Journal of Saudi Chemical Society (2014) **18**, 1022–1027

King Saud University

Journal of Saudi Chemical Society

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ORIGINAL ARTICLE

A simple procedure for synthesis of 3*H*-quinazolin-4-one hydrazones under mild conditions

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Received 10 October 2011; accepted 6 December 2011
Available online 16 December 2011

KEYWORDS

Hydrazones;
Condensation;
2-Hydrazino-3-phenyl-3*H*-quinazolin-4-ones;
Acetylation

Abstract Condensation of 6-bromo- and 6,8-dibromo-2-hydrazino-3-phenyl-3*H*-quinazolin-4-ones with D-sugars in the presence of a catalytic quantity of glacial acetic acid gave the corresponding hydrazones in good yields. Acetylation of hydrazones with acetic anhydride in anhydrous pyridine gave the corresponding acetyl hydrazones in high yields. Also, other hydrazones were synthesized from condensation of 2-hydrazino-3*H*-quinazolin-4-ones with aromatic aldehydes in the presence of a catalytic quantity of piperidine.

1. Introduction

Quinazoline derivatives exhibit a wide variety of pharmacological activities (Honda et al., 1979; Alafeefy, 2008; Al-Deeb and Alafeefy, 2008; Kadi, 2011). Therefore, methods for the synthesis and/or modification of this ring system are always of interest. They are important intermediates in the synthesis of a variety of valuable heterocyclic compounds (Shaban et al., 1991; El-Hiti, 2000; El-Hiti et al., 2011).

In view of the biological activities of both quinazolines and sugar moieties, we have reported convenient procedures for the synthesis of various quinazoline glycosides (Abdo et al., 1995; Abdel-Megeed et al., 1995, 2000; El-Brollosy et al., 2003) and hydrazones, Abdel-Megeed et al., 1999; El-Hiti et al., 2000; Saleh et al., 2003) as a continuation of our own interest in the synthesis and/or modification of quinazoline derivatives (Smith et al., 1995, 1996, 2003, 2004, 2005a,b; El-Hiti, 1997, 2003, 2004; El-Hiti and Abdel-Megeed, 2005; Abdel-Megeed et al., 2007).

In this work we now report the successful synthesis of a series of hydrazones containing 3*H*-quinazolin-4-one ring system under mild conditions.

2. Experimental

Melting points (°C, uncorrected) were determined on an electrothermal melting MEL-TEMP II apparatus. ^{1}H and ^{13}C NMR spectra were recorded in DMSO-d_6 using a Bruker AC400 spectrometer operating at 400 MHz for ^{1}H and 100 MHz for ^{13}C. Chemical shifts δ are reported in parts per

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Peer review under responsibility of King Saud University.

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http://dx.doi.org/10.1016/j.jscs.2011.12.009

million (ppm) relative to tetramethylsilane (TMS) and coupling constants *J* are in Hz. Assignments of signals are based on integration values, coupling patterns and expected chemical shift values and have not been rigorously confirmed. Signals with similar characteristics might be interchanged. IR Spectra were recorded on a Perkin-Elmer 1430 spectrometer using KBr disc technique. All samples were examined by thin layer chromatography (TLC), which was performed on EM silica gel F_{254} sheet (0.2 mm) with chloroform/acetone (5:2; v/v), hexane/acetone (5:2; v/v) or petroleum ether (40–60 °C)/acetone (5:2; v/v) as developing eluents. The spots were detected with UV Lamp Model UV GL-58.

*2.1. Synthesis of 6-bromo-2-hydrazino-3-phenyl-3H-quinazolin-4-one (**3**) and 6,8-dibromo-2-hydrazino-3-phenyl-3H-quinazolin-4-one (**4**)*

A mixture of **1** or **2** (10.0 mmol) and hydrazine hydrate (11.0 mmol) in EtOH (100 mL) was heated under reflux for 48 h. The solid obtained on cooling was collected by filtration, washed with EtOH and recrystallized from BuOH to give pure product.

