Gold(III) promoted formation of dihydroquinazolinones: double X–H activation by gold†

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An efficient 2-furyl gold–carbene promoted synthetic method was developed for the formation of dihydroquinazolinones from enynones by dual insertion of anthranilamides. In this organic transformation a new C–O and two C–N bond formations occurred and dihydroquinazolinones were obtained with a quaternary centre in moderate to very good yields in one-pot synthesis.

Introduction

Nitrogen-containing heterocyclic molecules such as quinazo-}
linones have gained much attention due to their wide range of biological and pharmacological applications. Dihy-
droquinazolinone derivatives like fenquizone, and quinet-
zone are drugs for edema and hypertension. It was reported that bouchardatine exhibits antiobesity activity, and pen-
ipanoid C exhibits tobacco mosaic virus inhibition (Fig. 1). Further, substituted dihydroquinazolinone derivatives displayed significant cytotoxic activity.

Hence, the development of new synthetic methods for the formation of dihydroquinazolinones is a limitless frontier. Cooperative catalysis has been established as a handy tool for the synthesis of several biologically valuable molecules and different procedures were reported for the synthesis of dihy-
droquinazolinone derivatives. Exploration of gold-catalyzed organic transformations has attracted much attention in recent years due to their broad functional group tolerance and selectivity for the formation of valuable heterocyclic molecules in one-pot reaction conditions. The recent literature indicating that exploitation of enynal/enynone has recognised as good donor–donor carbene precursors for C–H/X–H inser-
tion and cyclopropanation reactions. Several reports are available for synthesis of substituted furans from enynones in presence of metal catalysts via a 5-exo-dig cyclization.

Fig. 1 Selected examples of important molecules containing dihy-
droquinazolinone core skeleton.

Scheme 1 Synthetic transformations of enynones.
The reaction mechanism was proposed via (2-furyl) metal–carbene intermediate would react with one nucleophile to produce addition products (Scheme 1, eqn (1)). López and Vicente co-workers reported a method for synthesis of functionalized furans from enynones in the presence of zinc catalyst. Recently, Zhu et al. developed metal carbene promoted method for synthesis of vinyl-substituted dihydroindoles. Hashmi and co-workers studied the stabilization effects of gold carbene complexes.

Double insertion of isocyanides to enynones produced pyrrole-fused heterocyclic molecules via (2-furyl) metal–carbene intermediate was reported by Jia and Li co-workers (Scheme 1, eqn (2)). Very recently, we have reported formation of tetraarylmethane derivatives by reaction of enynones with indoles via (2-furyl) gold–carbene intermediate (Scheme 1, eqn (3)). Our current research efforts focused to explore the reactivity of enynones under gold catalysis. We envisioned that reaction of enynones (1) in the presence of gold-catalyst would produce gold–carbene complex I, which would react with anthranilamide (2) may give corresponding dihydroquinazolinone derivative 3 (Scheme 1, eqn (4)).

### Results and discussion

Accordingly, we have conducted an experiment by using substrates 1a and 2a in the presence of AuCl₃ (Scheme 2). Very interestingly, 21% yield of the corresponding product 2-(4-benzoyl-5-phenylfuran-2-yl)-2-phenyl-2,3-dihydroquinazolin-4(1H)-one 3a was observed. The product 3a was further...
confirmed by single crystal X-ray analysis. It is noteworthy that in this organic transformation two C-N bonds were formed by dual insertion of anthranilamide with a quaternary centre. This interesting observation encouraged us to optimize this reaction to get the better yields of the product 3a.

Gold(i) catalysts were screened with the substrates 1a with 2a to produce moderate yields of 3a along with 3a' (Table 1, entries 1 and 2). Whereas when experiments were conducted in the presence of AuBr₃ and KAuCl₄ moderate yields of product 3a was observed (Table 1, entries 3 and 4). Complex mixture was obtained when reaction was performed in the presence of IPrAuCl₃ (Table 1, entry 5). In the presence of AuCl₂PPh₃, poor yields of desired product 3a was found (Table 1, entry 6). Then reactions were conducted by utilizing gold catalysts in combination of selectfluor (Table 1, entries 7–10), yields of desired product 3a was not improved. Reactions were performed by employing KAuCl₄ in combination with K₂S₂O₈, CF₃COOH, Cu(OAc)₂, K₂CO₃, pyridine N-oxide and PhI(OAc)₂ (Table 1, entries 11–16), moderate yield of 3a was observed. Nevertheless, KAuCl₄ and FeCl₃ combination afforded very good yield of (81%) of product 3a (Table 1, entry 17). An experiment was conducted by utilizing only FeCl₃, poor yields of product 3a was observed along with 3a' (Table 1, entry 18). Reactions were screened by using series of solvents like toluene, MeOH, THF, DMF and DCE, none of them gave better yield than MeCN (Table 1, entries 19–23). When the gold-catalyst loading decreased from 10 mol% to 5 mol% and 7 mol%, the product yield also reduced to 52% and 58%, respectively (Table 1, entries 24 and 25). Two reactions were conducted without utilizing FeCl₃ and these cases poor yields of product 3a observed (Table 1, entries 26 and 27).

