Reactions of 2-aryl-4-arylidene-4*H*-oxazol-5-ones with 3-amino-1,2,4-triazole, 5-aminotetrazole, and 2-aminobenzimidazole

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The reactions of 3-amino-1,2,4-triazole, 5-aminotetrazole, and 2-aminobenzimidazole with 2-aryl-4-arylidene-4*H*-oxazol-5-ones (azlactones) were studied. The electron-releasing properties of the azole ring were demonstrated to influence the reaction pathway of azlactones with aminoazoles. The structures of the resulting compounds were established by ^{1}H and ^{13}C NMR spectroscopy using spin-spin decoupling and the nuclear Overhauser effect.

Key words: aminoazoles, 2-aryl-4-arylidene-4*H*-oxazol-5-ones, *N*-[2-aryl-1-(azol-5-ylcarbamoyl)vinyl]aroylamides, *N*-(7-aryl-5-oxo-4,5,6,7-tetrahydroazolo[1,5-*a*]pyrimidin-6-yl)aroylamides, cyclocondensation, NMR spectroscopy.

Arylideneoxazolones attract interest primarily as intermediates, which combine the properties of acid anhydrides and unsaturated carbonyl compounds and can react with substances containing labile hydrogen atoms, for example, with water,[1] alcohols,[2—4] amines,[5—7] and hydrogen halides.[8,9] However, data on their reactions with 1,3-dinucleophiles are lacking.

As part of continuing studies of the reactions of aminoazoles with α,β-unsaturated carbonyl compounds, we examined the reactions of 2-aryl-4-arylidene-4*H*-oxazol-5-ones **1—7** (Scheme 1) with 3-amino-1,2,4-triazole (**8**), 5-aminotetrazole (**9**), and 2-aminobenzimidazole (**24**).

The starting azlactones **1—7** were prepared by the reactions of *N*-aroyl derivatives of hippuric acid with aromatic aldehydes.[5,10]

We found that heating of azlactones **1—6** with amines **8** and **9** in DMF led to the oxazolone ring opening giving rise to *N*-[2-aryl-1-(1,2,4-triazol-5-ylcarbamoyl)vinyl]aroylamides **10—14** and *N*-[2-aryl-1-(tetrazol-5-ylcarbamoyl)vinyl]aroylamides **15—20**, respectively. Under analogous conditions, the reactions of arylideneoxazolones **1—4**, **6**, and **7** with 2-aminobenzimidazole **24** did not stop at acylation and afforded 1,2,3,4-tetrahydropyrimido[1,2-*a*]benzimidazol-2-ones **26—31**. *N*-[1-(Benzimidazol-2-ylcarbamoyl)-2-(2-methoxyphenyl)vinyl]benzamide **25** was isolated by performing the reaction of azlactone **3** with amine **24** at room temperature. Heating of **25** in DMF gave imidazopyrimidine **28**.

Compounds **10—20** remained unchanged upon refluxing both in pure DMF and in the presence of piperidine or triethylamine. Heterocyclization of triazole derivatives **10**, **13**, and **14** into the corresponding *N*-(5-oxo-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-*a*]pyrimidin-6-yl)benzamides **21—23** was performed by heating their solutions in DMF in the presence of acetic acid. Under these conditions, acylaminotetrazoles **15—20** did not undergo cyclization.

Compounds **21—23** were also prepared by the direct reactions of azlactones **1**, **4**, and **5**, respectively, with 3-amino-1,2,4-triazole (**8**) in DMF in the presence of catalytic amounts of acetic acid. The addition of acetic acid to the reaction mixtures of 5-aminotetrazole (**9**) with azlactones **1—6** has no effect on condensation, and the reactions also afforded aroylamides **15—20** as the only products.

The difference in the reactivity of aminoazoles **8**, **9**, and **24** with respect to arylideneoxazolones can be attributable to the fact that the triazole and tetrazole rings possess much stronger electron-withdrawing properties than the benzimidazole fragment. This leads to a decrease in the electron density on the reaction centers of aminoazoles **8** and **9** and, correspondingly, to a decrease in their nucleophilicity.