Compound **3**: Yield 72%, m.p. 260–262 °C; IR (KBr, cm^{-1}) 3256, 3599, 1660, 580; ^{1}H NMR (DMSO-d_6) δ: 8.21–7.13 (m, 8H, aromatics), 5.81 (s, 2H, NH_2, D_2O exchange), 5.40 (s, 1H, NH, D_2O exchange); ^{13}C NMR (DMSO-d_6) δ: 160.9 (C-4), 153.2 (C-2), 138.7 (C-8a), 137.5 (C-7), 134.5 (C-1 of Ph), 130.3 (C-4 of Ph), 129.3 (C-3/C-5 of Ph), 128.9 (C-2/C-6 of Ph), 128.9 (C-5), 123.6 (C-6), 119.4 (C-4a), 114.6 (C-8).

Compound **4**: Yield 67%, m.p. 240 °C; IR (KBr, cm^{-1}) 3282, 3575, 1674, 585; ^{1}H NMR (DMSO-d_6) δ: 8.17–7.11 (m, 7H, aromatics), 5.79 (s, 2H, NH_2, D_2O exchange), 5.43 (s, 1H, NH, D_2O exchange).

*2.2. Synthesis of D-sugar-1-(6-bromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**6**) and D-sugar-1-(6,8-dibromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**7**)*

A mixture of **3** or **4** (10.0 mmol), appropriate *D*-sugar **5** (11.0 mmol) and AcOH (0.2 mL) in EtOH (20 mL) was heated under reflux for 4 h. The yellowish white solid that separated on cooling was filtered, washed with H_2O (2 × 20 mL) then EtOH (2 × 20 mL) to give pure products **6** or **7**.

Compound **6a**: Yield 71%, m.p. 215–216 °C; IR (KBr, cm^{-1}) 3320, 2925, 1675, 1582, 583; ^{1}H NMR (DMSO-d_6) δ: 9.22 (s, 1H, NH, D_2O exchange), 8.01–6.83 (m, 8H, aromatics), 5.53 (d, J = 4.7 Hz, 1H, H-1 of glucose), 4.71–3.39 (m, 11H, other glucose protons).

Compound **6b**: Yield 69%, m.p. 194–195 °C; IR (KBr, cm^{-1}) 3323, 2923, 1674, 1611, 583; ^{1}H NMR (DMSO-d_6) δ: 9.50 (s, 1H, NH, D_2O exchange), 8.01–7.09 (m, 8H, aromatics), 5.68 (d, J = 4.9 Hz, 1H, H-1 of galactose), 4.51–3.50 (m, 11H, other galactose protons).

Compound **6c**: Yield 68%, m.p. 220–222 °C; IR (KBr, cm^{-1}) 3324, 2923, 1676, 1581, 582; ^{1}H NMR (DMSO-d_6) δ: 9.31 (s, 1H, NH, D_2O exchange), 7.88–6.89 (m, 8H, aromatics), 5.50 (d, J = 5.0 Hz, 1H, H-1 of mannose), 4.72–3.43 (m, 11H, other mannose protons).

Compound **6d**: Yield 68%, m.p. 240–241 °C; IR (KBr, cm^{-1}) 3325, 2923, 1667, 1602, 583; ^{1}H NMR (DMSO-d_6) δ: 9.49 (s, 1H, NH, D_2O exchange), 8.11–7.04 (m, 8H, aromatics), 5.72 (d, J = 5.1 Hz, 1H, H-1 of xylose), 4.81–3.69 (m, 9H, other xylose protons).

Compound **6e**: Yield 69%, m.p. 180–181 °C; IR (KBr, cm^{-1}) 3325, 2932, 1673, 1610, 585; ^{1}H NMR (DMSO-d_6) δ: 9.43 (s, 1H, NH, D_2O exchange), 7.94–7.03 (m, 8H, aromatics), 5.82 (d, J = 5.1 Hz, 1H, H-1 arabinose), 4.93–3.64 (m, 9H, other arabinose protons).

