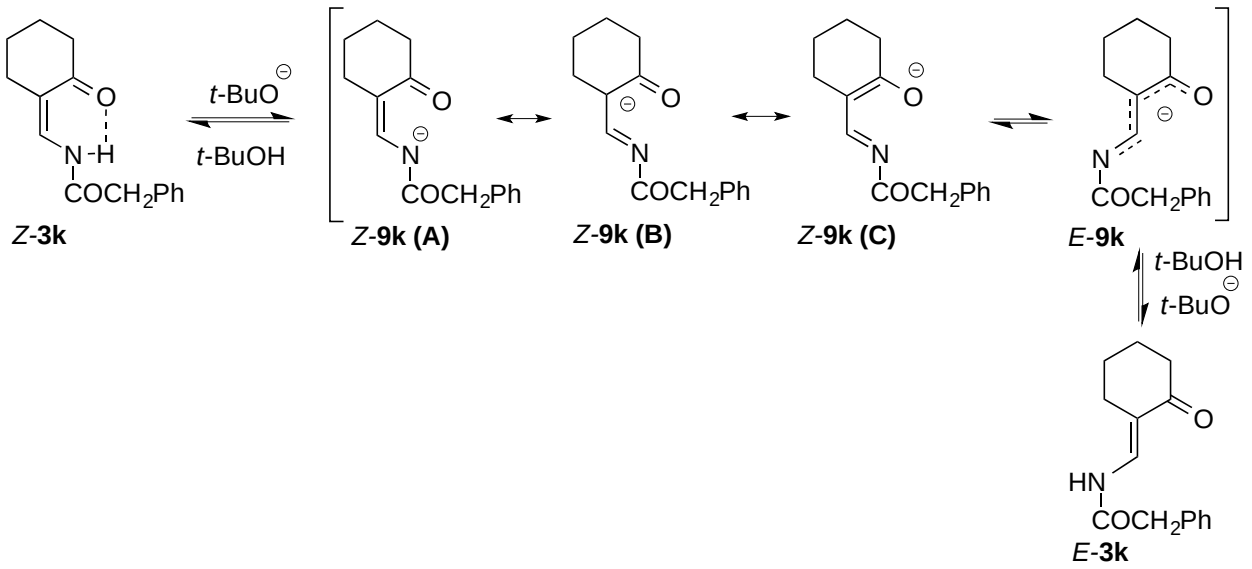
Entry	Product	Yield (%)	Entry	Product	Yield (%)	Entry	Product	Yield (%)
1	 (**7a**)	94	2	 **(7b)**	53	3	 (**7c**)	62
4	 **(7d)**	40	5	 **(7e)**	70	6	 (**7f**)	62

Table 2. Continued

Entry	Product	Yield (%)	Entry	Product	Yield (%)	Entry	Product	Yield (%)
7	 **(7g)**	73	8	 **(7h)**	65	9	 (**7i**)	60

The NH signals of *N*-(3-oxoalkenyl)amides **3b-j** are present in the low field region (δ(NH)~12) in the ^{1}H NMR spectra. This suggests a strong intramolecular hydrogen bonding between the amino group and the carbonyl oxygen. These compounds exist in the *Z-s-Z* form in $CDCl_3$ solution.[37,38] In contrast to compounds **3b-j**, a mixture of two isomers *Z*- and *E*-**3k** was obtained. *Z*- and *E*-isomers **3k** were separated by column chromatography on silica gel (Scheme 2). The isomer *Z*-**3k** had a broad NH signal at low field (δ(NH) = 11.32), but the NH signal of *E*-isomer **3k** was a doublet with a coupling constant $^3J_{NHCH}$ = 12.1 Hz and chemical shift 7.13 ppm. A broad absorbance of the NH of compound *Z*-**3k** was at 3200–3600 cm^{-1} in its IR spectrum. At the same time, the NH bond *E*-isomer **3k** is fixed as a narrow peak at 3246 cm^{-1}. According to ^{1}H NMR analysis, transformation of *Z*-isomer **3k** into a mixture of the *Z*- and *E*-isomers in the 6:1 ratio in solution of $CDCl_3$ takes two days. The addition of tetramethylguanidine led to rapid isomerization of compound **3k**. *Z*-Isomers **3k** are stabilized by an intramolecular hydrogen bond, which disappears after deprotonation of nitrogen atom. The nature of substituents influences both the charge distribution in mesomeric anion **9** and the stability of *Z*- and *E*-isomers. Alkyl groups stabilize a double bond. Therefore the double bond of anion *Z*-**9k** (R^3 = H) is more stable at the position C(2), C(3) (Scheme 2) in comparison with anions **9b-i** (R^3 =Alk) which have a more stable double bond at the position C(1), C(2) (Table 2). The stability of *Z*-and *E*-isomers **3** depends on the size of the substituents in the 3-oxoalkenyl fragment. Increasing the size of substituent R^3 should lead to a destabilization of the *E*-isomers of deprotonated *N*-(3-oxoalkenyl)amides **3** due to steric interactions with the carbonyl group, and vice versa - a decrease in the size of R^3 should stabilize the *E*-isomers, like the enaminones.[39-43] Thus the charge distribution in mesomeric anion **9** as well as the isomerization *Z*-**3k** to *E*-**3k** prevent the intramolecular ring closure of *N*-(3-oxoalkenyl)amide **3k** to a corresponding 2(1*H*)-pyridinone. Obviously, for this reason the cyclization of amides *Z*-**3k** does not occur under the action of potassium *tert*-butoxide in THF. In this case, the reaction product was a mixture of *Z*- and *E*-isomers **3k**.



