# The Chemical and Structural Properties of 2-Aminobenzylamine Derivatives

Kamal M. El-Shaieb[a] and Peter G. Jones[b]

[a] Chemistry Department, Faculty of Science, Minia University, El-Minia, A. R. Egypt

[b] Institute of Inorganic and Analytical Chemistry, Hagenring 30, Technische Universität Braunschweig, D-38106 Braunschweig, Germany

Reprint requests to Dr. Kamal M. El-Shaieb. Fax: +2-086-2342601.
E-mail: kmelshaieb@yahoo.com

*Z. Naturforsch.* **2013**, *68b*, 913 – 923 / DOI: 10.5560/ZNB.2013-3107
Received April 8, 2013

We report a conventional and simple method to synthesize quinazoline and benzodiazepine derivatives by treatment of 2-aminobenzylamine (**1**) with several $\pi$-electron-deficient compounds. Quinazoline derivatives were obtained by treatment of **1** with either tetracyanoethylene or 7,7,8,8-tetracyanoquinodimethane. Benzodiazepine derivatives were formed when **1** was reacted with either 2,3-dichloro-1,4-naphthoquinone or 2,3-dichloro-5,6-dicyano-1,4-benzoquinone. Fused quinazoline and benzoazepine derivatives were formed by dry heating of **1** with either naphthalic anhydride, *cis*-1,2,3,6-tetrahydrophthalic anhydride, tetrachlorophthalic anhydride and/or diphenic anhydride, respectively. The NMR spectra and the mass spectrometric data as well as elemental analyses of all new products are consistent with the proposed structures. The structures of the quinazoline derivatives **26** and **28** were further confirmed by single-crystal X-ray structure determination. Plausible mechanisms for the formation of several products are discussed.

*Key words:* 2-Aminobenzylamine, $\pi$ Acceptors, Anhydrides, Quinazolines, Diazepines

## Introduction

Nitrogen-containing heterocycles are an important class of compounds in medicinal chemistry [1]. There has been considerable interest in the development of reliable and simple preparative methods for the production of quinazolines [2], because they and their ring-fused derivatives display a broad spectrum of biological activities [3 – 5], like *e. g.* antitubercular, analgesic, anti-inflammatory, and anti-bacterial, analgesic, and many other properties. These examples clearly demonstrate the remarkable potential of quinazoline derivatives as a source of useful pharmacophores for new drug development. Substituted quinazolines have been synthesized by a number of methods involving several substrates such as 2-amino-*N*′-aryl-benzamidines [6 – 8] and 2-aminobenzylamine [9, 10]. A further important class of heterocycles is furnished by benzodiazepines [11]. Benzodiazepine derivatives are widely distributed in nature, and they represent a class of heterocycles which possesses a wide range of biological applications. Many of them are for example widely used as antileukaemic, antiplatelet, anticonvulsant, and neuroleptic agents [12]. Some heterocycles containing benzodiazepine moieties were also found to have anti-inflammatory, antiviral, anti-HIV-1, antimicrobial, and antitumour activities [13]. Apart from their biological importance, benzodiazepines are valuable synthons for the preparation of fused ring compounds, such as triazolo, thiazolo, imidazo and pyrimidobenzo-diazepines [14]. Although many methods for synthesizing benzodiazepine ring systems have been reported, they continue to receive much attention [11]. We found that reaction of either 2,3-dichloro-1,4-naphthoquinone (DCHNQ) or 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ) with 2-amino-*N*′-arylbenzamidines led to the formation of benzodiazepine derivatives *via* charge-transfer (CT) complexes [15]. The formation of CT complexes has long been recognized as an important phenomenon in many chemical processes [16], and this approach led us to synthesize several heterocyclic systems that could not be obtained by usual methods [17].

## Results and Discussion

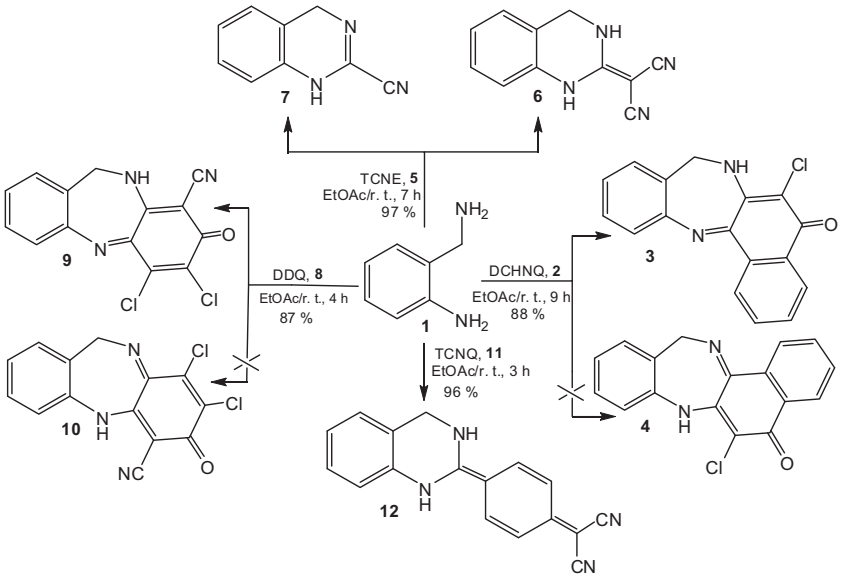
On continuing our studies in electron donor-acceptor interactions (EDA) as well as developing conventional approaches for the synthesis of *N*-heterocycles, we reported very recently highly efficient approaches for the synthesis of quinazoline [6–8] and benzodiazepine [12] derivatives. The study was initiated by investigating the reaction of 2-aminobenzylamine (**1**) with the acceptors **2**, **5**, **8**, and **11**. We have found that treatment of **1** with TCNE **5** and/or TCNQ **11** in anhydrous ethyl acetate leads to the formation of deep-red or green complexes. The color gradually disappears to give rise to the formation of the quinazoline derivatives **6**, **7** and **12**, respectively, in excellent yield (Scheme 1). The assigned molecular structures of the new compounds were based on spectroscopic analysis including IR, NMR, MS, and elemental analysis data.

The product **6** displayed in its IR spectrum strong absorption maxima at $\nu = 3272$ and $3218\,cm^{-1}$ indicating the presence of two different NH groups; the CN groups absorb at $\nu = 2207$ and $2175\,cm^{-1}$.

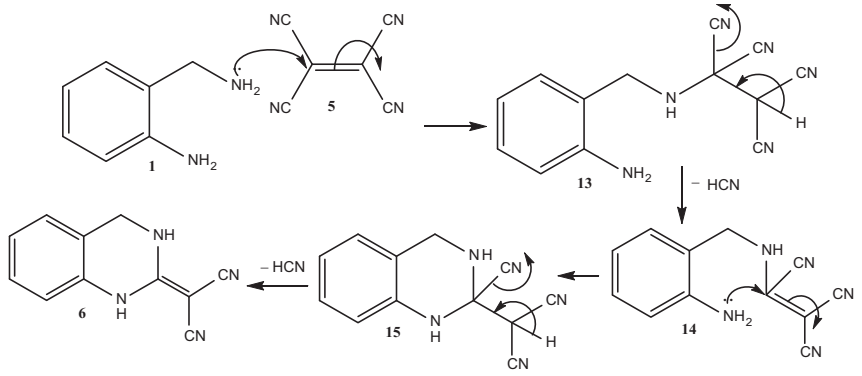
Furthermore, the IR spectrum reveals no absorption bands characteristic of $NH_2$ groups. The $^1H$ NMR spectrum of **6** exhibits, besides the signals from the aromatic protons, two sharp singlets at $\delta = 10.26$ and 8.32 ppm for the two different N*H* groups, and a doublet at $\delta = 4.38$ ppm is assigned to the aliphatic C$H_2$ protons with a coupling constant of $J = 1.00$ Hz. The $^{13}C$ NMR spectrum showed signals from eleven distinct carbon atoms, two of them at $\delta = 119.16$ and 117.62 ppm characteristic of cyano groups. The signal at $\delta = 41.12$ ppm was assigned to the carbon atom of the *C*$H_2$ group. Both MS and elemental analysis confirm the molecular formula of **6** as $C_{11}H_8N_4$.