The above experiments concludes that Table 1, entry 17 is the best suitable reaction conditions. Then substrate scope was tested by utilizing different enynones (1a–k) with anthranilamide 2a under the optimal conditions. These results are incorporated in the Table 2.

The substrates which are bearing electron-donating groups such as 1b and 1c were tested with 2a to provide 76% and 72% yields of corresponding products 3b and 3c, respectively. Electron-withdrawing functional group containing enynone such as 1d react with 2a to give the corresponding dihydroquinazolinone derivative 3d in 68% yield. Substrates bearing electron-donating groups like 1e and 1f reacted with 2a to produce 75% and 74% yields of the corresponding dihydroquinazolinone derivatives 3e and 3f, respectively. Both electron-donating and electron-withdrawing functional groups containing enynone like 1g reacted with 2a to generate the corresponding product 3g in 71% yield. The substrates which are having electron-donating groups at ortho position of R¹ like 1h (R² = 3-Me-C₆H₄), 1i (R² = 3-Me-C₆H₄, R¹ = 4-Me-C₆H₄) and 1j (R¹ = 3-Me-C₆H₄) reacted with 2a to provide the corresponding products 3h, 3i and 3j in moderate yields, respectively (Table 2, entries 8–10). Alkyl substitution at R² position containing substrate like 1k produced the product 3k in 43% yield (Table 2, entry 11).

Further, experiments were conducted to check the scope of dihydroquinazolinone derivatives by utilizing different substituted anthranilamides (2b–e) with enynones (1a–f). These results were included in the Table 3. The enynone 1a was tested with electron-donating functional group containing anthranilamide 2b to give 3l in 72% yield. Electron-withdrawing functional group containing anthranilamides such as 2c, 2d, and 2e reacted with 1a to produce the corresponding dihydroquinazolinone derivatives 3m, 3n, and 3o in 65%, 63% and 60% yields, respectively (Table 3, entries 2–4). Enynones bearing
electron donating groups such as 1b and 1c reacted with 2b to provide corresponding products 3p and 3q in 72% and 70% yields, respectively (Table 3, entries 5 and 6). Fluorine substituted enynone such as 1d reacted with 2b, 2c, 2d and 2e to provide the corresponding dihydroquinazolinone derivatives such as 3r, 3s, 3t and 3u in 69%, 63%, 61% and 62% yields, respectively (Table 3, entries 7 and 10). Electron-donating substitutions containing enynones such as 1e and 1f reacted with 2b under optimized reaction conditions to give the corresponding products 3v and 3w in 70% and 68% yields, respectively (Table 3, entries 11 and 12).

2-Amino-6-phenyl-4-(trifluoromethyl)nicotinamide 2f reacted with 1a to produce the corresponding product 3t in 58% yield (Scheme 3, eqn (1)). An experiment was conducted by employing a phosphorus substituted enynone like 1l with 2a to provide the corresponding dihydroquinazolinone derivative 3y in 68% yield (Scheme 3, eqn (2)). Further, one more experiment was conducted in gram scale by utilizing 1a and 2a under optimized reaction conditions to give the corresponding product 3a in 74% yield (Scheme 3, eqn (3)).

Control experiments were conducted to clarify the reaction mechanism (Scheme 4). The substrate 1a was tested under optimized conditions to produce good yields of product 3a’ (Scheme 4, eqn (1)). A reaction was conducted by utilizing 3a’ with anthranilamide 2a in the presence of gold-catalyst to provide 32% yield of product 3a (Scheme 4, eqn (2)). Without using catalyst one reaction was conducted by using 3a’ and 2a,
in this case product 3a was not observed (Scheme 4, eqn (3)). Then 1a was tested with 2-(prop-2-yn-1-olxy)benzohydrazide 4 to give 62% yield of product 5. The structure of the compound 5a further characterized by single crystal X-ray analysis.22

Formation of dihydroquinazolinones can be proposed by the reaction mechanism as shown in Scheme 5. Gold catalyst would coordinate with enyne 1a may form complex-A, which would further generate 2-furyl gold carbene complex via intramolecular of 5-exo-dig cyclised zwiterionic complex-B.24 Then, it would produce ketone (3a),13,14 which would coordinate with ferric chloride as a lewis acid in a regioselective fashion then it would react with anthranilamide (2a) may generate IM-I. Subsequent activation of IM-I by metal catalyst may lead to cyclization to form intermediate IM-II, which would finally afford the product 3a.