The compounds were identified by ^{1}H and ^{13}C NMR spectroscopy. The ^{1}H NMR spectra of compounds **10—20** show singlets of the ethylene proton at δ 7.6—7.8 and singlets of the CH groups of the triazole ring (for compounds **10—14**) at δ 7.3. In addition, the NMR spectra

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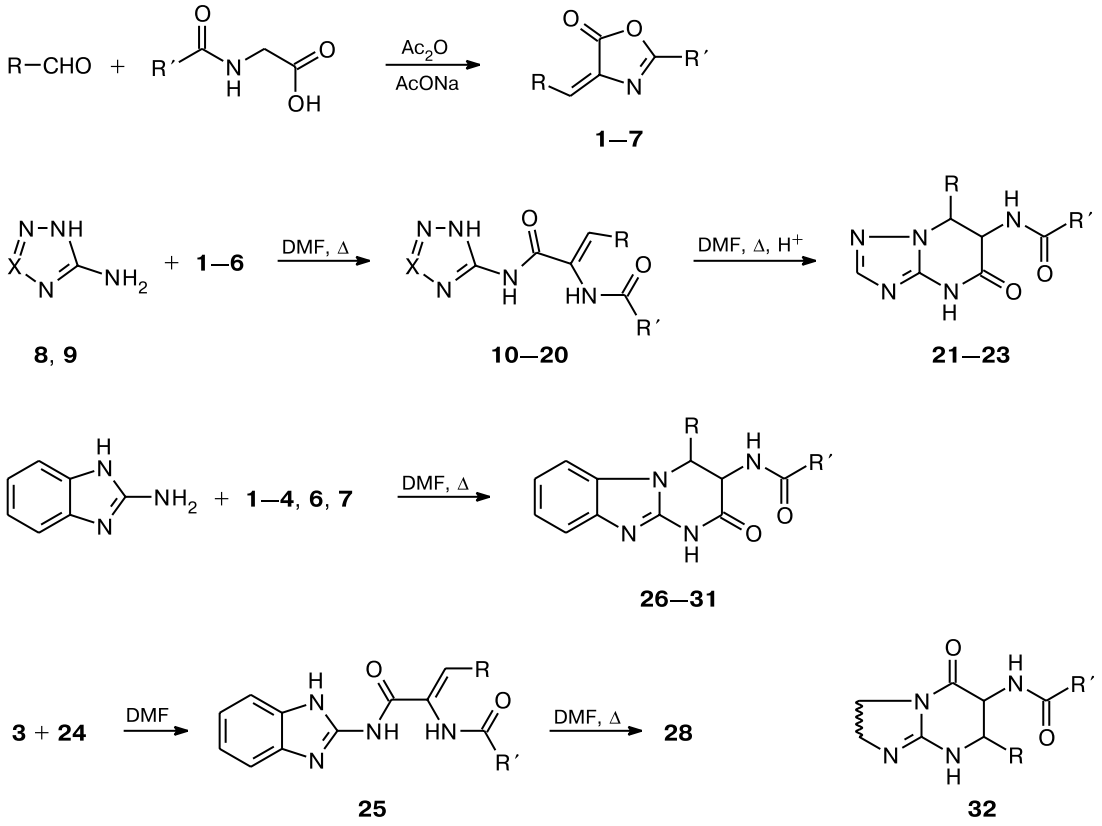
Scheme 1



8, **10—14:** X = CH
9, **15—20:** X = N

Compound	R	R′
1, **10**, **15**, **21**, **26**	Ph	Ph
2, **11**, **16**, **27**	3,4-$(MeO)_2C_6H_3$	Ph
3, **12**, **17**, **25**, **28**	2-$MeOC_6H_4$	Ph
4, **13**, **18**, **22**, **29**	2,4,5-$(MeO)_3C_6H_2$	Ph
5, **14**, **19**, **23**	4-$NO_2C_6H_4$	Ph
6, **20**, **30**	2-$MeOC_6H_4$	4-$MeOC_6H_4$
7, **31**	4-$MeOC_6H_4$	2-MeC_6H_4

have signals for the protons of three NH groups at δ 10.2, 11.8, and 13.5, multiplets for the aromatic protons, and signals of other functional groups. The assignment of the signals of the amino groups was made based on analysis of the experimental data on the deuterium exchange.