Compound **7a**: Yield 70%, m.p. 276–277 °C; IR (KBr, cm^{-1}) 3323, 3063, 1685, 1583, 585; ^{1}H NMR (DMSO-d_6) δ: 9.21 (s, 1H, NH, D_2O exchange), 7.99–6.91 (m, 7H, aromatics), 5.52 (d, J = 4.8 Hz, 1H, H-1 of glucose), 4.82–3.71 (m, 11H, other glucose protons).

Compound **7b**: Yield 68%, m.p. 230–231 °C; IR (KBr, cm^{-1}) 3327, 2926, 1677, 1588, 584; ^{1}H NMR (DMSO-d_6) δ: 9.36 (s, 1H, NH, D_2O exchange), 8.01–6.92 (m, 7H, aromatics), 5.59 (d, J = 4.7 Hz, 1H, H-1 of galactose), 5.44–3.53 (m, 11H, other galactose protons).

Compound **7c**: Yield 61%, m.p. 244–245 °C; IR (KBr, cm^{-1}) 3328, 2927, 1672, 1590, 583; ^{1}H NMR (DMSO-d_6) δ: 9.40 (s, 1H, NH, D_2O exchange), 8.01–6.74 (m, 7H, aromatics), 5.53 (d, J = 5.1 Hz, 1H, H-1 of mannose), 5.40–3.83 (m, 11H, other mannose protons); ^{13}C NMR (DMSO-d_6) δ: 160.3 (C-4), 159.2 (C-1 of mannose), 147.2 (C-2), 138.7 (C-8a), 138.0 (C-7), 136.5 (C-1 of Ph), 129.4 (C-4 of Ph), 128.9 (C-3/C-5 of Ph), 128.4 (C-2/C-6 of Ph), 128.3 (C-5), 123.6 (C-8), 119.4 (C-4a), 116.3 (C-6), 73.4 (C-2 of mannose), 72.2 (C-3 of mannose), 70.5 (C-4 of mannose), 69.0 (C-5 of mannose), 64.0 (C-6 of mannose).

Compound **7d**: Yield 57%, m.p. 194–195 °C; IR (KBr, cm^{-1}) 3326, 2924, 1678, 1588, 584; ^{1}H NMR (DMSO-d_6) δ: 9.57 (s, 1H, NH, D_2O exchange), 8.11–7.03 (m, 7H, aromatics), 5.72 (d, J = 5.2 Hz, 1H, H-1 xylose), 5.31–3.79 (m, 9H, other xylose protons).

Compound **7e**: Yield 68%, m.p. 265–266 °C; IR (KBr, cm^{-1}) 3324, 2926, 1675, 1591, 586; ^{1}H NMR (DMSO-d_6) δ: 9.42 (s, 1H, NH, D_2O exchange), 8.14–6.93 (m, 7H, aromatics), 5.63 (d, J = 5.0 Hz, 1H, H-1 of arabinose), 5.22–3.63 (m, 9H, other arabinose protons).

*2.3. Synthesis of per-O-acetyl-D-sugar-1-acetyl-1-(6-bromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**8**) and per-O-acetyl-D-sugar-1-(6,8-dibromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**9**)*

A cold (0 °C) solution of **6** or **7** (1.0 g) in anhydrous pyridine (5 mL) was treated with freshly distilled Ac_2O (5 mL). The reaction mixture was stirred overnight at room temperature then poured onto iced H_2O (100 mL). The solid obtained was filtered, washed repeatedly with H_2O (4 × 20 mL) and recrystallized from EtOH to give pure products **8** or **9**.

Compound **8a**: Yield 68%, m.p. 160–161 °C; IR (KBr, cm^{-1}) 2927, 1748, 1700, 1611, 585; ^{1}H NMR (DMSO-d_6) δ: 8.13–6.53 (m, 8H, aromatics), 5.61 (d, J = 4.6 Hz, 1H, H-1 of glucose), 5.33–3.81 (m, 6H, other glucose protons), 2.22–0.84 (6s, 18H, 6CH_3CO).