Scheme 2. Interaction of *N*-(3-oxoalkenyl)phenylacetamide **3k** with potassium *t*-butoxide.

Conclusions

The intramolecular aldol-type condensation of bifunctional compounds containing carbonyl and amide groups is a general method for the synthesis of 2(1*H*)-pyridinones and derivatives. This approach can be successfully used for the preparation of 2(1*H*)-quinolinones, 5,6-dihydro-2(1*H*)-pyridinones. The transformations of *N*-(3-oxoalkenyl)amides in basic medium depend on structural, electronic factors and the reaction conditions. However, availability and diversity of starting materials and a simple experimental procedure make the *N*-(3-oxoalkenyl)phenylacetamides convenient precursors for the synthesis of 3-phenyl-2(1*H*)-pyridinones.

Experimental Section

General. The ^{1}H and ^{13}C NMR spectra were recorded on a Bruker ARX-300 or a Bruker DRX-400 instruments, with TMS as internal standard. The IR spectra were recorded on a INFRALUM FT-801 spectrometer. The mass spectra were recorded on an Agilent 6890 gas chromatograph coupled with a 5973N quadrupole mass-selective electron impact (EI) detector or the Thermo Scientific DSQ II GC/MS with TRACE GC Ultra (70 eV, evaporator temperature 200-250 ^{o}C). The reaction course and purity of the products were checked by thin-layer chromatography on Sorbfil UV-254 plates. Compounds **8a**,[39] **8b**,[40] **8c,d,**[41] **8e**,[40] **8j**,[42] **8k**[43] and **3j**[37] were prepared as previously reported. The physical constants and spectral data of the compounds **3b**, **7b** were given earlier by us.[34] Copies of NMR spectra are given in the supporting information.

General procedures for the synthesis of enaminones (8e-i). A slow stream of ammonia gas was passed through a solution of the relevant 1,3-diketone (0.1 mol) in dry toluene (20 mL) and a catalytic amount of formic acid. The mixture was heated under reflux and the H_2O formed was removed azeotropically using a Dean-Stark apparatus. After cooling the reaction mixture a precipitate of enaminone was filtered off and recrystallized.

3-Amino-1-(4-methylphenyl)but-2-en-1-one (8f). Yield 85%, light crystals, mp 89-90 ^oC (toluene) (lit.[41] mp 89-90 °C). IR (KBr): ν, cm^{-1} 3297, 3143 (NH_2), 1602 (C=O), 1535 (C=C). ^1H NMR (400 MHz, $CDCl_3$): δ 2.02 (3H, s, CH_3), 2.37 (3H, s, 4-CH_3), 5.39 (1H, br.s, NH_2), 5.71 (1H, s, –CH=), 7.17–7.23 (2H, m, H Ar), 7.75–7.81 (2H, m, H Ar), 10.16 (1H, br.s, NH_2). ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.4 (4-CH_3), 22.8 (=C-$\underline{C}H_3$), 92.1 (=CH–), 127.2, 128.9, 137.6, 141.1 (Ar), 162.8 (=$\underline{C}$-CH_3), 189.3 ($\underline{C}$OAr).