The ^{13}C NMR spectrum of compound **16** shows signals for all C nuclei present in the molecule (see the Experimental section).

In addition to the signals of the terminal substituents and aromatic protons, the ^1H NMR spectra of compounds **21—31** have signals for the methine protons and two amino groups. The signal of the C(3)H group (δ_H 5.7—5.8) appears as a doublet of doublets, and the signals of C(4)H (δ_H 5.5—6.5) and the exocyclic amino group (δ_H 7.5—9) appear as doublets. It should be noted that this set of signals in the ^1H NMR spectra corresponds both to structures **21—31** and compounds **32**, which could be generated in the case of the opposite direction of the reaction.

The structures of compounds **21—31** were unambiguously established using spin-spin decoupling and the nuclear Overhauser effect. It was found that suppression of the doublet of the exocyclic amino group or the doublet of C(4)H leads to a change in the multiplicity of the signal for C(3)H. In turn, suppression of the doublet of doublets of C(3)H changes the multiplicities of the signals of both the amino group and C(4)H. The nuclear Overhauser effect measurements demonstrated that C(3)H, C(4)H, and the proton of the exocyclic secondary amino group are in spatial proximity to each other. Additional irradiation at the resonance frequency of C(3)H

causes an increase in the integral intensities of the signals of C(4)H and NH, whereas double resonance at the frequency of the signal of the endocyclic amino group does not change the integral intensities of the signals for other protons.

The ^{13}C NMR spectrum of compound **27** is also consistent with the proposed structure.

The above-mentioned spectroscopic data provide evidence that the reactions of arylideneoxazolones **1—7** with 3-amino-1,2,4-triazole (**8**) and 2-aminobenzimidazole (**24**) afford compounds **21—31**, whereas an alternative pathway giving rise to imidazopyrimidines **32** does not take place.

To summarize, the reaction of aminoazoles with 2-aryl-4-arylidene-4*H*-oxazol-5-ones provides a convenient approach to the synthesis of both *N*-[2-aryl-1-(azolylcarbamoyl)vinyl]aroylamides and *N*-(7-aryl-5-oxo-4,5,6,7-tetrahydroazolo[1,5-*a*]pyrimidin-6-yl)aroylamides or *N*-(4-aryl-2-oxo-1,2,3,4-tetrahydropyrimido[1,2-*a*]benzimidazol-3-yl)aroylamides.

Experimental

The ^1H and ^{13}C NMR spectra were recorded on a Varian Mercury VX-200 spectrometer (200 MHz for ^1H and 50 MHz for ^{13}C) in DMSO-d_6 with Me_4Si as the internal standard. The deuterium exchange was carried out with the use of methanol-d_4. The purity of the compounds was checked by TLC on Silufol UV-254 plates using chloroform, ethyl acetate, or their mixtures as the solvents. The melting points were measured on a Kofler hot-stage apparatus.

2-Aryl-4-arylidene-4*H*-oxazol-5-ones **1—7** were synthesized according to a known procedure.[5,10] The melting points and spectroscopic characteristics of compounds **1—7** are identical to the data published in the literature.[11—14]

***N*-[2-(2-Methoxyphenyl)-1-(1,2,4-triazol-3-ylcarbamoyl)vinyl]benzamide (12).** A mixture of 3-amino-1,2,4-triazole (**8**) (0.84 g, 0.01 mol) and azlactone **3** (3.10 g, 0.01 mol) was heated until a homogeneous mixture formed. Then three drops of DMF were added and the mixture was heated until a precipitate formed (~5 min). After cooling of the reaction mixture, acetone (5 mL) was added. The precipitate was filtered off and washed with hot acetone. The yield was 57% (2.25 g), m.p. 257—258 °C. Found (%): N, 19.29. $C_{19}H_{17}N_5O_3$. Calculated (%): N, 19.27. ^1H NMR, δ: 3.43 (s, 3 H, OMe); 7.38 (s, 1 H, N=CH—N); 7.73 (s, 1 H, C=CH); 7.35—8.29 (m, 9 H, Ar); 10.20, 11.76, and 13.43 (all s, 1 H each, ArCONH, CONH, N—NH).