Compound **8b**: Yield 68%, m.p. 100–101 °C; IR (KBr, cm^{-1}) 2924, 1747, 1697, 1612, 614; ^{1}H NMR (DMSO-d_6) δ: 8.10–6.54 (m, 8H, aromatics), 5.52 (d, J = 4.8 Hz, 1H, H-1 of galactose), 5.44–3.42 (m, 6H, other galactose protons), 2.11–0.82 (6s, 18H, 6CH_3CO).

Compound **8c**: Yield 65%, m.p. 180–181 °C; IR (KBr, cm^{-1}) 2924, 1739, 1676, 1600, 582; ^1H NMR (DMSO-d_6) δ: 8.09–6.84 (m, 8H, aromatics), 5.53 (d, J = 4.9 Hz, 1H, H-1 of mannose), 5.22–3.51 (m, 6H, other mannose protons), 2.11–1.02 (6s, 18H, $6CH_3CO$).

Compound **8d**: Yield 62%, m.p. 150–151 °C; IR (KBr, cm^{-1}) 2925, 1743, 1696, 1602, 595; ^1H NMR (DMSO-d_6) δ: 8.08–6.63 (m, 8H, aromatics), 5.57 (d, J = 5.0 Hz, 1H, H-1 of xylose), 5.42–3.64 (m, 5H, other xylose protons), 2.19–0.91 (5s, 15H, $5CH_3CO$).

Compound **8e**: Yield 66%, m.p. 90–91 °C; IR (KBr, cm^{-1}) 2935, 1747, 1699, 1604, 601; ^1H NMR (DMSO-d_6) δ: 8.02–6.91 (m, 8H, aromatics), 5.90 (d, J = 5.1 Hz, 1H, H-1 of arabinose), 5.41–3.73 (m, 5H, other arabinose protons), 2.21–1.00 (5s, 15H, $5CH_3CO$).

Compound **9a**: Yield 73%, m.p. 260–261 °C; IR (KBr, cm^{-1}) 3324, 2925, 1747, 1705, 1592, 581; ^1H NMR (DMSO-d_6) δ: 9.22 (s, 1H, NH, D_2O exchange), 8.03–6.94 (m, 7H, aromatics), 5.52 (d, J = 4.8 Hz, 1H, H-1 of glucose), 5.11–3.64 (m, 6H, other glucose protons), 2.22–0.74 (5 s, 15H, $5CH_3CO$).

Compound **9b**: Yield 65%, m.p. 200–201 °C; IR (KBr, cm^{-1}) 3325, 2924, 1748, 1707, 1591, 583; ^1H NMR (DMSO-d_6) δ: 9.38 (s, 1H, NH, D_2O exchange), 8.12–6.94 (m, 7H, aromatics), 5.54 (d, J = 4.9 Hz, 1H, H-1 of galactose), 5.24–3.63 (m, 6H, other galactose protons), 2.21–0.64 (5s, 15H, $5CH_3CO$).

Compound **9c**: Yield 63%, m.p. 220–221 °C; IR (KBr, cm^{-1}) 3326, 2926, 1743, 1697, 1601, 584; ^1H NMR (DMSO-d_6) δ: 9.32 (s, 1H, NH, D_2O exchange), 8.11–6.94 (m, 7H, aromatics), 5.54 (d, J = 4.6 Hz, 1H, H-1 of mannose), 5.23–3.54 (m, 6H, other mannose protons), 2.20–0.74 (5s, 15H, $5CH_3CO$).

Compound **9d**: Yield 53%, m.p. 180–181 °C; IR (KBr, cm^{-1}) 3323, 2926, 1748, 1707, 1591, 605; ^1H NMR (DMSO-d_6) δ: 9.41 (s, 1H, NH, D_2O exchange), 8.12–6.94 (m, 7H, aromatics), 5.54 (d, J = 4.9 Hz, 1H, H-1 of xylose), 5.65–3.55 (m, 5H, other xylose protons), 2.14–0.64 (4s, 12H, $4CH_3CO$).