The $^1H$ NMR of compound **7** reveals, in addition to the three multiplets assignable to the N*H* group and aromatic protons, a singlet at $\delta = 4.73$ ppm for the two aliphatic methylene protons. The IR spectrum shows bands at $\nu = 3278$ and $2180\,cm^{-1}$ characteristic for NH and CN, respectively. Formation of the quinazolines **6** and **7** is suggested to proceed according to the mechanism outlined in Schemes 2 and 3. In this case the reaction sequence would start by the formation of the adducts **13**–**15**. Losing a molecule of either HCN or malononitrile furnishes the quinazolines **6** and **7**, respectively.

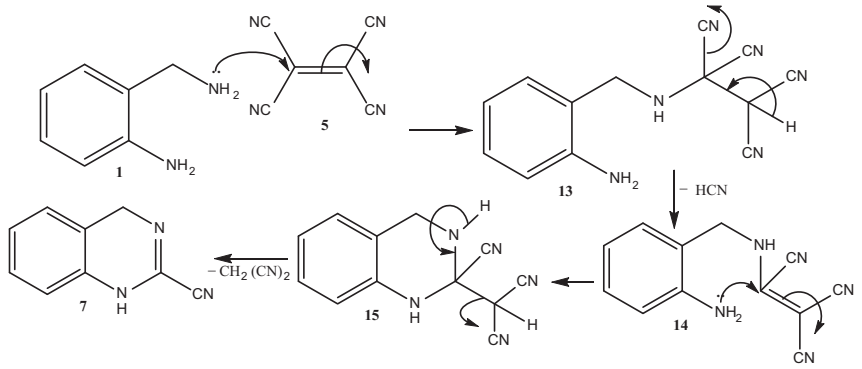
Addition of the electron donor **1** to the electron acceptor **11** in anhydrous ethyl acetate at room temperature led to an EDA complex with a deep-red color, which gradually disappeared to give a single new reaction product **12** (TLC analysis). The mass spectrum and elemental analysis suggest the molecular formula of **12** as $C_{17}H_{12}N_4$. The IR spectrum of **12** shows sharp absorptions characteristic of differ-




Scheme 1. Reaction of 2-aminobenzylamine (**1**) with $\pi$-electron-deficient compounds.



Scheme 2. Mechanism for the formation of quinazoline derivative **6**.



Scheme 3. Mechanism for the formation of quinazoline derivative **7**.

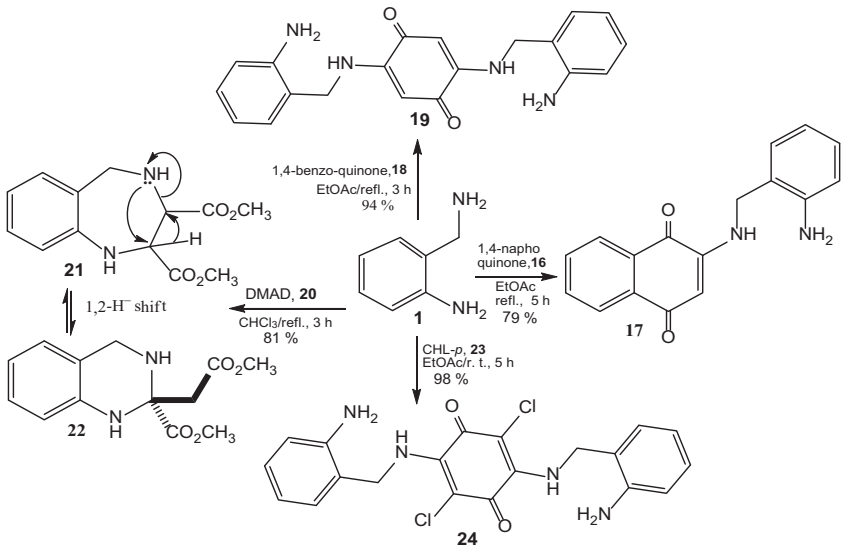
ent cyano groups at $\nu = 2185$ and 2133 $cm^{-1}$, and of the NH groups at $\nu = 3271$, 3245 $cm^{-1}$. Its $^1$H NMR spectrum reveals, apart from the aromatic protons, two broad singlets at $\delta = 11.51$ and 10.12 ppm for the two different N*H* groups; the cyclohexadiene protons appear as two doublets at $\delta = 7.60$ and 6.90 ppm with similar coupling constants of $J = 8.81$ Hz, and the two aliphatic methylene protons as a singlet at $\delta =$ 4.76 ppm.

Benzonaphthodiazepine derivative **3** rather than **4** is formed on stirring compound **1** with DCHNQ **2** in ethyl acetate at room temperature. One indication of this is the coupling between the methylene protons (doublet at $\delta = 4.33$ ppm with $J = 5.25$ Hz) and the adjacent $sp^3$ N*H* proton (triplet at $\delta = 8.38$ ppm with $J = 5.10$ Hz); in **4** both of the N*H* and methylene protons should appear as two different singlets. The IR spectrum of **3** displayed absorptions at $\nu = 3284$ (NH) and 1630 $cm^{-1}$ (CO). In its $^{13}$C NMR spectrum, the *C*=O and *C*=N carbon atoms appear at $\delta = 174.59$ and 151.14 ppm, respectively, while the methylene carbon appears at $\delta = 44.80$ ppm. The brutto formula $C_{17}H_{11}ClN_2O$ of **3** was confirmed by the mass spectrum, which exhibited the molecular ion at $m/z = 294$ (100 %).

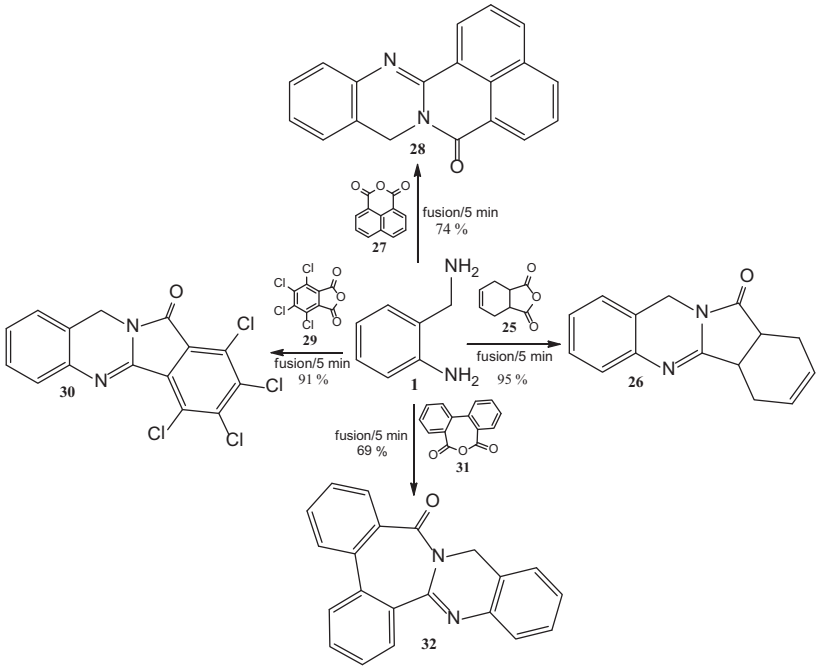
Treatment of **1** with DDQ **7** in ethyl acetate led to the dibenzodiazepine **8** rather than to its isomer **9**, the evidence for which is related to the case of **3** and **4** (see above). The IR spectrum of compound **8** showed three strong absorptions at $\nu = 3296$, 2209 and 1663 $cm^{-1}$, indicating the presence of NH, CN and CO groups, respectively. In the $^1$H NMR spectrum the N*H* group appeared as a triplet at $\delta = 9.85$ ppm with $J = 5.16$ Hz, whereas the methylene protons form a doublet at $\delta =$ 4.37 ppm with $J = 5.27$ Hz. The $^{13}$C NMR spectrum of **8** showed fourteen distinct resonances, of which those at $\delta = 172.15$ and 45.24 ppm are assigned to the carbonyl and the methylene carbon atoms, respectively. The molecular formula of dibenzodiazepine **8** is supported by elemental analysis and the mass spectrum with the expected molecular ion peak as the base peak.