Compounds **10**, **11**, **13**, and **14** were synthesized analogously.

***N*-[2-Phenyl-1-(1,2,4-triazol-3-ylcarbamoyl)vinyl]benzamide (10).** The yield was 63%, m.p. 212—213 °C. Found (%): N, 21.05. $C_{18}H_{15}N_5O_2$. Calculated (%): N, 21.01. ^1H NMR, δ: 7.30 (s, 1 H, N=CH—N); 7.65 (s, 1 H, C=CH); 7.21—8.32 (m, 10 H, Ar); 10.18, 11.69, and 13.42 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[2-(3,4-Dimethoxyphenyl)-1-(1,2,4-triazol-3-ylcarbamoyl)vinyl]benzamide (11).** The yield was 61%, m.p. 255—256 °C. Found (%): N, 17.85. $C_{20}H_{19}N_5O_4$. Calculated (%): N, 17.80. ^1H NMR, δ: 3.48 and 3.51 (both s, 3 H each, OMe); 7.35 (s, 1 H, N=CH—N); 7.69 (s, 1 H, C=CH); 7.38—8.27 (m, 8 H, Ar); 10.25, 11.78, and 13.47 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[1-(1,2,4-Triazol-3-ylcarbamoyl)-2-(2,4,5-trimethoxyphenyl)vinyl]benzamide (13).** The yield was 58%, m.p. 215—217 °C. Found (%): N, 16.51. $C_{21}H_{21}N_5O_5$. Calculated (%): N, 16.54. ^1H NMR, δ: 3.48, 3.49, and 3.51 (all s, 3 H each, OMe); 7.33 (s, 1 H, N=CH—N); 7.71 (s, 1 H, C=CH); 6.92—7.92 (m, 7 H, Ar); 10.22, 11.82, and 13.45 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[2-(4-Nitrophenyl)-1-(1,2,4-triazol-3-ylcarbamoyl)vinyl]benzamide (14).** The yield was 67%, m.p. 221—222 °C. Found (%): N, 22.19. $C_{18}H_{14}N_6O_4$. Calculated (%): N, 22.21. ^1H NMR, δ: 7.41 (s, 1 H, N=CH—N); 7.80 (s, 1 H, C=CH); 7.36—8.30 (m, 9 H, Ar); 10.22, 11.75, and 13.44 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[2-Phenyl-1-(tetrazol-5-ylcarbamoyl)vinyl]benzamide (15).** A mixture 5-aminotetrazole (**9**) (0.85 g, 0.01 mol) and azlactone **1** (0.01 mol) was heated until a homogenous mixture formed. Then three drops of DMF were added, and the mixture was heated until a precipitate formed (~5 min). After cooling of the reaction mixture, acetone (5 mL) was added. The precipitate was filtered off and washed with hot acetone. The yield was 54% (0.18 g), m.p. 243—245 °C. Found (%): N, 25.19. $C_{17}H_{14}N_6O_2$. Calculated (%): N, 25.14. ^1H NMR, δ: 7.70 (s, 1 H, C=CH); 6.85—8.17 (m, 10 H, Ar); 10.01, 11.95, and 15.41 (all s, 1 H each, ArCONH, CONH, N—NH).

Compounds **16—20** were prepared analogously.

***N*-[2-(3,4-Dimethoxyphenyl)-1-(tetrazol-5-ylcarbamoyl)vinyl]benzamide (16).** The yield was 78%, m.p. 259—260 °C. Found (%): N, 25.19. $C_{17}H_{14}N_6O_2$. Calculated (%): N, 25.14. ^1H NMR, δ: 3.46 and 3.48 (both s, 3 H each, OMe); 7.69 (s, 1 H, C=CH); 6.95—8.15 (m, 8 H, Ar); 10.10, 12.08, and 15.15 (all s, 1 H each, ArCONH, CONH, N—NH). ^{13}C NMR, δ: 55.0, 55.5 (OMe); 111.6 (C̲H=C); 124.2 (CH=C̲); 112.8, 126.0, 127.7, 128.1 131.5 (*o*-CH_{Ar}, *m*-CH_{Ar}); 133.3 (*p*-CH_{Ar}); 164.5, 166.0 (C=O); 125.8, 132.5 (*ipso*-C_{Ar}), 148.3, 150.1, 151.5 (quaternary C).