3-Amino-1-(4-chlorophenyl)but-2-en-1-one (8g). Yield 75%, light crystals, mp 128-129 ^oC (toluene) (lit.[42] mp 128-129 °C). IR (KBr): ν, cm^{-1} 3285, 3146 (NH_2), 1603 (C=O), 1529 (C=C). ^1H NMR (400 MHz, $CDCl_3$): δ 2.04 (3H, s, CH_3), 5.40 (1H, br.s, NH_2), 5.66 (1H, s, –CH=), 7.33–7.39 (2H, m, H Ar), 7.77–7.83 (2H, m, H Ar), 10.20 (1H, br.s, NH_2). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.8 (=C-$\underline{C}H_3$), 92.0 (=CH–), 128.4,128.5, 136.9, 138.6 (Ar), 163.5 (=$\underline{C}$-CH_3), 187.9 ($\underline{C}$OAr); Anal. Calcd for: $C_{10}H_{10}ClNO$ C, 61.39; H, 5.15; N, 7.16. Found: C, 61.42; H, 5.14; N, 7.20%.

3-Amino-1-(2-naphthyl)but-2-en-1-one (8h). Yield 83%, light crystals, mp 148-149 ^oC (toluene); IR (KBr): ν, cm^{-1} 3293, 3138 (NH_2), 1612 (C=O), 1530 (C=C). ^1H NMR (400 MHz, $CDCl_3$): δ 2.09 (3H, s, CH_3), 5.25 (1H, br.s, NH_2), 5.90 (1H, s, –CH=), 7.47–7.55 (2H, m, H Ar), 7.83–7.89 (2H, m, H Ar), 7.91–7.96 (1H, m, H Ar), 7.98–8.02 (1H, m, H Ar), 8.39 (1H, s, H-1′ Ar), 10.28 (1H, br.s, NH_2). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.8 (=C-$\underline{C}H_3$), 92.6 (=CH–), 124.2, 126.2, 127.1, 127.5, 127.6, 127.9, 129.2, 132.9, 134.7, 137.6 (Ar), 162.8 (=$\underline{C}$-CH_3), 189.3 ($\underline{C}$OAr); Anal. Calcd for: $C_{14}H_{13}NO$ C, 79.59; H, 6.20; N, 6.63. Found: C, 79.53; H, 6.21; N, 6.59%.

3-Amino-1-(1-naphthyl)but-2-en-1-one (8i). Yield 87%, light crystals, mp 159-160 ^oC (toluene); IR (KBr): ν, cm^{-1} 3287, 3133 (NH_2), 1617 (C=O), 1524 (C=C). ^1H NMR (400 MHz, $CDCl_3$): δ 1.98 (3H, s, CH_3), 5.46 (1H, br.s, NH_2), 5.50 (1H, s, –CH=), 7.43–7.54 (3H, m, H Ar), 7.61–7.65 (1H, m, H Ar), 7.83–7.88 (2H, m, H Ar), 8.41–8.45 (1H, m, H Ar), 10.22 (1H, br.s, NH_2). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.6 (=C-$\underline{C}H_3$), 97.0 (=CH–), 124.8, 125.1, 125.9, 126.1, 126.4, 128.2, 129.7, 130.3, 133.8, 140.4 (Ar), 162.8 (=$\underline{C}$-CH_3); 193.7 ($\underline{C}$OAr); Anal. Calcd for: $C_{14}H_{13}NO$ C, 79.59; H, 6.20; N, 6.63. Found: C, 79.59; H, 6.17; N, 6.64%.

General procedure for the synthesis of *N*-(3-oxoalkenyl)phenylacetamides (3a-k). Phenylacetyl chloride 1.623 g (10.5 mmol) was added dropwise to a solution of enamino ketone (10.0 mmol) and anhydrous pyridine (1 mL) in absolute $CHCl_3$ (15 mL) with stirring. The mixture was stirred for 1 h with cooling in ice and for 4-10 h at room temperature. $CHCl_3$ (10 mL) was then added and the reaction mixture was washed with 10% aq HCl solution (30 mL) and with H_2O until the wash water gave a neutral reaction. The organic phase was dried with

anhydrous Na_2SO_4, and the $CHCl_3$ distilled off. The compound was recrystallized from an EtOAc–petroleum ester mixture 4/7 (**3b**,**h**,**i**,**j**), *i*-PrOH (**3f**,**g**) or purified by column chromatography (Al_2O_3, CH_2Cl_2-hexane (**3c-e**), hexane–EtOAc 5/1 (*Z*-**3a**) or on silica gel L 40/100, $CHCl_3$–EtOAc 1/1 (*E,Z*-**3k**).