Compound **9e**: Yield 59%, m.p. 250–251 °C; IR (KBr, cm^{-1}) 3324, 2930, 1742, 1699, 1590, 588; ^1H NMR (DMSO-d_6) δ: 9.45 (s, 1H, NH, D_2O exchange), 8.05–6.94 (m, 7H, aromatics), 5.55 (d, J = 5.1 Hz, 1H, H-1 of arabinose), 5.35–3.74 (m, 5H, other arabinose protons), 2.20–0.65 (4s, 12H, $4CH_3CO$).

*2.4. Synthesis of aldehyde-1-(6-bromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**11**) and aldehyde-1-(6,8-dibromo-3-phenyl-4-oxoquinazolin-2-yl)hydrazones (**12**)*

A mixture of **3** or **4** (10.0 mmol), appropriate aromatic aldehyde **10** (11.0 mmol) and piperidine (1.0 mL) in dry EtOH (30 mL) was heated under reflux for 5 h. The solid obtained on cooling was filtered, washed with EtOH (2 × 20 mL) and recrystallized from EtOH to give pure products **11** or **12**. The physical properties of **11** and **12** are recorded in Table 1.

Compound **11a**: Yield 70%, m.p. 300–301 °C; IR (KBr, cm^{-1}) 3426, 1665, 1558, 619; ^1H NMR (DMSO-d_6) δ: 9.23 (s, 1H, NH, D_2O exchange), 7.81–6.56 (m, 13H, aromatics), 5.55 (s, 1H, CH).

Compound **11b**: Yield 66%, m.p. 256–266 °C; IR (KBr, cm^{-1}) 3321, 1674, 1571, 649; ^1H NMR (DMSO-d_6) δ: 9.22 (s, 1H, NH, D_2O exchange), 8.11 (s, 1H, OH, D_2O exchange), 7.92–6.95 (m, 12H, aromatics), 6.72 (s, 1H, CH).

Table 1 Synthesis of hydrazones of **11** and **12** according to Scheme 3.

Product	X	Ar	m.p. (°C)	Yield (%)[a]
11a	H	C_6H_5	300–301	70
11b	H	$-HOC_6H_4$	265–266	66
11c	H	$-MeOC_6H_4$	223–224	67
11d	H	$-ClC_6H_4$	250–251	63
11e	H	$-NO_2C_6H_4$	280–281	73
11f	H	$-Me_2NC_6H_4$	210–211	59
12a	Br	C_6H_5	190–191	65
12b	Br	$-HOC_6H_4$	280–281	59
12c	Br	$-MeOC_6H_4$	235–236	61
12d	Br	$-ClC_6H_4$	285–286	58
12e	Br	$-NO_2C_6H_4$	240–241	68
12f	Br	$-Me_2NC_6H_4$	250–251	55

[a] Yield for pure product after crystallization from ethanol.

Compound **11c**: Yield 67%, m.p. 223–224 °C; IR (KBr, cm^{-1}) 3460, 1679, 1595, 581; ^1H NMR (DMSO-d_6) δ: 8.44 (s, 1H, NH, D_2O exchange), 8.13–6.94 (m, 12H, aromatics), 5.73 (s, 1H, CH), 3.82 (s, 3H, OMe).

Compound **11d**: Yield 63%, m.p. 250–251 °C; IR (KBr, cm^{-1}) 3321, 1683, 1593, 513; ^1H NMR (DMSO-d_6) δ: 9.24 (s, 1H, NH, D_2O exchange), 8.12–6.95 (m, 12H, aromatics), 5.57 (s, 1H, CH).

Compound **11e**: Yield 73%, m.p. 250–251 °C; IR (KBr, cm^{-1}) 3346, 1680, 1616, 582; ^1H NMR (DMSO-d_6) δ: 9.23 (s, 1H, NH, D_2O exchange), 8.20–6.96 (m, 12H, aromatics), 5.58 (s, 1H, CH).