***N*-[2-(2-Methoxyphenyl)-1-(tetrazol-5-ylcarbamoyl)vinyl]benzamide (17).** The yield was 61%, m.p. 261—263 °C. Found (%): N, 23.10. $C_{18}H_{16}N_6O_3$. Calculated (%): N, 23.07. ^1H NMR, δ: 3.41 (s, 3 H, OMe); 7.65 (s, 1 H, C=CH); 6.78—8.06 (m, 9 H, Ar); 9.95, 12.03, and 15.35 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[1-(Tetrazol-5-ylcarbamoyl)-2-(2,4,5-trimethoxyphenyl)vinyl]benzamide (18).** The yield was 63%, m.p. 208—210 °C. Found (%): N, 25.19. $C_{20}H_{20}N_6O_5$. Calculated (%): N, 25.14. ^1H NMR, δ: 3.50, 3.51, and 3.56 (all s, 3 H each, OMe); 7.61 (s, 1 H, C=CH); 6.81—7.88 (m, 7 H, Ar); 10.05, 11.98, and 15.50 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[2-(4-Nitrophenyl)-1-(tetrazol-5-ylcarbamoyl)vinyl]benzamide (19).** The yield was 68%, m.p. 254—255 °C. Found (%): N, 25.89. $C_{17}H_{13}N_7O_4$. Calculated (%): N, 25.85. ^1H NMR, δ: 7.13 (s, 1 H, C=CH); 6.87—8.09 (m, 9 H, Ar); 10.03, 12.01, and 15.46 (all s, 1 H each, ArCONH, CONH, N—NH).

***N*-[2-(2-Methoxyphenyl)-1-(tetrazol-5-ylcarbamoyl)vinyl]-4-methoxybenzamide (20).** The yield was 65%, m.p. 244—246 °C. Found (%): N, 21.32. $C_{19}H_{18}N_6O_4$. Calculated (%): N, 21.31. ^{1}H NMR, δ: 3.53 and 3.65 (both s, 3 H each, OMe); 7.01 (s, 1 H, C=CH); 6.91—8.10 (m, 8 H, Ar); 10.06, 11.97, and 15.55 (all s, 1 H each, ArCONH, CONH, N—NH).

N*-[7-(4-Nitrophenyl)-5-oxo-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-*a*]pyrimidin-6-yl]benzamide (23).** ***A. Glacial acetic acid (two drops) was added to a mixture of azlactone **5** (0.30 g, 0.01 mol) and 3-amino-1,2,4-triazole (**8**) (0.84 g, 0.01 mol) in DMF (0.1 mL), and the mixture was heated for 10 min. The precipitate that formed was filtered off and washed with propan-2-ol. The yield was 34% (0.13 g), m.p. 268—270 °C.

B. Glacial acetic acid (two drops) was added to a solution of *N*-[2-(4-nitrophenyl)-1-(1,2,4-triazol-3-ylcarbamoyl)vinyl]benzamide (**12**) (0.38 g, 0.01 mol) in DMF (0.2 mL) and the reaction mixture was heated for 15 min. The precipitate that formed was filtered off and washed with propan-2-ol. The yield was 40% (0.15 g), m.p. 268—270 °C.

Found (%): N, 22.19. $C_{18}H_{14}N_6O_4$. Calculated (%): N, 22.21. ^{1}H NMR, δ: 5.62 (dd, 1 H, C(3)H, J = 7.4 Hz, J = 6.8 Hz); 5.89 (d, 1 H, C(4)H, J = 7.4 Hz); 7.80 (s, 1 H, N=CH—N); 6.47—7.79 (m, 9 H, Ar); 8.21 (d, 1 H, NH, J = 6.8 Hz); 12.04 (br.s, 1 H, N(1)H).

Compounds **21** and **22** were synthesized according to the methods ***A*** and ***B***, respectively.