Heating of **1** with NQ **16** and/or BQ **18** in anhydrous ethyl acetate produced **17** and **19**, respectively (Scheme 4). The $^1$H NMR spectrum showed all the expected signals; in addition to the aromatic signals a triplet at $\delta \approx 8.14$ ppm for the N*H* protons, and a singlet of two protons at $\delta \approx 5.00$ ppm for the N$H_2$ protons were observed. In addition, a characteristic doublet at $\delta \approx 4.20$ ppm with a coupling constant $J \approx 6.00$ Hz for the methylene protons was noted. The IR spectra showed bands at $\nu \approx 3420$, 3390, 3220 cm$^{-1}$ for the $NH_2$ and NH groups, whereas the carbonyl groups absorbed at $\nu \approx 1670$ and 1629 cm$^{-1}$. Furthermore, the structure assigned for the products **17** and **19** were fully supported by their mass spectra, which showed their molecular formulae $C_{17}H_{14}N_2O_2$ and $C_{20}H_{20}N_4O_2$, respectively.

Heating of **1** with dimethyl acetylenedicarboxylate (DMAD, **20**) in anhydrous chloroform provides quinazoline derivative **22**, which might be in equilibrium with its diazepine isomer **21** (Scheme 4). The results obtained from elemental microanalysis and the IR, $^1$H NMR, $^{13}$C NMR and mass spectroscopic data are in agreement with the assigned structure. The IR spectrum showed two strong absorptions at $\nu = 3362$ and 1729 cm$^{-1}$, indicating the presence of NH and CO groups, respectively. However, the $^1$H NMR spectrum shows two broad resonances at $\delta = 4.88$ and 2.60 ppm for the two exchangeable protons of the two N*H* groups. Moreover, there are two singlets at $\delta =$ 3.76 and 3.70 ppm for the methyl ester protons and a doublet at $\delta = 3.94$ ppm for the -NH–C$H_2$- protons with $J = 4.35$ Hz. The $^1$H NMR spectrum of **21** displayed two doublets at $\delta = 3.05$ and 2.85 ppm with similar coupling constants $J = 15.63$ Hz referred to the AB spin system, along with characteristic resonances for the four aromatic protons. The presence of the AB spin system in the $^1$H NMR spectrum indicates the formation of the product **21**. As the result of the expected instability of the benzodiazepine derivative **21**, due to its angle strain, one can expect its rearrangement into the more stable quinazoline derivative **22**. Isomerization between **21** and **22** has been noticed and confirmed by measuring the NMR spectrum of the sample after 1 h. The data reveals the presence of a quaternary carbon atom resonating in the $^{13}$C NMR spectrum at $\delta = 70.63$ ppm characteristic for the NH–*C*–NH carbon atom, in addition to the presence of a signal at $\delta = 43.24$ ppm for the methylene carbon of the *C*$H_2CO_2CH_3$ group. The two carbonyl groups of the product have been recorded in the $^{13}$C NMR spectrum at $\delta = 172.31$ and 170.22 ppm. Furthermore, the methylene carbon atom of the *C*$H_2$NH group resonates at $\delta = 42.48$ ppm, and the methyl ester carbon atoms at $\delta = 52.92$ and 51.93 ppm. The mass spectrum of this sample gives the molecular ion peak at $m/z = 264$ which is in accordance with the molecular mass (264.11) of compound **22**.




Scheme 4. Reactions of **1** with CHL-*p*, NQ, BQ and DMAD.




Scheme 5. Reactions of compound **1** with selected anhydrides.

The substituted quinone **24** was obtained in excellent yield by the reaction of 2-amino-benzylamine (**1**) with CHL-*p* **23** in absolute ethyl acetate at room temperature (Scheme 4). The deep-green solution initally formed rapidly turned to yellowish green while a precipitate was formed. The structure of **24** was confirmed by elemental analysis and spectral data. Three IR bands at $\nu = 3421, 3349, 3326\,\mathrm{cm}^{-1}$ are assignable to $NH_2$ and NH groups, while the carbonyl group appeared at $\nu = 1638\,\mathrm{cm}^{-1}$. In the $^1$H NMR spectrum the N*H* and N$H_2$ protons appeared at $\delta = 8.18$ and 5.11 ppm, respectively; six multiplets in the region at $\delta = 7.17 - 6.50$ ppm are assigned to aromatic protons, and a singlet at $\delta = 4.80$ ppm to the methylene protons. In the $^{13}$C NMR spectrum signals of most of the carbon atoms of $CH_2$, *C*H, *C* =O and the aromatic system could be assigned and were in agreement with the proposed structure (see spectral data in the Experimental Section). Both mass spectrum and elemental analysis confirm the molecular formula of the product **24** as $C_{20}H_{18}Cl_2N_4O_2$.

Recently, we have reported that diamino compounds can be employed as key starting materials for the synthesis of diverse nitrogen bridgehead compounds [6–8, 15]. This prompted us to investigate the reactivity of **1** towards electrophilic reagents such as naphthalic anhydride, tetrahydrophthalic anhydride, diphenic anhydride, and tetrachlorophthalic anhydride (Scheme 5). The conditions for the reaction between the reactant **1** and the anhydrides **25**, **27**, **29**, and **31** were investigated in a number of trials, and the optimized conditions were identified by varying the reagent concentrations, the reaction time and/or temperature. We found that the optimum conditions for this reaction involve mixing both reaction partners in equimolar amounts and heating them at 200 °C for 5 min. This reaction sequence is conveniently performed in one-pot procedures without isolation of the intermediates. Previous studies on refluxing **1** with anhydride **27** in absolute EtOH afforded a mixture of two products in low yields [18]. Thus when a mixture of **1** and anhydride **25** were dry heated at 200 °C for 5 minutes, the quinazoline derivative **26** was formed as colorless crystals in excellent yield (Scheme 5).

The structure of product **26** was corroborated by its elemental analysis and IR, $^1$H and $^{13}$C NMR spectroscopic data. The mass spectrum of **26** displayed the molecular ion ($M^+$) at $m/z = 238$ as the base peak. The $^1$H NMR spectrum of **26** exhibited four differ-

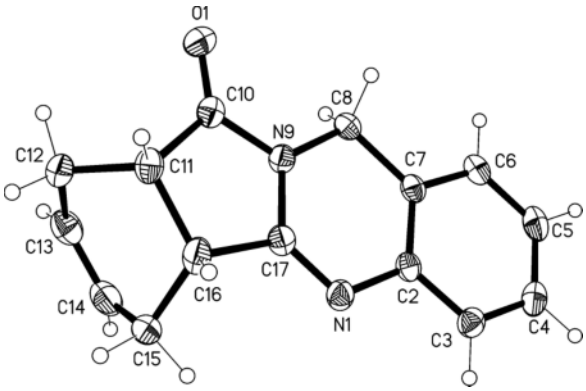



Fig. 1. The molecular structure of **26** in the solid state with the crystallographic numbering scheme adopted.