Compound **11f**: Yield 59%, m.p. 210–211 °C; IR (KBr, cm^{-1}) 3323, 1678, 1593, 616; ^1H NMR (DMSO-d_6) δ: 8.66 (s, 1H, NH, D_2O exchange), 8.03–6.84 (m, 12H, aromatics), 5.54 (s, 1H, CH), 2.8 (s, 6H, NMe_2).

Compound **12a**: Yield 65%, m.p. 190–191 °C; IR (KBr, cm^{-1}) 3426, 1667, 1615, 614; ^1H NMR (DMSO-d_6) δ: 9.25 (s, 1H, NH, D_2O exchange), 8.41–7.25 (m, 12H, aromatics), 6.75 (s, 1H, CH).

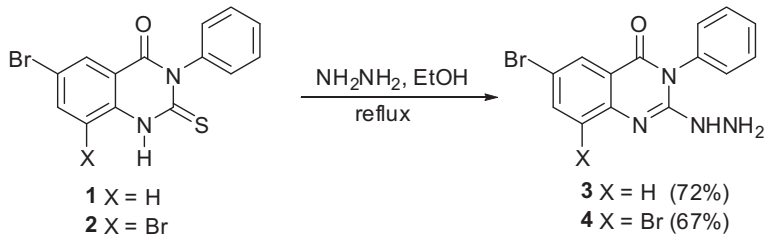
Compound **12b**: Yield 59%, m.p. 280–281 °C; IR (KBr, cm^{-1}) 3438, 1695, 1604, 541; ^1H NMR (DMSO-d_6) δ: 9.60 (s, 1H, NH, D_2O exchange), 8.22 (s, 1H, OH, D_2O exchange), 7.91–6.84 (m, 11H, aromatics), 5.84 (s, 1H, CH).

Compound **12c**: Yield 61%, m.p. 235–236 °C; IR (KBr, cm^{-1}) 3438, 1675, 1612, 582; ^1H NMR (DMSO-d_6) δ: 9.60 (s, 1H, NH, D_2O exchange), 8.22–7.15 (m, 11H, aromatics), 5.71 (s, 1H, CH), 3.80 (s, 3H, OMe).