***N*-(5-Oxo-7-phenyl-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-*a*]pyrimidin-6-yl)benzamide (21).** The yield was 42%, m.p. 263—265 °C. Found (%): N, 21.04. $C_{18}H_{15}N_5O_2$. Calculated (%): N, 21.01. ^{1}H NMR, δ: 5.66 (dd, 1 H, C(3)H, J = 7.6 Hz, J = 7.6 Hz); 5.91 (d, 1 H, C(4)H, J = 7.6 Hz); 7.78 (s, 1 H, N=CH—N); 6.50—7.75 (m, 10 H, Ar); 8.18 (d, 1 H, NH, J = 7.6 Hz); 12.02 (br.s, 1 H, N(1)H).

***N*-[5-Oxo-7-(2,4,5-trimethoxyphenyl)-4,5,6,7-tetrahydro-1,2,4-triazolo[1,5-*a*]pyrimidin-6-yl)benzamide (22).** The yield was 38%, m.p. 265—267 °C. Found (%): N, 16.57. $C_{21}H_{21}N_5O_5$. Calculated (%): N, 16.54. ^{1}H NMR, δ: 3.66, 3.67, and 3.75 (all s, 3 H each, OMe); 5.68 (dd, 1 H, C(3)H, J = 7.2 Hz, J = 6.4 Hz); 5.95 (d, 1 H, C(4)H, J = 7.2 Hz); 8.01 (s, 1 H, N=CH—N); 6.68—8.15 (m, 7 H, Ar); 9.61 (d, 1 H, NH, J = 6.4 Hz); 11.55 (br.s, 1 H, N(1)H).

***N*-[1-(Benzimidazol-2-ylcarbamoyl)-2-(2-methoxyphenyl)vinyl]benzamide (25).** A mixture of azlactone **3** (0.28 g, 0.01 mol) and 2-aminobenzimidazole **22** (0.13 g, 0.01 mol) in DMF (0.3 mL) was kept at room temperature for 12 h. The precipitate that formed was filtered off and washed with a small amount of acetone. The yield was 56%, m.p. 279—280 °C. Found (%): N, 13.56. $C_{24}H_{20}N_4O_3$. Calculated (%): N, 13.58. ^{1}H NMR, δ: 3.85 (s, 3 H, OMe); 7.49 (s, 1 H, C=CH); 6.80—8.10 (m, 13 H, Ar); 9.96, 12.12, and 15.62 (all s, 1 H each, ArCONH, CONH, N=C—NH).

N*-[4-(2-Methoxyphenyl)-2-oxo-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]benzamide (28).** ***A. A mixture of 2-aminobenzimidazole (**22**) (0.13 g, 0.01 mol) and azlactone **3** (0.28 g, 0.01 mol) was heated until a melt formed and then DMF (0.1 mL) was added. The reaction mixture was heated for 7 min and then cooled, after which acetone (5 mL) was added and the reaction mixture was kept at room temperature for one day. The precipitate that formed was filtered off and washed with acetone. The yield was 84%, m.p. 257—258 °C.

B. A solution of *N*-[1-(benzimidazol-2-ylcarbamoyl)-2-(2-methoxyphenyl)vinyl]benzamide (**23**) (0.41 g, 0.01 mol) in DMF (0.3 mL) was heated for 15 min and then cooled, after which acetone (5 mL) was added and the reaction mixture was kept at room temperature for one day. The precipitate that formed was filtered off and washed with acetone. The yield was 75%, m.p. 257—258 °C.

Found (%): N, 13.60. $C_{24}H_{20}N_4O_3$. Calculated (%): N, 13.58. ^{1}H NMR, δ: 3.57 (s, 3 H, OMe); 5.69 (dd, 1 H, C(3)H, J = 8.2 Hz, J = 7.2 Hz); 6.26 (d, 1 H, C(4)H, J = 8.2 Hz); 6.78—7.65 (m, 13 H, Ar); 8.02 (d, 1 H, NH, J = 7.2 Hz); 11.84 (br.s, 1 H, N(1)H).

Compounds **26**, **27**, and **29—31** were synthesized according to the procedure ***A***.