***N*-methyl-*N*-[(*Z*)-1-methyl-3-oxo-but-1-enyl]-2-phenylacetamide (*Z*-3a).** Yield 45%, light yellow oil; IR (KBr): ν, cm^{-1} 1693 (C=O), 1612 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 2.14 (3H, s, $COCH_3$), 2.22 (3H, s, =C-CH_3), 3.06 (3H, s, N-CH_3), 3.70 (2H, s, CH_2), 5.97 (1H, s, =CH), 7.20-7.35 (5H, m, C_6H_5). ^{13}C NMR (100 MHz, $CDCl_3$): δ 18.5 (=C-$\underline{C}H_3$), 32.0 (N-CH_3), 34.2 (CO$\underline{C}H_3$), 41.4 (C_6H_5-$\underline{C}H_2$), 124.7 (=$\underline{C}$HCO), 126.9 (C_6H_5, C-4'), 128.7 (C_6H_5, C-2', C-6'), 128.8 (C_6H_5, C-3', C-5'), 135.3 (C_6H_5, C-1'), 154.2 (=C-N), 169.8 (NHCO), 197.7 (C=O); Anal. Calcd for: $C_{14}H_{17}NO_2$ C, 72.70; H, 7.41; N, 6.06. Found: C, 72.77; H, 7.46; N, 6.12%.

***N*-[(Z)-2-acetylcyclohex-1-enyl]-2-phenylacetamide (*Z*-3c).** Yield 78%, light yellow oil; IR ($CHCl_3$): ν, cm^{-1} 3250-3150 (NH), 1697 (C=O), 1632 (C=O), 1585 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 1.54-1.66 (4H, m, 2H-4, 2H-5), 2.17 (3H, s, CH_3), 2.31-2.39 (2H, m, 2H-3), 2.91–3.03 (2H, m, 2H-2), 3.61 (2H, s, CH_2), 5.30 (1H, s, =CH), 7.22-7.29 (5H, m, C_6H_5), 12.86 (1H, br. s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.5 (CH_2), 22.0 (CH_2), 26.0 (CH_2), 28.6 (CH_2), 29.0 (CH_3); 46.1 (C_6H_5-$\underline{C}H_2$); 111.8 (=$\underline{C}$CO); 127.2 (C_6H_5, C-'), 128.8 (C_6H_5, C-2', C-6'), 129.4 (C_6H_5, C-3',C-5'), 133.4 (C_6H_5, C-1'), 152.6 (=C-N), 170.5 (NHCO), 202.5 (C=O); Anal. Calcd for: $C_{16}H_{19}NO_2$ C, 74.68; H, 7.44; N, 5.44. Found: C, 74.73; H, 7.36; N, 5.35%.

***N*-{1-[2-oxocyclopent-(*Z*)-ylidene]-ethyl}-2-phenylacetamide (*Z*-3d).** Yield 67%, light yellow crystals, mp 56-57 ^oC. IR (KBr): ν, cm^{-1} 3250-3150 (NH), 1717 (C=O), 1683 (C=O), 1615 (N–C=O. ^1H NMR (400 MHz, $CDCl_3$): δ 1.79-1.97 (2H, m, 2H-4), 2.24-2.48 (5H, m, 2H-5, CH_3), 2.49-2.66 (2H, m, 2H-3), 3.66 (2H, s, CH_2), 7.22-7.47 (5H, m, C_6H_5), 12.01 (1H, br.s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 18.5 (CH_3), 19.6 (CH_2), 27.2 (CH_2), 39.7 (CH_2), 45.6 (C_6H_5-$\underline{C}H_2$), 114.5 (=$\underline{C}$CO), 127.3 (C_6H_5, C-4'), 128.8 (C_6H_5, C-2',C-6'), 129.4 (C_6H_5, C-3',C-5'), 134.0 (C_6H_5, C-1'), 148.1 (=C-N), 170.4 (NHCO), 207.8 (C=O); Anal. Calcd for: $C_{15}H_{17}NO_2$ C, 74.05; H, 7.04; N, 5.76. Found: C, 74.12; H, 7.07; N, 5.81%.

***N*-[(*Z*)-1-methyl-3-oxo-3-phenylprop-1-en-1-yl]-2-phenylacetamide (*Z*-3e).** Yield 85%, white crystals, mp 75-76 ^oC. IR (KBr): ν, cm^{-1} 3250-3150 (NH), 1702 (C=O), 1624 (C=O), 1591 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 2.49 (3H, d, *J* 0.9 Hz, CH_3), 3.72 (2H, s, CH_2), 6.02 (1H, d, *J* 0.9 Hz, CH=), 7.26–7.55 (5H, m, Ar-H), 7.84–7.91 (5H, m, Ar-H), 12.88 (1H, br. s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.4 (=C-$\underline{C}H_3$); 45.6 (CO$\underline{C}H_2$); 102.1 (=CH), 127.3, 127.6, 128.5, 128.8, 129.4, 132.3, 133.8, 138.7 (Ar), 157.3 (=C-N), 170.7 (NHCO), 191.4 (COAr); Anal. Calcd for: $C_{18}H_{17}NO_2$ C, 77.40; H, 6.13; N, 5.01. Found: C, 77.48; H, 6.18; N, 5.07%.