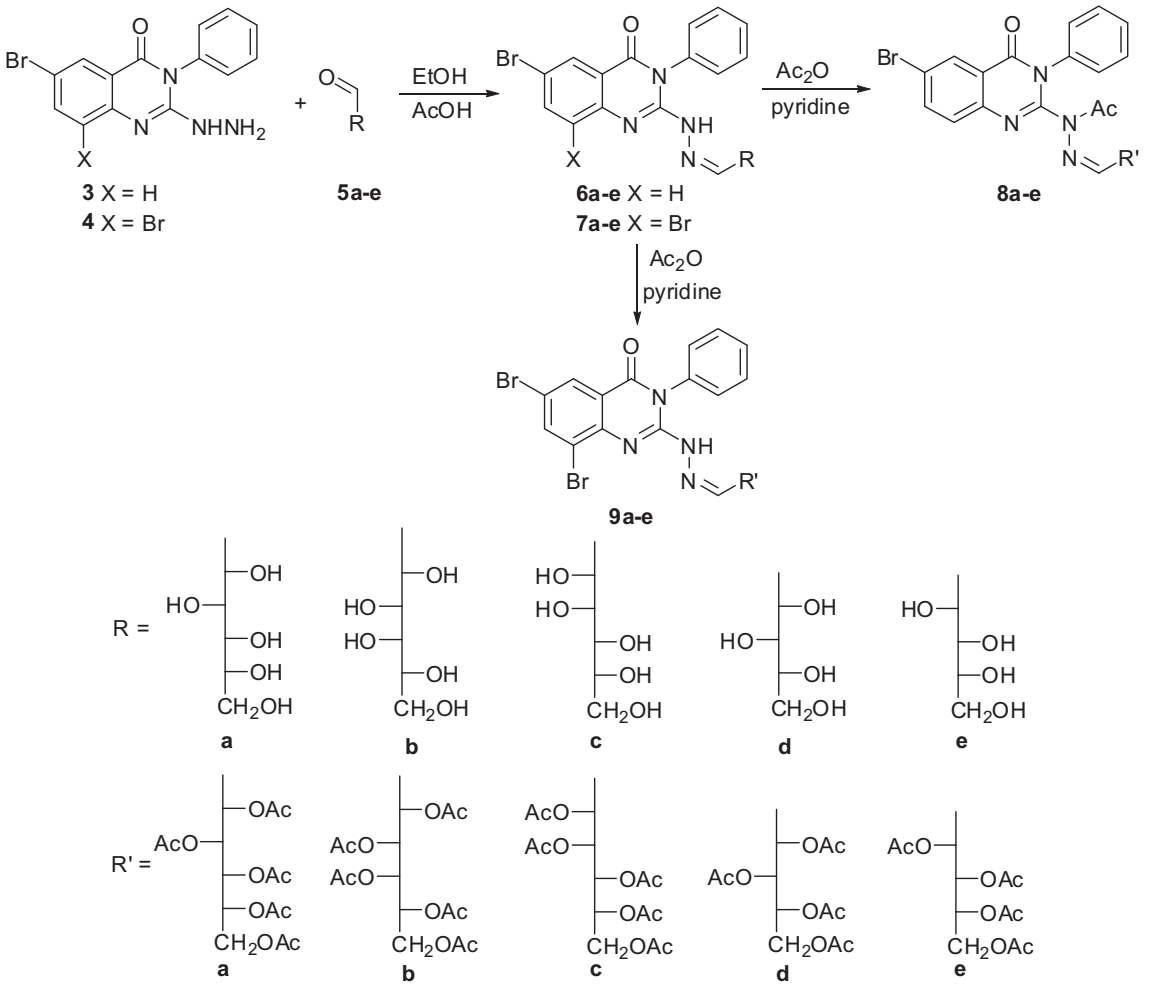
Compound **12d**: Yield 58%, m.p. 285–286 °C; IR (KBr, cm^{-1}) 3421, 1777, 1614, 584; ^1H NMR (DMSO-d_6) δ: 9.52 (s, 1H, NH, D_2O exchange), 8.32–7.05 (m, 11H, aromatics), 5.82 (s, 1H, CH).

Compound **12e**: Yield 68%, m.p. 240–241 °C; IR (KBr, cm^{-1}) 3328, 1678, 1614, 583; ^1H NMR (DMSO-d_6) δ: 9.34 (s, 1H, NH, D_2O exchange), 8.34–6.96 (m, 11H, aromatics), 5.55 (s, 1H, CH).

Compound **12f**: Yield 55%, m.p. 250–252 °C; IR (KBr, cm^{-1}) 3426, 1674, 1604, 580; ^1H NMR (DMSO-d_6) δ: 9.62 (s, 1H, NH, D_2O exchange), 8.26–6.94 (m, 11H, aromatics), 5.82 (s, 1H, CH), 3.01 (s, 6H, NMe_2).



Scheme 1 Synthesis of 2-hydrazino-3-phenyl-3*H*-quinazolin-4-ones **3** and **4**.



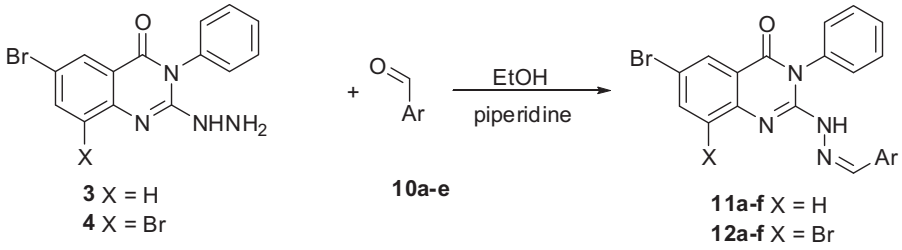
Scheme 2 Synthesis of hydrazones **6** and **7** and their acetyl derivatives **8** and **9**.