***N*-[2-Oxo-4-phenyl-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]benzamide (26).** The yield was 58%, m.p. 223—225 °C. Found (%): N, 14.63. $C_{23}H_{18}N_4O_2$. Calculated (%): N, 14.65. ^{1}H NMR, δ: 5.70 (dd, 1 H, C(3)H, J = 7.5 Hz, J = 7.5 Hz); 6.12 (d, 1 H, C(4)H, J = 7.5 Hz); 6.89—7.90 (m, 14 H, Ar); 8.20 (d, 1 H, NH, J = 7.5 Hz); 11.75 (br.s, 1 H, N(1)H).

***N*-[4-(3,4-Dimethoxyphenyl)-2-oxo-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]benzamide (27).** The yield was 84%, m.p. 257—258 °C. Found (%): N, 12.64. $C_{25}H_{22}N_4O_4$. Calculated (%): N, 12.66. ^{1}H NMR, δ: 3.45 and 3.65 (both s, 3 H each, OMe); 5.67 (dd, 1 H, C(3)H, J = 7.6 Hz, J = 7.6 Hz); 6.10 (d, 1 H, C(4)H, J = 7.6 Hz); 6.40—7.81 (m, 12 H, Ar); 8.22 (d, 1 H, NH, J = 7.6 Hz); 11.96 (br.s, 1 H, N(1)H). ^{13}C NMR, δ: 55.7, 56.0 (OMe); 53.2 (C(4)); 55.2 (C(3)); 110.2, 112.1, 112.5, 118.0, 119.0, 121.9, 122.6, 128.3, 129.0 (*o*-CH_{Ar}, *m*-CH_{Ar}); 132.8 (*p*-CH_{Ar}); 167.1, 167.3 (C=O); 127.9, 134.1 (*ipso*-C_{Ar}); 132.8, 142.1, 147.9, 149.1, 149.5 (quaternary C).

***N*-[2-Oxo-4-(2,4,5-trimethoxyphenyl)-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]benzamide (29).** The yield was 56%, m.p. 216—218 °C. Found (%): N, 11.90. $C_{26}H_{24}N_4O_5$. Calculated (%): N, 11.86. ^{1}H NMR, δ: 3.37, 3.83, and 3.84 (all s, 3 H each, OMe); 5.88 (dd, 1 H, C(3)H, J = 7.4 Hz, J = 7.2 Hz); 6.05 (d, 1 H, C(4)H, J = 7.4 Hz); 6.67—8.81 (m, 11 H, Ar); 9.83 (d, 1 H, NH, J = 7.2 Hz); 11.84 (br.s, 1 H, N(1)H).

***N*-[4-(2-Methoxyphenyl)-2-oxo-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]-4-methoxybenzamide (30).** The yield was 60%, m.p. 220—221 °C. Found (%): N, 12.65. $C_{25}H_{22}N_4O_4$. Calculated (%): N, 12.66. ^{1}H NMR, δ: 3.52 and 3.71 (both s, 3 H each, OMe); 5.65 (dd, 1 H, C(3)H, J = 7.6 Hz, J = 7.6 Hz); 6.22 (d, 1 H, C(4)H, J = 7.6 Hz); 6.70—7.74 (m, 12 H, Ar); 7.76 (d, 1 H, NH, J = 7.6 Hz); 11.69 (br.s, 1 H, N(1)H).

***N*-[4-(4-Methoxyphenyl)-2-oxo-1,2,3,4-tetrahydrobenz[6,7]imidazo[1,2-*a*]pyrimidin-3-yl]-2-methylbenzamide (31).** The yield was 62%, m.p. 219—220 °C. Found (%): N, 13.16. $C_{25}H_{22}N_4O_3$. Calculated (%): N, 13.14. ^{1}H NMR, δ: 2.25 (s, 3 H, Me); 3.71 (s, 3 H, OMe); 5.10 (dd, 1 H, C(3)H, J = 7.3 Hz, J = 7.6 Hz); 5.58 (d, 1 H, C(4)H, J = 7.6 Hz); 6.13—7.65 (m, 12 H, Ar); 8.90 (d, 1 H, NH, J = 7.3 Hz); 11.65 (br.s, 1 H, N(1)H).

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