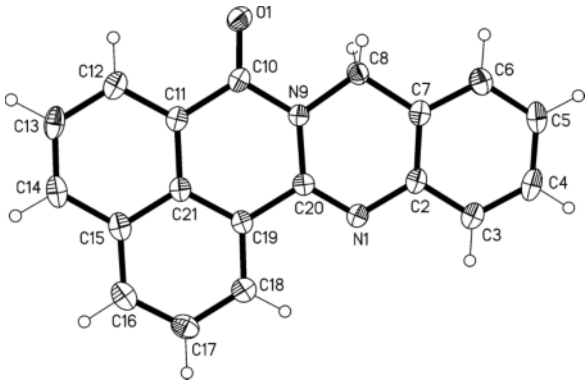



Fig. 2. The molecular structure of **28** in the solid state with the crystallographic numbering scheme adopted.

ent multiplets at $\delta = 7.24 - 7.00$ ppm for the aromatic protons, along with five different characteristic multiplets for the six protons of the cyclohexene moiety at $\delta = 3.38 - 2.24$ ppm. Furthermore, the methylene proton resonated as a sharp singlet at $\delta = 4.79$ ppm. The proton-decoupled $^{13}$C NMR spectrum of **26** showed 14 distinct resonance lines in agreement with the proposed structure. Unambiguous evidence for the proposed structure of **26** was finally obtained by single-crystal X-ray-diffraction analysis as shown in Fig. 1.

Similarly, heating of **1** with naphthalic anhydride (NA, **27**) in the absence of solvents for 5 minutes at 200 °C afforded the quinazoline derivative **28** as orange crystals in excellent yield as shown in Scheme 5. The structure of the quinazoline derivative **28** was established by conventional spectroscopic methods as well as elemental analysis. It exhibits two strong IR absorption bands at 1694 and 1623 cm$^{-1}$ for the CO and C=N groups, respectively. The same carbon atoms resonated in the $^{13}$C NMR at $\delta = 171.16$ and 160.73 ppm, respectively. The $^{1}$H NMR spectrum of **28** shows characteristic signals in the aromatic region at $\delta = 8.51 - 6.26$ ppm. A singlet at $\delta = 5.06$ ppm was assigned to the methylene protons. The carbon atom of this methylene group resonated in the $^{13}$C NMR at $\delta = 40.33$ ppm. The mass spectrum exhibits the molecular ion peak at $m/z = 284$. Moreover, the structure of **28** was confirmed by an X-ray diffraction study (Fig. 2).

The quinazoline derivative **30** was formed when one equivalent of **1** was fused with one equivalent of the anhydride **29**; its structure was fully supported by elemental analysis and its mass spectrum, which confirmed the molecular formula $C_{15}H_6Cl_4N_2O$ (369.92).

The reactivity of **1** towards the anhydride **31** was also studied; as illustrated in Scheme 5 the reaction furnishes the dibenzoazepinoquinazoline **32**. Its structure was assigned on the basis of the NMR data and was supported by IR data. The $^{1}$H NMR spectrum of the quinazoline derivative **32** revealed six characteristic multiplets at $\delta = 7.68 - 6.55$ ppm assigned to the twelve aromatic protons. Furthermore, a singlet was present at $\delta = 4.14$ ppm due to the methylene protons. The methylene carbon signal of **32** appeared in the $^{13}$C NMR spectrum at $\delta = 40.78$ ppm, and the carbonyl carbon atom resonated at $\delta = 175.11$ ppm. The molecular formula of compound **32** is supported by elemental analysis and a mass spectrum that gave the expected molecular ion peak and fragmentation patterns (see Experimental Section).

## Conclusion

Herein, we have reported a facile and efficient synthesis of substituted quinazoline and benzodiazepine derivatives by using a one-pot reaction of diverse $\pi$ acceptors and anhydrides with 2-aminobenzylamine. In all cases the reaction of two components proceeded rapidly to afford the corresponding desired products in excellent yields. The reaction conditions are mild, the work-up is simple, and the steps are automation-friendly. The structures of two selected examples (**26** and **28**) were elucidated by single X-ray diffraction analysis. The mechanism of formation of the obtained products **5**, **6**, **36**, and **37** has been rationalized.

Table 1. Crystallographic data for compounds **26** and **28**.

| Compound | **26** | **28** |
|---|---|---|
| Formula | $C_{15}H_{14}N_2O$ | $C_{15}H_{14}N_2O$ |
| $M_r$ | 238.28 | 278.33 |
| Habit | colorless tablet | colorless tablet |
| Crystal size, mm$^3$ | $0.35\times0.3\times0.2$ | $0.3\times0.15\times0.1$ |
| Crystal system | monoclinic | monoclinic |
| Space group | $P2_1/n$ | $P2_1/n$ |
| Temperature, °C | −133 | −173 |
| $a$, Å | 6.1268(2) | 5.2041(6) |
| $b$, Å | 10.5576(3) | 17.6877(16) |
| $c$, Å | 18.0859(5) | 14.1232(16) |
| $\beta$, deg | 90.357(3) | 90.03(2) |
| $V$, Å$^3$ | 1169.85 | 1300.0 |
| $Z$ | 4 | 4 |
| $D_{calcd.}$, Mg m$^{-3}$ | 1.35 | 1.45 |
| $\mu(MoK_\alpha)$, mm$^{-1}$ | 0.1 | 0.1 |
| $F(000)$, e | 504 | 592 |
| $2\theta_{max}$, deg | 60 | 56.6 |
| Refl. measured / unique / $R_{int}$ | 41055 / 3412 / 0.028 | 30329 / 3215 / 0.044 |
| Ref. parameters | 163 | 200 |
| $R[F>4\sigma(F)]$ | 0.041 | 0.041 |
| $wR(F^2$, all refls.) | 0.107 | 0.106 |
| $S$ | 1.02 | 1.06 |
| $\Delta\rho_{max}$, e Å$^{-3}$ | 0.35 | 0.31 |

## Experimental Section

*General.* All reagents were purchased from Acros and Aldrich companies and were used without further purification. The melting points were measured in capillary tubes without corrections using a Büchi 530 melting point apparatus. The NMR spectra were recorded on a Bruker AM 400 MHz spectrometer with TMS as internal standard; the coupling constants are given in Hz. The mass spectra (EI) were performed using a Finnigan MAT 8430 spectrometer. IR spectra were run from KBr discs using a Bruker Tensor 27 instrument. Chromatography columns were prepared from Merck silica gel 230 – 240 mesh.

*Synthesis of 6-chloro-7,8-dihydro-5H-benzo[e]naphtho[2,1-b][1,4]diazepin-5-one (**3**)*

To 227 mg (1.0 mmol) of **2** in anhydrous ethyl acetate (10 mL) at room temperature was added dropwise a solution of **1** (122 mg, 1.0 mmol) in anhydrous ethyl acetate (10 mL). The color of the reaction mixture changed to deep red. The mixture was stirred at room temperature for 9 h. A red precipitate separated. The resulting precipitate was filtered, washed with ethyl acetate, dried and recrystallized from EtOH to give the diazepine derivative **3**.