***N*-[(*Z*)-1-methyl-3-(4-methylphenyl)-3-oxoprop-1-en-1-yl]-2-phenylacetamide *(Z*-3f).** Yield 70%, white crystals, mp 104-105.^oC. IR (KBr): ν, cm^{-1} 3463 (NH), 1703 (C=O), 1607 (C=O), 1588 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 2.19 (3H, s, 4-CH_3), 2.49 (3H, s, CH_3), 3.73 (2H, s, CH_2), 6.15 (1H, s, –CH=), 7.26–7.30 (2H, m, Ar-H), 7.36–7.39 (3H, m, Ar-H), 7.76–7.82 (4H, m, Ar-H), 12.93 (1H, s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 21.6 (4-CH_3), 22.5 (=C-$\underline{C}H_3$), 45.7 (CO$\underline{C}H_2$), 102.1 (=CH–); 127.1, 127.8, 128.8, 129.1, 129.4, 133.9, 136.1, 143.2 (Ar), 156.9 (=$\underline{C}$-

CH_3), 170.7 ($\underline{C}OCH_2$), 191.2 ($\underline{C}OAr$); Anal. Calcd for: $C_{19}H_{19}NO_2$ C, 77.79; H, 6.53; N, 4.77. Found: C, 77.83; H, 6.55; N, 4.80%.

***N*-[(*Z*)-3-(4-chlorophenyl)-1-methyl-3-oxoprop-1-en-1-yl]-2-phenylacetamide (*Z*-3g).** Yield 65%, white crystals, mp 110-111 °C. IR (KBr): ν, cm^{-1} 3455 (NH), 1708 (C=O), 1618 (C=O), 1594 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$)): δ 2.49 (3H, s, CH_3), 3.74 (2H, s, CH_2), 5.97 (1H, s, –CH=), 7.35–7.44 (7H, m, Ar-H), 7.79–7.83 (2H, m, Ar-H), 12.84 (1H, s, NH). ^{13}C NMR (100 MHz, $CDCl_3$)): δ 22.6 (=C-$\underline{C}H_3$), 45.7 (CO$\underline{C}H_2$), 101.7 (=CH–), 127.4, 128.8, 128.9, 129.1, 129.5, 133.7, 137.0, 138.7 (Ar), 158.0 (=$\underline{C}$-CH_3), 170.8 ($\underline{C}OCH_2$), 189.9 ($\underline{C}OAr$); Anal. Calcd for: $C_{18}H_{16}ClNO_2$ C, 68.90; H, 5.14; N, 4.46. Found: C, 68.94; H, 5.12; N, 4.51%.

***N*-[(*Z*)-1-methyl-3-(2-naphthyl)-3-oxoprop-1-en-1-yl]-2-phenylacetamide (*Z*-3h).** Yield 70%, white crystals, mp 99-100 °C. IR (KBr): ν, cm^{-1} 3460 (NH), 1710 (C=O), 1620 (N–C=O), 1598 (C=C). ^1H NMR (400 MHz, $CDCl_3$): δ 2.55 (3H, s, CH_3), 3.77 (2H, s, CH_2), 6.20 (1H, s, –CH=), 7.29–7.45 (5H, m, H Ar), 7.51–7.61 (2H, m, Ar-H), 7.85–8.00 (4H, m, Ar-H), 8.41 (1H, s, H-1′ Ar), 12.97 (1H, s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.5 (=C-$\underline{C}H_3$), 45.7 (CO$\underline{C}H_2$), 102.3 (=CH–), 123.9, 126.7, 127.4, 127.7, 128.1, 128.4, 128.8, 128.9, 129.4, 132.7, 133.9, 135.3, 136.1 (Ar), 157.3 (=$\underline{C}$-CH_3), 170.7 ($\underline{C}OCH_2$), 191.2 (COAr); Anal. Calcd for: $C_{22}H_{19}NO_2$ C, 80.22; H, 5.81; N, 4.25. Found: C, 80.25; H, 5.80; N, 4.19%.