3. Results and discussion

6-Bromo-2-hydrazino-3-phenyl-3*H*-quinazolin-4-one (**3**) and 6,8-dibromo-2-hydrazino-3-phenyl-3*H*-quinazolin-4-one (**4**) were synthesized in 72% and 67% yields, respectively, from reactions of 6-bromo-3-phenyl-2-thioxo-3*H*-quinazolin-4-one (**1**) and 6,8-dibromo-3-phenyl-2-thioxo-3*H*-quinazolin-4-one (**2**) with hydrazine hydrate in boiling ethanol (Scheme 1).

Condensation of **3** and **4** with equimolar amounts of monosaccharides **5a**–**e** (D-glucose, D-galactose, D-mannose, D-xylose and D-arabinose) in ethanol and in the presence of a catalytic amount of glacial acetic acid under reflux conditions for 4 h afforded the corresponding hydrazones **6a**–**e** and **7a**–**e**, respectively (Scheme 2) in 57–71% yields.

The IR spectra of **6** and **7** showed absorption bands at 3320–3428 cm^{-1} region due to the stretching vibration of NH groups and hydroxyl groups of the sugar residue. The bands at 1667–1685 cm^{-1} and 582–586 cm^{-1} region are due to C=O and C–Br bonds, respectively. The ^1H NMR spectra of **6** and **7** are characterized by the presence of CH=N protons as doublets and resonated within the 5.50–5.82 ppm region. The NH protons resonated as exchangeable singlet signals at 9.21–9.57 ppm region.



Scheme 3 Synthesis of hydrazones **11** and **12**.

Acetylation of hydrazones **6a**–**e** with freshly distilled acetic anhydride in anhydrous pyridine at room temperature gave the corresponding acetylated hydrazones **8a**–**e** (Scheme 2) in 62–68% yields. The spectroscopic data of **8** indicated that peracetylation had taken place at both the polyol residue and NH groups. However, acetylation **7a**–**e** under similar conditions gave the corresponding acetylated hydrazones **9a**–**e** (Scheme 2) in 53–73% yields in which peracetylation had taken place only at the polyol residue without affecting the NH groups.

The IR spectra of **8** and **9** showed strong absorption bands at 1739–1748 cm^{-1} due to the stretching vibrations of ester (O–C═O) carbonyl groups. The IR spectra of **8** showed the absence of absorption bands due to NH groups. While, IR spectra of **9** showed absorption bands at 3323–3326 cm^{-1} due to the stretching vibration of NH groups. The ^1H NMR spectra of **8** showed the absence of any exchangeable signals due to the NH protons. While, ^1H NMR spectra of **9** showed the presence of exchangeable singlet signals that resonated at 9.22–9.45 ppm region due to the NH protons.

Our attention was next turned to attempt reactions of **3** and **4** with aromatic aldehydes. It was found that reactions of **3** and **4** with aromatic aldehydes **10a**–**f**, such as benzaldehyde, 4-hydroxybenzaldehyde, 4-anisaldehyde, 4-chlorobenz-aldehyde, 4-nitrobenzaldehyde and 4-(*N*,*N*-dimethylamino)benzaldehyde, in dry ethanol and in the presence of few drops of piperidine as a catalyst gave the corresponding hydrazones **11a**–**f** and **12a**–**f**, respectively (Scheme 3) in good yields. The physical data of **11** and **12** are recorded in Table 1.

The IR spectra of **11** and **12** showed strong absorption bands that appeared at 1674–1677 cm^{-1} region due to the stretching vibrations of the quinazolinone carbonyl group. They also showed absorption bands within the 3321–3460 cm^{-1} region due to the stretching vibrations of the NH groups and absorption bands at 513–649 cm^{-1} region are due to C–Br bonds. ^1H NMR spectra of **11** and **12** showed the presence of CH═N protons that resonated at 5.57–6.75 ppm region, while the NH protons resonated as exchangeable singlet signals at 8.44–9.62 ppm region.

Acknowledgement

The author expresses his sincere thanks and gratitude to Professor Keith Smith, School of Chemistry, Cardiff University, UK for recording the NMR spectra.

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