Red powder (yield: 259 mg, 88 %), m. p. > 240 °C. – IR (film): $\nu$ = 3284 (NH), 3066, 2923, 2853 (CH), 1630 (CO), 1590 (C=N), 1539 (C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, [D$_6$]DMSO): $\delta$ = 4.33 (d, 2H, CH$_2$, $J$ = 5.25 Hz), 7.41 – 7.56 (m, 4H), 7.96 – 7.98 (dd, 1H, $J$ = 1.72, 7.54 Hz), 7.66 – 7.73 (m, 2H), 8.12 – 8.14 (dd, 1H, $J$ = 1.70, 7.66 Hz), 8.38 (t, 1H, NH, $J$ = 5.10 Hz). – $^{13}$CNMR (100 MHz, [D$_6$]DMSO): $\delta$ = 44.80 (CH$_2$), 103.41 (C), 106.04 (C), 124.61 (CH), 125.74 (CH), 127.14 (CH), 129.06 (CH), 129.11 (C), 129.15 (CH), 130.85 (CH), 131.55 (CH), 132.03 (CH), 135.11 (C), 145.44 (C), 149.41 (C), 151.14 (C=N), 174.59 (CO). – MS (EI, 70 eV): $m/z$(%) = 297 (5), 296 (30), 294 (100) [M]$^+$, 293 (16), 268 (5), 260 (10), 259 (16) [M − HCl]$^+$, 251 (4), 233 (2), 232 (8), 231 (9), 229 (18), 217 (4), 203 (10), 190 (2), 176 (5), 165 (4), 156 (4), 134 (2), 128 (4), 115 (10), 102 (16), 89 (12), 77 (4). – $C_{17}H_{11}ClN_2O$ (294.06): calcd. C 69.28, H 3.76, N 9.50; found C 69.17, H 3.75, N 9.43.

*Reaction of 2-aminobenzylamine (1) with 1,1,2,2-tetracyanoethylene (**5**)*

To 128 mg (1.0 mmol) of **5** in anhydrous ethyl acetate (20 mL), 122 mg (1.0 mmol) of **1** in 10 mL of anhydrous ethyl acetate was added with stirring within 1/2 h. The mixture was stirred for 7 h at room temperature, during which time a crystalline colorless product separated. The resulting solid material was filtered, and the precipitate was washed with ethyl acetate, dried and recrystallized from ethanol to give the quinazoline derivative **6**. The filtrate was concentrated to dryness, and the residue was subjected to column chromatography using $CH_2Cl_2$ as an eluent to give a red zone which was removed and extracted to give the quinazoline derivative **7**.

*2-(3,4-Dihydroquinazolin-2(1H)-ylidene)malononitrile (**6**)*

Colorless powder (yield: 147 mg, 75 %), m. p. > 240 °C. – IR (film): $\nu$ = 3272, 3218 (NH), 3168, 3059, 2944, 2870 (CH), 2207, 2175 (CN), 1596, 1552 (C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, [D$_6$]DMSO): $\delta$ = 4.38 (d, 2H, CH$_2$, $J$ = 1.00 Hz), 7.04 – 7.09 (m, 1H), 7.15 (d, 1H, $J$ = 7.41 Hz), 7.19 – 7.25 (m, 2H), 8.32 (s, 1H, NH), 10.26 (s, 1H, NH). – $^{13}$CNMR (100 MHz, [D$_6$]DMSO): $\delta$ = 41.12 (CH$_2$), 115.95 (CH), 117.62 (C), 119.16 (C), 124.08 (CH), 125.91 (CH), 128.16 (CH), 133.71 (C), 158.05 (C). – MS (EI, 70 eV): $m/z$(%) = 197 (7) [M + 1]$^+$, 196 (75) [M]$^+$, 195 (100) [M − 1]$^+$, 168 (22) [M − HCN]$^+$, 141 (10), 129 (7), 116 (4), 104 (6), 97 (4), 89 (2), 77 (8). – $C_{11}H_8N_4$ (196.07): calcd. C 67.34, H 4.11, N 28.55; found C 67.25, H 4.13, N 28.49.

*1,4-Dihydroquinazolin-2-carbonitrile (**7**)*

Deep-red powder (yield: 35 mg, 22 %), m. p. 121 – 123 °C. – IR (film): $\nu$ = 3278 (NH), 3135, 3059, 2976, 2864 (CH), 2180 (CN), 1695, 1619, 1575 (C=N, C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ = 4.73 (s, 2H, CH$_2$), 6.91 (d, 1H, $J$ = 7.41 Hz), 7.09 – 7.13 (m,

2H), 7.20 – 7.24 (m, 1H), 7.31 – 7.37 (m, 1H). – $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta = 42.02$ ($CH_2$), 108.83 (C), 126.01 (CH), 126.41 (CH), 128.12 (C), 128.55 (CH), 128.59 (CH), 129.68 (CH), 147.49 (C). – MS (EI, 70 eV): $m/z(\%) = 158$ (5) $[M+1]^+$, 157 (40) $[M]^+$, 156 (100) $[M-1]^+$, 129 (22) $[M-HCN]^+$, 102 (6), 77 (10). – $C_9H_7N_3$ (157.06): calcd. C 68.78, H 4.49, N 26.74; found C 68.61, H 4.44, N 26.60.

*Synthesis of 6,7-dichloro-8-oxo-10,11-dihydro-8H-dibenzo[b,e][1,4]diazepine-9-carbonitrile (**9**)*

To 227 mg (1.0 mmol) of **8** in anhydrous ethyl acetate (15 mL) was added dropwise a solution of compound **1** (122 mg, 1.0 mmol) in anhydrous ethyl acetate (10 mL) at room temperature. The color of the reaction mixture changed to deep red. The mixture was stirred at room temperature for 4 h. A red precipitate separated, which was filtered and washed with ethyl acetate, dried and recrystallized from EtOH to give the diazepine derivative **9**.

Deep-red powder (yield: 264 mg, 87 %), m. p. 201 – 204 °C. – IR (film): $\nu =$ 3296 (NH), 3015, 2921, 2875 (CH), 2209 (CN), 1663 (CO), 1559 (C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, $[D_6]$DMSO): $\delta = 4.37$ (d, 2H, $J = 5.27$ Hz, $CH_2$), 7.34 – 7.39 (m, 1H), 7.45 – 7.49 (m, 1H), 7.59 (d, 2H, $J = 2.96$ Hz), 9.85 (t, 1H, NH, $J = 5.16$ Hz). – $^{13}$CNMR (100 MHz, $[D_6]$DMSO): $\delta = 45.24$ ($CH_2$), 79.75 (C), 114.79 (C), 127.59 (CH), 128.94 (CH), 129.57 (CH), 130.22 (C), 134.57 (C), 131.46 (CH), 140.11 (C), 144.51 (C), 147.35 (C), 155.60 (C=N), 172.15 (CO). – MS (EI, 70 eV): $m/z(\%) = 307$ (30) $[M+4]^+$, 305 (75) $[M+2]^+$, 303 (100) $[M]^+$, 289 (35), 277/275 (35/55) $[M-HCN]^+$, 269 (20), 255 (10), 240 (35) $[M-(HCN+HCl)]^+$, 213 (10), 204 (10) $[M-(HCN+2HCl)]^+$, 179 (10), 177 (12), 163 (4), 153 (6), 137 (8), 104 (6), 77 (8). – $C_{14}H_7Cl_2N_3O$ (303.00): calcd. C 55.29, H 2.32, N 13.82; found C 55.18, H 2.34, N 13.70.