***N*-[(*Z*)-1-methyl-3-(1-naphthyl)-3-oxoprop-1-en-1-yl]-2-phenylacetamide (*Z*-3i)**. Yield 63%, white crystals, mp 58-59 °C. IR (KBr): ν, cm^{-1} 3455 (NH), 1708 (C=O), 1618 (C=O), 1594 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 2.51 (3H, s, CH_3), 3.80 (2H, s, CH_2), 5.88 (1H, s, –CH=), 7.31–7.61 (8H, m, Ar-H), 7.67–7.71 (1H, m, Ar-H), 7.86–7.96 (2H, m, H Ar), 8.34–8.39 (1H, m, Ar-H), 12.90 (1H, s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.4 (=C-$\underline{C}H_3$), 45.8 (CO$\underline{C}H_2$), 106.6 (=CH–), 124.6, 125.6, 126.3, 126.4, 127.2, 127.3, 127.5, 128.5, 129.0, 129.6, 130.0, 131.5, 133.9, 138.3 (Ar), 157.1 (=$\underline{C}$-CH_3), 170.9 ($\underline{C}OCH_2$), 195.7 ($\underline{C}OAr$); Anal. Calcd for: $C_{22}H_{19}NO_2$ C, 80.22; H, 5.81; N, 4.25. Found: C, 80.18; H, 5.82; N, 4.29%.

***N*-[2-oxo-cyclohex-(*Z*,*E*)-ylidenemethyl]-2-phenylacetamide (*Z*,*E*-3k).** Yield 39%, light-yellow oil. Individual isomers were isolated by column chromatography (silicagel L 40/100, $CHCl_3$-EtOAc). *Z*-**3k**. White crystals, mp 85-86 °C (hexane). IR ($CHCl_3$): ν, cm^{-1} 3248 (NH), 1698 (C=O), 1655 (C=O), 1583 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$)): δ 1.63–1.80, 2.31–2.45 (8H, m, $(CH_2)_4$), 3.67 (2H, s, CH_2), 7.22 (1H, dt, *J* 10.7, 1.5 Hz, =CH), 7.28–7.42 (5H, m, C_6H_5), 11.32 (1H, m, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.4, 23.3, 28.8, 39.4 $(CH_2)_4$, 44.1 (C_6H_5-$\underline{C}H_2$), 113.5 (=$\underline{C}$CO), 127.7, 129.1, 129.4, 133.5 (C_6H_5), 133.9 (=CHN), 170.1 (NHCO), 203.7 (C=O). Anal. Calcd for: $C_{15}H_{17}NO_2$ C, 74.05; H, 7.04; N, 5.76. Found: C, 73.93; H, 7.06; N, 5.77%. *E*-**3k.** White crystals, mp 137-138 °C (EtOAc); IR ($CHCl_3$): ν, cm^{-1} 3412 (NH), 1712 (C=O), 1670 (C=O), 1551 (N–C=O). ^1H NMR (400 MHz, $CDCl_3$): δ 1.62–1.80, 2.01–2.07, 2.32–2.38 (8H, m, $(CH_2)_4$), 3.72 (2H, s, CH_2), 7.13 (1H, d, *J* 12,1 Hz, NH), 7.25–7.42 (5H, m, C_6H_5), 7.94 (1H, dt, *J* 12.1, 2.0 Hz, =CH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 22.5, 22.6, 24.0, 39.5 $(CH_2)_4$, 43.9 (C_6H_5-$\underline{C}H_2$), 116.5 (=$\underline{C}$CO), 128.0 (=CHN), 129.1, 129.4, 129.7, 133.5 (C_6H_5), 168.6 (NHCO), 199.0 (C=O); Anal. Calcd for: $C_{15}H_{17}NO_2$ C, 74.05; H, 7.04; N, 5.76. Found: C, 73.94; H, 7.04; N, 5.76%.

1,4,6-Trimethyl-3-phenyl-1*H*-pyridin-2-one (**7a**)**.** Powdered KOH 0.112 g (1.5 mmol) was added to a solution of compound Z-**3a** 0.232 g (1.0 mmol) in EtOH. The mixture was stirred for 15 min and then H_2O (10 mL) added, evaporated to 2/3, and cooled. The solid product was filtered off, and recrystallized from a mixture of solvents: EtOH-H_2O yielding 0.20 g (94%) of product with mp 118–119 ^oC. IR (KBr): ν, cm^{-1} 1642 (NC=O). ^1H NMR (400 MHz, $CDCl_3$): δ 1.99 (3H, s, 4-CH_3), 2.35 (3H, s, 6-CH_3), 3.53 (3H, s, 1-CH_3), 6.01 (1H, s, H-5), 7.22-7.42 (5H, m, C_6H_5). ^{13}C NMR (100 MHz, $CDCl_3$): δ 20.3 (6-CH_3), 20.8 (4-CH_3), 31.4 (1-CH_3), 109.4 (C-5), 127.0 (C_6H_5, C-4'), 127.8 (C-3), 128.1 (C_6H_5, C-3', C-5'), 130.1 (C_6H_5, C-2', C-6'), 136.8 (C_6H_5, C-1'), 143.5 (C-6), 146.0 (C-4), 163.0 (C-2); *m/z* 213 $[M]^{+\cdot}$; Anal. Calcd for: $C_{14}H_{15}NO$ C, 78.84; H, 7.09; N, 6.57. Found: C, 78.89; H, 7.13; N, 6.54%.