**Conclusion**

In conclusion, we have established gold-catalyzed reaction of enynes with dual insertion of anthranilamides to produce a novel approach for synthesis of dihydroquinazolinones. It is significant that in this organic transformation new C–O and two C–N bonds were formed with a quaternary centre with good functional group tolerance.

**Experimental section**

**General information**

Reactions were carried out in oven dried reaction flasks under nitrogen atmosphere and also solvents and reagents were transferred by oven-dried syringes to ambient temperature. TLC was performed on Merck silica gel aluminium sheets using UV as a visualizing agent. Solvents were removed under reduced pressure. Columns were packed as slurry of silica gel in hexane and ethyl acetate solvent mixture. The elution was assisted by applying pressure with an air pump.13C NMR spectra were recorded on 75, 100 and 125 MHz spectrometers.1HNMR spectra were recorded on 300, 400 and 500 MHz spectrometers in appropriate solvents using TMS as internal standard. The following abbreviations were used to explain multiplicities: s = singlet, d = doublet, dd = double doublet, dt = doublet of triplet, td = triplet of doublet, t = triplet, m = multiplet, br s = broad singlet. All reactions were performed under nitrogen atmosphere with freshly distilled and dried solvents. All solvents were distilled using standard procedures. Unless otherwise noted, reagents were obtained from Aldrich, Alfa Aesar, and TCI used without further purification. Synthesis of enynes (1a–I) were prepared by following reported procedures.25

**General procedure for synthesis of dihydroquinazolinone derivatives (3a)**

To a 10 mL round-bottomed flask equipped with magnetic stir bar the substrate 2-aminobenzamide 2a (0.45 mmol, 61 mg, 1.5 equiv.) was taken and dissolved in dry CH3CN (3 mL) at 80 °C (oil bath) after that 1,3-diphenyl-2-(3-phenylprop-2-yn-1-ylidene)propane-1,3-dione 1a (0.3 mmol, 100 mg, 1 equiv.) was added. To this reaction mixture KAuCl4 (10 mol%, 11 mg) and FeCl3 (0.6 mmol, 97 mg, 2.0 equiv.) was added and stirred at 80 °C for 5 h under nitrogen atmosphere. Progress of the reaction was monitored by using TLC. After completion of the reaction, the reaction mixture was filtered through celite plug and washed with ethyl acetate. The ethyl acetate layer was concentrated under reduced pressure to get crude residue which was purified by column chromatography through silica gel using hexane and ethyl acetate as eluent (10 : 3.5) to give 113 mg of the product 2-(4-benzoyl-5-phenylfuran-2-yl)-2-phenyl-2,3-dihydroquinazolin-4(1H)-one 3a (81% yield). The same reaction was conducted on a gram scale by utilizing 1a (1 g) and 2a (0.61 g) produced the corresponding product 3a in 74% yield (1.03 g). A similar experimental procedure was adopted for the synthesis of all the furan containing dihydroquinazolinones (3b–y) and 5.

2-(4-Benzoyl-5-phenylfuran-2-yl)-2-phenyl-2,3-dihydroquinazolin-4(1H)-one (3a). Rf: 0.5; hexane : ethyl acetate mixture (10 : 3.5); yellow solid with 113 mg (81%) yield; melting point: 188–190 °C; 1H NMR (500 MHz, CDCl3); δ 7.93 (dd, J = 7.7, 1.2 Hz, 1H), 7.73–7.66 (m, 2H), 7.63–7.56 (m, 4H), 7.52–7.48 (m, 1H), 7.47–7.42 (m, 3H), 7.37–7.31 (m, 3H), 7.30–7.26 (m, 3H), 6.97–6.87 (m, 1H), 6.79–6.70 (m, 2H), 6.35 (s, 1H), 5.09 (br s, 1H), ppm; 13C NMR (100 MHz, CDCl3); δ 191.1, 164.0, 156.3, 153.1, 145.2, 140.4, 137.5, 134.4, 133.0, 129.7, 129.6, 129.3, 129.0, 128.8, 128.4, 128.3, 128.2, 127.4, 126.9, 120.9, 119.8, 115.1, 114.9, 114.0, 72.7 ppm; IR(KBr): ν = 3368, 3057, 2922, 1654, 1613, 1485, 1367, 1262 cm–1; HRMS (ESI-TOF) m/z: [M + H]+ calecd for C31H21N2O4H 471.1703, found 471.1705.