*Synthesis of 2-(4-(3,4-dihydroquinazolin-2(1H)-ylidene)cyclohexa-2,5-dien-1-ylidene)malononitrile (**12**)*

To 204 mg (1.0 mmol) of **11** in anhydrous ethyl acetate (30 mL) was added dropwise a solution of **1** (122 mg, 1.0 mmol) in anhydrous ethyl acetate (10 mL) at room temperature. The color of the reaction mixture changed to green. The mixture was stirred at room temperature for 3 h. A yellowish-green precipitate separated, which was filtered and washed with ethyl acetate, dried and recrystallized from DMF to give the quinazoline derivative **12**.

Yellowish-green powder (yield: 261 mg, 96 %), m. p. > 240 °C. – IR (film): $\nu =$ 3271, 3245 (NH), 3155, 3025, 2937, 2871 (CH), 2185, 2133 (CN), 1598, 1566 (C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, $[D_6]$DMSO): $\delta = 4.76$ (s, 2H, $CH_2$), 6.90 (d, 2H, $J = 8.81$ Hz), 7.20 – 7.27 (m, 3H), 7.32 – 7.38 (m, 1H), 7.60 (d, 2H, $J = 8.81$ Hz), 10.12 (br s, 1H, NH), 11.51 (br s, 1H, NH). – $^{13}$CNMR (100 MHz, $[D_6]$DMSO): $\delta = 41.62$ ($CH_2$), 112.14 (C), 116.84 (CH), 117.38 (2CH), 118.55 (C), 122.93 (C), 126.07 (CH), 126.46 (CH), 128.63 (CH), 128.85 (2CH), 132.61 (C), 149.92 (C), 156.48 (C). – MS (EI, 70 eV): $m/z(\%) = 273$ (15) $[M+1]^+$, 272 (100) $[M]^+$, 243 (30) $[M-HCN]^+$, 215 (10), 205 (6), 178 (10), 164 (2), 151 (5), 141 (10), 129 (12), 122 (10), 114 (5), 104 (25), 95 (3), 85 (3), 76 (20). – $C_{17}H_{12}N_4$ (272.11): calcd. C 74.98, H 4.44, N 20.58; found C 74.87, H 4.41, N 20.47.

*Reactions of 2-aminobenzylamine (**1**) with 1,4-naphthoquinone (**16**) and/or 1,4-benzoquinone (**18**)*

In a typical experiment, reactant **1** (122 mg, 1.0 mmol) was added dropwise to a well-stirred solution of 1,4-naphthoquinone (**16**) (158 mg, 1 mmol) and/or 1,4-benzoquinone (**18**) (108 mg, 1.0 mmol) in anhydrous ethylacetate (15 mL) at room temperature. The color of the parent quinone disappeared, and the solution turned brown. The solution was heated under reflux conditions for 3 – 5 h. The progress of the reaction was monitored *via* TLC. The volume of the solution was reduced to half under reduced pressure. A brown precipitate appeared which was filtered, dried at room temperature and recrystallized from EtOH to obtain the desired products **17** and **19**.

*2-((2-Aminobenzyl)amino)naphthalene-1,4-dione (**17**)*

Deep-brown powder (yield: 220 mg, 79 %), m. p. 132 – 134 °C. – IR (film): $\nu =$ 3425, 3393, 3223 ($NH_2$,NH), 3074, 3028, 2959, 2827 (CH), 1671, 1601 (CO), 1568 (C=C) cm$^{-1}$. – $^1$H NMR (400 MHz, $[D_6]$DMSO): $\delta = 4.22$ (d, 2H, $CH_2$, $J = 6.03$ Hz), 5.08 (br s, 2H, $NH_2$), 6.50 (t, 1H, $J = 6.46$ Hz), 6.61 – 6.64 (dd, 1H, $J = 1.16$, 8.52 Hz), 7.22 (s, 1H), 6.94 – 6.98 (m, 2H), 7.63 (t, 1H, $J = 7.47$ Hz), 7.73 (t, 1H, $J = 7.51$ Hz), 8.04 (t, 2H, $J = 5.60$ Hz), 8.13 (t, 1H, NH, $J = 6.12$ Hz). – $^{13}$CNMR (100 MHz, $[D_6]$DMSO): $\delta = 41.74$ ($CH_2$), 115.67 (CH), 119.01 (C), 122.82 (C), 125.37 (C), 126.58 (CH), 127.02 (CH), 127.11 (CH), 128.00 (C), 128.46 (CH), 130.74 (CH), 131.66 (CH), 133.12 (CH), 142.16 (C), 151.10 (C), 172.54 (CO), 182.53 (CO). – MS (EI, 70 eV): $m/z(\%) = 280$ (4) $[M+2]^+$, 279 (16) $[M+1]^+$, 278 (54) $[M]^+$, 261 (6) $[M-NH_3]^+$, 185 (12), 172 (32), 156 (6), 121 (10), 106 (68), 93 (5), 76 (10). – $C_{17}H_{14}N_2O_2$ (278.11): calcd. C 73.37, H 5.07, N 10.07; found C 73.19, H 5.03, N 9.93.

*2,5-Bis((2-aminobenzyl)amino)cyclohexa-2,5-diene-1,4-dione (**19**)*

Brown powder (yield: 327 mg, 94 %), m. p. 219 – 222 °C. – IR (film): $\nu$ = 3429, 3403, 3352, 3329 ($NH_2$,NH), 3074, 3028, 2917, 2873 (CH), 1629 (CO), 1583 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, [$D_6$]DMSO): $\delta$ = 4.20 (d, 4H, $2CH_2$, $J$ = 6.24 Hz), 5.14 (s, 4H, $NH_2$), 5.19 (s, 2H), 6.50 (t, 2H, $J$ = 6.46 Hz), 6.61 – 6.64 (dd, 2H, $J$ = 1.16, 8.52 Hz), 6.94 – 6.98 (m, 4H), 8.15 (t, 2H, 2NH, $J$ = 6.20 Hz). – $^{13}$CNMR (100 MHz, [$D_6$]DMSO): $\delta$ = 42.46 ($2CH_2$), 93.06 (2CH), 114.94 (2CH), 115.74 (2CH), 119.18 (2C), 128.00 (2CH), 128.46 (2CH), 146.16 (2C), 151.10 (2C), 177.53 (2CO). – MS (EI, 70 eV): $m/z$(%) = 350 (2) $[M+2]^+$, 349 (10) $[M+1]^+$, 348 (40) $[M]^+$, 331 (6) $[M-NH_3]^+$, 314 (4), 295 (5), 282 (2), 242 (48), 225 (30), 198 (5), 181 (2), 169 (5), 121 (5), 106 (100), 93 (5), 77 (12). – $C_{20}H_{20}N_4O_2$ (348.16): calcd. C 68.95, H 5.79, N 16.08; found C 68.79, H 5.71, N 15.89.

*Synthesis of dimethyl 2,3,4,5-tetrahydro-1H-benzo[e][1,4]diazepine-2,3-dicarboxylate (**21**)/methyl 2-(2-methoxy-2-oxoethyl)-1,2,3,4-tetrahydroquinazoline-2-carboxylate (**22**)*

Into a 250 mL two-necked round bottom flask containing a solution of **20** (142 mg, 1.0 mmol) in absolute $CHCl_3$ (10 mL), a solution of **1** (122 mg, 1.0 mmol) in absolute $CHCl_3$ (10 mL) was added dropwise with stirring. The mixture was stirred at room temperature for 1 h, and at reflux for 3 h monitored by TLC. The solvent was evaporated under vacuum, and the residue was purified by dissolving in $CH_2Cl_2$ (5 mL) and then subjected to preparative plate chromatography (silica gel, diethyl ether). The obtained products **21** and/or **22** were detected by spectral analyses.