General procedure for the synthesis of 3-phenylpyridin-2(1*H*)-one (7b-i). KO*t*-Bu (0.084 g, 0.75 mmol) was added with ice-cooling and stirring to a solution of phenylacetamide **3** (0.5 mmol) in absolute THF (4 ml). After 5-18 h (monitored by TLC) the solvent was evaporated, the residue triturated with H_2O and then neutralized with aq 5% AcOH solution. The product was filtered off, and recrystallized from MeOH (**7g**), EtOH (**7b-e**) or *i*-PrOH (**7f,h,i**).

4-Methyl-3-phenyl-5,6,7,8-tetrahydro-1*H*-quinolin-2-one (7c). The mixture was stirred for 5 hours. Yield 62%, white crystals, mp above 280 °C. IR (KBr): ν, cm^{-1} 1631 (NC=O), 3278 (NH). ^1H NMR (400 MHz, $CDCl_3$): δ 1.63-1.86 (4H, m, 2H-6, 2H-7), 1.93 (3H, s, CH_3), 2.35-2-2.61 (4H, m, 2H-5, 2H-8), 7.18-7.34 (3H, m, C_6H_5), 7.35-7.45 (2H, m, C_6H_5), 12.12 (1H, br. s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 17.3 (4-CH_3), 21.6, 22.8, 24.6, 27.1 (4CH_2), 114.1 (C-4), 126.8, 128.0, 130.4, 136.8, 136.8 (C_6H_5), 128.3 (C-3), 140.9 (C-6), 149.1 (C-4), 162.8 (C-2); *m/z* 239 $[M]^{+\cdot}$. Anal. Calcd for: $C_{16}H_{17}NO$ C, 80.30; H, 7.16; N, 5.85. Found: C, 80.38; H, 7.22; N, 5.91%.

1-Methyl-4-phenyl-2,5,6,7-tetrahydro-3*H*-cyclopenta[c]pyrindin-3-one (7d). The reaction mixture was stirred for 7 h. Yield 40%, white crystals, mp > 280 °C. IR (KBr): ν, cm^{-1} 1640 (NC=O), 3276 (NH). ^1H NMR (400 MHz, $CDCl_3$): δ 1.97 (2H, tt (q), *J* 7.2, 7.3 Hz, CH_2), 2.26 (3H,s, CH_3), 2.69 (2H, t, *J* 7.2 Hz, CH_2), 2.78 (2H , t, *J* 7.3 Hz, CH_2), 7.24-7.34, 7.35-7.53 (5H, m, C_6H_5), 12.78 (1H, br. s, NH). ^{13}C NMR (100 MHz, $CDCl_3$): δ 16.8 (6-CH_3), 25.6, 28.7, 33.5 (3CH_2), 121.2 (C-5), 123.0 (C-3), 126.8, 127.8, 129.8, 136.0 (C_6H_5), 137.3 (C-6), 157.8 (C-4), 164.1 (C-2); *m/z* 225 $[M]^{+\cdot}$. Anal. Calcd for: $C_{15}H_{15}NO$ C, 79.97; H, 6.71; N, 6.22. Found: C, 80.04; H, 6.82; N, 6.27%.

6-Methyl-3,4-diphenylpyridin-2(1*H*)-one (7e). The reaction mixture was stirred for 7 h.Yield 70%, white crystals, mp 228-229 C; IR (KBr): ν, cm^{-1} 3476, 2787 (NH), 1628 (N–C=O). ^1H NMR (400 MHz, DMSO-d_6): δ 2.32 (3H, s, 6-CH_3), 6.13 (1H, s, H-5), 7.00–7.24 (10H, m, H Ar), 12.86 (1H, br.s, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ 18.9 (6-CH_3), 108.7 (C-5), 126.6 (C-3), 126.1, 127.5, 131.3, 135.4, 127.5, 127.9, 128.9, 139.7 (Ar), 143.7 (C-6), 152.2 (C-4), 164.7 (C-2); Anal. Calcd for: $C_{18}H_{15}NO$ C, 82.73; H, 5.79; N, 5.36. Found: C, 82.76; H, 5.80; N, 5.42%.