Colorless oil (yield: 214 mg, 81 %). – IR (film): $\nu$ = 3362 (NH), 3008, 2953, 2894 (CH), 1729 (CO), 1609, 1592 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 2.60 (br s, 1H, NH), 2.85 (d, 1H, $J$ = 15.63 Hz, -C*H*–$CO_2CH_3$), 3.05 (d, 1H, $J$ = 15.63 Hz, -C*H*–$CO_2CH_3$), 3.70 (s, 3H, $-CH_3$), 3.94 (d, 2H, $J$ = 4.35 Hz, $-CH_2$), 3.76 (s, 3H, $-CH_3$), 4.84 (br s, 1H, 1NH), 6.60 – 6.57 (dd, 1H, $J$ = 0.92, 7.97 Hz), 6.72 – 6.67 (ddd, 1H, $J$ = 1.14 Hz), 6.89 – 6.86 (dd, 1H, $J$ = 1.02, 7.50 Hz), 7.00 (t, 1H, $J$ = 7.00 Hz). – $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 42.48 ($CH_2$), 43.24 ($CH_2$), 51.93 ($CH_3$), 52.92 ($CH_3$), 70.63 (C), 115.18 (CH), 118.47 (CH), 119.93 (C), 125.83 (CH), 127.38 (CH), 141.10 (C), 170.22 (CO), 172.31 (CO). – MS (EI, 70 eV): $m/z$(%) = 265 (4) $[M+1]^+$, 264 (18) $[M]^+$, 232 (7), 206 (14), 205 (96), 199 (6), 191 (18), 190 (62), 173 (10), 157 (38), 145 (10), 130 (40), 129 (100), 106 (46), 77 (20).

*Synthesis of 2,5-bis((2-aminobenzyl)amino)-3,6-dichlorocyclohexa-2,5-diene-1,4-dione (**24**)*

A solution of compound **1** (122 mg, 1.0 mmol) in anhydrous ethyl acetate (10 mL) was added to a magnetically stirred solution of 2,3,5,6-tetrachloro-1,4-benzoquinone (**23**) (246 mg, 1.0 mmol) in anhydrous ethyl acetate (20 mL). The yellow color of the solution changed to deep green and then to yellowish green. The reaction mixture was stirred at room temperature for 5 h. After completion of the reaction (followed by TLC), the formed precipitate was collected by filtration, washed and recrystallized from EtOH to afford the product **24**.

Pale-brown powder (yield: 408 mg, 98 %), m. p. 164 – 167 °C. – IR (film): $\nu$ = 3421, 3349, 3326 ($NH_2$,NH), 3025, 2996, 2921, 2849 (CH), 1638 (CO), 1564 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, [$D_6$]DMSO): $\delta$ = 4.80 (s, 4H, $2CH_2$), 5.11 (br s, 4H, $2NH_2$), 6.50 – 6.59 (m, 2H), 6.64 – 6.69 (m, 1H), 6.70 – 6.72 (m, 1H), 6.86 – 6.90 (m, 1H), 6.95 – 6.99 (m, 1H), 7.03 – 7.07 (m, 1H), 7.15 – 7.17 (dd, 1H, $J$ = 1.20, 7.49 Hz), 8.18 (br s, 2H, 2 NH). – $^{13}$CNMR (100 MHz, [$D_6$]DMSO): $\delta$ = 39.49 ($CH_2$), 114.90 (C), 115.62 (CH), 116.05 (CH), 116.18 (CH), 117.28 (CH), 122.06 (C), 121.37 (C), 126.91 (C), 127.30 (CH), 127.87 (CH), 129.39 (CH), 130.52 (CH), 144.43 (C), 146.95 (C). – MS (EI, 70 eV): $m/z$(%) = 420/418/416 (2/4/6) [$M^{4+}$/$M^{2+}$/$M^+$], 380 (4) $[M-HCl]^+$, 362 (2) $[M-(HCl+H_2O)]^+$, 346 (2), 313/311/309 (10/16/12), 277/275 (5/8), 259 (7), 208 (6), 179 (3), 171 (4), 143 (3), 131 (4), 118 (7), 106 (100), 93 (5), 77 (12). – $C_{20}H_{18}Cl_2N_4O_2$ (416.08): calcd. C 57.57, H 4.35, N 13.43; found C 57.47, H 4.33, N 13.35.

*General procedures for the reaction of **1** with anhydrides **25**, **27**, **29**, and **31***

In a fusion tube provided with a condenser, a mixture of compound **1** (122 mg, 1.0 mmol) and 1.0 mmol of the anhydride (either **25**, **27**, **29** or **31**) was heated in an oil bath at 200 °C for 5 min. Then the mixture was cooled to room temperature and poured into 20 mL $Et_2O$. The obtained solids were collected and recystallized from EtOH or DMF to give the products **26**, **28**, **30** and **32**.

*1,4,4a,12a-Tetrahydroisoindolo[1,2-b]quinazolin-12(10H)-one (**26**)*

Colorless crystals (yield: 226 mg, 95 %), m. p. 154 – 156 °C. – IR (film): $\nu$ = 3039, 2944, 2898 (CH), 1730 (CO), 1641 (C=N), 1599, 1578 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 2.24 – 2.30 (m, 1H), 2.34 – 2.40 (m, 1H), 2.61 – 2.69 (m, 2H), 3.01 – 3.06 (m, 1H), 3.32 – 3.38 (m, 1H), 4.79 (s, 2H, $CH_2$), 5.90 – 5.93 (m, 2H), 7.00 – 7.03 (m, 1H), 7.10 – 7.14 (m, 1H), 7.21 – 7.24

(m, 2H). – $^{13}$CNMR (100 MHz, $CDCl_3$): $\delta$ = 23.25 (C $H_2$), 25.27 (C $H_2$), 37.36 (CH), 38.66 (CH), 41.55 ($CH_2$), 126.09 (CH), 126.40 (CH), 126.42 (CH), 127.73 (CH), 128.06 (CH), 128.75 (CH), 140.39 (C), 160.73 (C=N), 178.36 (CO). – MS (EI, 70 eV): $m/z(\%)$ = 240 (3) $[M+2]^+$, 239 (12) $[M+1]^+$, 238 (100) $[M]^+$, 233 (2), 223 (10), 209 (25), 195 (8), 183 (18), 168 (6), 155 (5), 144 (6), 132 (12), 116 (3), 104 (12), 97 (4), 91 (6), 76 (14). – $C_{15}H_{14}N_2O$ (238.11): calcd. C 75.61, H 5.92, N 11.76; found C 75.50, H 5.90, N 11.67.

*Benzo[4,5]isoquinolino[1,2-b]quinazolin-7(9H)-one (**28**)*

Orange crystals (yield: 210 mg, 74 %), m. p. 211 – 215 °C. – IR (film): $\nu$ = 3064, 2967 (CH), 1737, 1694 (CO), 1623 (C=N), 1588, 1560 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, $[D_6]$DMSO): $\delta$ = 5.06 (s, 2H, $CH_2$), 6.46 (t, 1H, $J$ = 7.60 Hz), 6.69 (d, 1H, $J$ = 7.47 Hz), 6.96 – 7.02 (m, 1H), 7.18 – 7.28 (m, 1H), 7.68 – 7.91 (m, 3H), 8.36 – 8.51 (m, 3H). – $^{13}$CNMR (100 MHz, $[D_6]$DMSO): $\delta$ = 40.33 ($CH_2$), 114.33 (CH), 115.98 (CH), 118.84 (C), 119.71 (C), 121.68 (C), 125.76 (CH), 126.60 (CH), 127.15 (CH), 127.21 (CH), 127.87 (CH), 128.28 (C), 130.95 (CH), 132.38 (CH), 134.45 (CH), 135.30 (C), 146.14 (C), 160.73 (C=N), 171.16 (CO). – MS (EI, 70 eV): $m/z(\%)$ = 286 (3) $[M+2]^+$, 285 (14) $[M+1]^+$, 284 (65) $[M]^+$, 283 (100), 255 (12), 227 (5), 198 (4), 180 (2), 152 (8), 142 (6), 127 (10), 114 (2), 99 (2), 76 (5). – $C_{19}H_{12}N_2O$ (284.09): calcd. C 80.27, H 4.25, N 9.85; found C 80.21, H 4.26, N 9.78.

*1,2,3,4-Tetrachloroisoindolo[1,2-b]quinazolin-12(10H)-one (**30**)*

Yellow powder (yield: 336 mg, 91 %), m. p. 159 – 161 °C. – IR (film): $\nu$ = 3054, 2916 (CH), 1722 (CO), 1633 (C=N), 1601 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, $[D_6]$DMSO): $\delta$ = 4.37 (s, 2H, $CH_2$), 6.27 – 6.29 (dd, 2H, $J$ = 1.18, 7.96 Hz), 7.58 – 7.60 (dd, 2H, $J$ = 1.57, 7.60 Hz). – $^{13}$CNMR (100 MHz, $[D_6]$DMSO): $\delta$ = 38.82 ($CH_2$), 116.39 (CH), 118.31 (CH), 119.75 (C), 126.83 (C), 127.92 (C), 128.67 (C), 128.98 (C), 129.21 (C), 129.50 (C), 130.25 (CH), 133.27 (CH), 139.07 (C), 146.31 (C), 163.54 (C). – MS (EI, 70 eV): $m/z(\%)$ = 374 (15), 372 (52), 370 (100), 368 (68) $[M]^+$, 336 (8), 308 (6), 299 (5), 271 (6), 236 (6), 209 (3), 186 (6), 172 (2), 153 (5), 135 (3), 119 (4), 102 (5), 76 (4). – $C_{15}H_6Cl_4N_2O$ (369.92): calcd. C 48.43, H 1.63, N 7.53; found C 48.29, H 1.60, N 7.42.

*Dibenzo[3,4:5,6]azepino[2,1-b]quinazolin-9(11H)-one (**32**)*

Colorless powder (yield: 214 mg, 69 %), m. p. 88 – 91 °C. – IR (film): $\nu$ = 3059, 2929, 2872 (CH), 1709 (CO), 1596 (C=N), 1540 (C=C) $cm^{-1}$. – $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ = 4.14 (s, 2H, $CH_2$), 6.50 – 6.55 (m, 3H), 6.57 – 6.85 (m, 1H), 6.99 – 7.18 (m, 3H), 7.28 – 7.37 (m, 3H), 7.46 – 7.51 (m, 1H), 7.62 – 7.68 (m, 1H). – $^{13}$CNMR (100 MHz, $CDCl_3$): $\delta$ = 40.78 ($CH_2$), 116.18 (CH), 118.42 (CH), 127.10 (CH), 127.71 (CH), 127.83 (CH), 128.96 (CH), 129.89 (CH), 130.00 (CH), 130.19 (CH), 130.67 (CH), 134.65 (C), 139.48 (C), 175.11 (CO). – MS (EI, 70 eV): $m/z(\%)$ = 312 (2) $[M+2]^+$, 311 (8) $[M+1]^+$, 310 (26) $[M]^+$, 309 (15) $[M-1]^+$, 282 (30), 281 (65), 254 (10), 223 (4), 206 (4), 197 (8), 181 (18), 152 (22), 144 (10), 121 (60), 106 (30), 76 (12). – $C_{21}H_{14}N_2O$ (310.11): calcd. C 81.27, H 4.55, N 9.03; found C 81.12, H 4.51, N 8.90.

*X-Ray structure determinations*

Numerical details are presented in Table 1. *Data collection and reduction*: Crystals were mounted in inert oil on glass fibers and transferred to the cold gas stream of an Oxford Diffraction Xcalibur E diffractometer. Measurements were performed using monochromated Mo$K_\alpha$ radiation ($\lambda$ = 0.71073 Å). Absorption corrections were applied for **28** using the multi-scan method. *Structure refinement*: The structures were refined anisotropically against $F^2$ (program SHELXL-97) [19]. Hydrogen atoms were included using a riding model. Compound **28** was refined as a non-merohedral twin (by 180° rotation about the *a* axis).

CCDC 922329 (**26**) and 922330 (**28**) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre *via* www.ccdc.cam.ac.uk/data_request/cif.

*Acknowledgement*

K. M. El-Shaieb gratefully acknowledges the financial support of the German Research foundation (DFG). Professor H. Hopf is thanked for his support and helpful discussions.

[1] N. P. Abida, R. Arpana, I. Mohd, *Intern. J. Pharm. & Biol. Arch.* **2011**, 1651 – 1657.
[2] B. Kaur, R. Kaur, *Arkivoc* **2007**, *xiii*, 315 – 323.
[3] J. Kunes, J. Bazant, M. Pour, K. Waisser, M. Slosarek, J. Janota, *IL Farmaco* **2000**, 725 – 729.
[4] P. M. S. Bedi, V. Kumar, M. P. Mahajan, *Bioorg. Med. Chem. Lett.* **2004**, 5211 – 5213.
[5] D. J. Brown, *The Chemistry of Heterocyclic Compounds*; J. Wiley & Sons: New York, **1996**.

[6] K. M. El-Shaieb, M. A. Ameen, F. F. Abdel-Latif, A. H. Mohamed, *Z. Naturforsch.* **2012**, *67b* 1144 – 1150.
[7] K. M. El-Shaieb, H. Hopf, P. G. Jones, *Z. Naturforsch.* **2009**, *64b*, 858 – 864.
[8] K. M. El-Shaieb, P. G. Jones, *Z. Naturforsch.* **2009**, *64b*, 945 – 951.
[9] A. J. L. Pombeiro, V. Yu. Kukushkin in *Comprehensive Coordination Chemistry II*, (Eds.: J. A. McCleverty, T. C. Meyer), Vol. 1, Elsevier, Amsterdam, **2004**, p. 631.
[10] B. Han, X.-L. Yang, C. Wang, Y.-W. Bai, T.-C. Pan, X. Chen, W. Yu, *J. Org. Chem.* **2012**, 1136 – 1142.
[11] N. K. Terret, D. Bojanic, J. R. Merson, P. T. Stephenson, *Bioorg Med Chem Lett.* **1992**, *2*, 1745 – 1750.
[12] R. Ramajayam, R. Giridhar, M. R. Yadav, R. Balaraman, H. Djaballah, D. Shum, C. Radu, *Eur. J. Med. Chem.* **2008**, *43*, 2004 – 2010.
[13] R. Kumar, Y. C. Joshi, *Arkivoc* **2007**, *xiii*, 142 – 149.
[14] R. Janciene, A. Vektariene, Z. Stumbreviciute, L. Kosychova, A. Klimavicius, B. D. Puodziunaite, *Heteroatom Chem.* **2004**, *15*, 363 – 368.
[15] F. F. Abdel-Latif, K. M. El-Shaieb, A. G. El-Deen, *Z. Naturforsch.* **2011**, *66b*, 965 – 971.
[16] K. M. El-Shaieb, M. A. Ameen, F. F. Abdel-Latif, A. H. Mohamed, *J. Chem. Res.* **2012**, *36*, 528 – 531.
[17] K. M. El-Shaieb, *J. Chin. Chem. Soc.* **2007**, *54*, 1353 – 1358.
[18] E. F. M. Stephenson, *J. Chem. Soc.* **1954**, 2354 – 2357.
[19] G. M. Sheldrick, *Acta Crystallogr.* **2008**, *A64*, 112 – 122.