6-Methyl-4-(4-methylphenyl)-3-phenylpyridin-2(1*H*)-one (7f). The reaction mixture was stirred for 10 h.Yield 62%, light crystals, mp 275 °C (decomp.); IR (KBr): ν, cm^{-1} 3453, 2767 (NH), 1621 (N–C=O). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.20 (3H, s, 4′-CH_3), 2.21 (3H, s, 6-CH_3), 6.02 (1H, s, H-5), 6.90–7.02 (6H, m, H Ar), 7.09–7.17 (3H, m, H Ar), 11.75 (1H, br.s, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ 18.9 (6-CH_3), 21.1 (4′-CH_3), 107.2 (C-5), 126.7 (C-3), 125.90, 127.7, 129.0, 129.1, 131.6, 136.7, 137.1, 137.2 (Ar), 144.2 (C-6), 150.8 (C-4), 162.9 (C-2); Anal. Calcd for: $C_{19}H_{17}NO$ C, 82.88; H, 6.22; N, 5.09. Found: C, 82.90; H, 6.25; N, 5.13%.

4-(4-Chlorophenyl)-6-methyl-3-phenylpyridin-2(1*H*)-one (7g). The reaction mixture was stirred for 7 h. Yield 73%, light crystals, mp 287 °C (decomp.); IR (KBr): ν, cm^{-1} 3451, 2765 (NH), 1622 (N–C=O). ^{1}H NMR (400 MHz, DMSO- d_6): δ 2.21 (3H, s, 6-CH_3), 6.04 (1H, s, H-5), 6.98–7.07 (4H, m, H Ar), 7.11–7.19 (3H, m, H Ar), 7.22–7.27 (2H, m, H Ar), 11.85 (1H, br.s, NH). ^{13}C NMR (100 MHz, DMSO- d_6): δ 18.9 (6-CH_3), 106.9 (C-5), 126.9 (C-3), 126.3, 127.9, 128.5, 131.0, 131.5, 132.7, 136.2, 138.8 (Ar), 144.7 (C-6), 149.7 (C-4), 162.8 (C-2); Anal. Calcd for: $C_{18}H_{14}ClNO$ C, 73.10; H, 4.77; N, 4.74. Found: C, 73.06; H, 4.80; N, 4.79%.

6-Methyl-4-(2-naphthyl)-3-phenylpyridin-2(1*H*)-one (7h). The reaction mixture was stirred for 18 h. Yield 65%, light crystals, mp 260 °C (decomp.); IR (KBr): ν, cm^{-1} 3440, 2775 (NH), 1624 (N–C=O). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.25 (3H, s, 6-CH_3), 6.17 (1H, s, H-5), 7.03–7.13 (6H, m, H Ar), 7.43–7.48 (2H, m, H Ar), 7.65 (1H, s, H-1′ Ar), 7.72–7.81 (3H, m, H Ar), 11.74 (1H, br.s, NH). ^{13}C NMR (100 MHz, DMSO-d_6) : δ 18.9 (6-CH_3), 107.4 (C-5), 126.7 (C-3), 126.7, 126.8, 127.2, 127.5, 127.7, 127.9, 128.1, 128.4, 131.6, 132.4, 133.4, 133.0, 136.5, 137.6 (Ar), 144.5 (C-6), 150.8 (C-4), 162.9 (C-2); Anal. Calcd for: $C_{22}H_{17}NO$ C, 84.86; H, 5.50; N, 4.50. Found: C, 84.87; H, 5.52; N, 4.43%.

6-Methyl-4-(1-naphthyl)-3-phenylpyridin-2(1*H*)-one (7i). The reaction mixture was stirred for 18 h. Yield 60%, light crystals, mp 235 °C (decomp.). IR (KBr): ν, cm^{-1} 3429, 2768 (NH), 1628 (N–C=O). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.22 (3H, s, 6-CH_3), 6.05 (1H, s, H-5), 6.97–7.720 (12H, m, H Ar), 11.82 (1H, br.s, NH). ^{13}C NMR (100 MHz, DMSO-d_6): δ 18.9 (6-CH_3), 107.2 (C-5), 126.7 (C-3), 126.1, 127.7, 128.4, 129.1, 131.6, 136.5 140.0 (Ar), 144.4 (C-6), 150.9 (C-4), 162.9 (C-2); Anal. Calcd for: $C_{22}H_{17}NO$ C, 84.86; H, 5.50; N, 4.50. Found: C, 84.90; H, 5.55; N, 4.58%.

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