**Crystal data for 3a.** C19H121N2O5 (M =470.50 g mol–1); triclinic, space group P1 (no. 2), α = 8.2197(2) Å, b = 10.4608(2) Å, c = 14.4752(3) Å, α = 77.0948(8)°, β = 79.6924(8)°, γ = 81.5655(9)°, V = 1188.21(4) Å3, Z = 2, T = 294.15 K, μ(MoKα) = 0.085 mm–1, Δcalc 1.317 g cm–3, 35683 reflections measured (4.64° ≤ 2θ ≤ 61.01°), 7216 unique (Rint = 0.0618, Rsigma = 0.0518) which were used in all calculations. The final R1 was 0.0634 (I > 2σ(I)) and wR2 was 0.1669 (all data). CCDC 1863534.

2-(4-Benzoyl-5-phenylfuran-2-yl)-2-(p-tolyl)-2,3-dihydroquinazolin-4(1H)-one (3b). Following the general procedure, 100 mg (0.285 mmol, 1.0 equiv.) of 1b, 58 mg (0.428 mmol, 1.5 equiv.) of 2a, 10 mg (0.10 mol%) of KAuCl4 and 92 mg (0.571 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.5; hexane/ethyl acetate mixture 10/3.5), 105 mg
of 3b was obtained in 76% yield as a yellow solid. Mp: 166–168 °C; 1H NMR (500 MHz, CDCl3): δ 7.92 (dd, J = 7.8, 1.2 Hz, 1H), 7.74–7.65 (m, 2H), 7.64–7.56 (m, 2H), 7.53–7.43 (m, 3H), 7.39–7.31 (m, 3H), 7.30–7.21 (m, 5H), 6.94–6.87 (m, 1H), 6.75–6.70 (m, 2H), 6.31 (s, 1H), 5.06 (br s, 1H), 2.38 (s, 3H) ppm; 13C NMR (100 MHz, CDCl3): δ 191.1, 163.8, 156.2, 154.3, 153.4, 139.7, 137.6, 137.5, 134.2, 132.9, 129.6, 129.4, 129.2, 128.37, 128.31, 128.2, 127.4, 126.8, 120.9, 119.7, 115.1, 114.8, 113.8, 72.5, 21.0 ppm; IR(KBr): ν = 3376, 3068, 2922, 1654, 1612, 1484, 1368, 1267 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ cale for C14H12N2O3H 248.0859, found 248.0862.

2-(4-Benzoyl-5-phenylfurazan-2-yl)-2-(4-methoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one (3c). Following the general procedure, 100 mg (0.273 mmol, 1.0 equiv.) of 1, 55 mg (0.409 mmol, 1.5 equiv.) of 2a, 10 mg (10 mol%) of KAuCl4 and 88 mg (0.546 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.5; hexane/ethyl acetate mixture 10/3.5), 95 mg of 3c was obtained in 72% yield as a yellow solid. Mp: 111–113 °C; 1H NMR (400 MHz, CDCl3): δ 7.92 (d, J = 7.3 Hz, 1H), 7.75–7.46 (m, 6H), 7.39–7.23 (m, 7H), 7.01–6.88 (m, 3H), 6.79–6.65 (m, 2H), 6.28 (s, 1H), 5.04 (br s, 1H), 3.83 (s, 3H), ppm; 13C NMR (100 MHz, CDCl3): δ 191.1, 163.9, 160.5, 156.3, 153.3, 145.3, 137.5, 134.3, 133.0, 132.5, 129.1, 128.4, 128.3, 128.2, 127.4, 120.9, 119.8, 115.2, 114.8, 114.0, 113.9, 72.4, 55.3 ppm; IR(KBr): ν = 3285, 3058, 2925, 1655, 1609, 1507, 1368, 1254 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ cale for C14H12N2O3H 513.2186, found 513.2182.

2-(4-Benzoyl-5-phenylfurazan-2-yl)-2-(4-fluorophenyl)-2,3-dihydroquinazolin-4(1H)-one (3d). Following the general procedure, 100 mg (0.282 mmol, 1.0 equiv.) of 1d, 57 mg (0.423 mmol, 1.5 equiv.) of 2a, 10 mg (10 mol%) of KAuCl4 and 91 mg (0.564 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.5; hexane/ethyl acetate mixture 10/3.5), 95 mg of 3d was obtained in 68% yield as a yellow solid. Mp: 138–140 °C; 1H NMR (400 MHz, CDCl3): δ 7.91 (d, J = 7.4 Hz, 1H), 7.73–7.47 (m, 7H), 7.39–7.24 (m, 6H), 7.17–7.06 (m, 2H), 6.98–6.87 (m, 1H), 6.80–6.69 (m, 2H), 6.45 (s, 1H), 5.07 (br s, 1H) ppm; 13C NMR (100 MHz, CDCl3): δ 190.1, 163.8, 162.2 (d, JCF = 250.148 Hz), 156.4, 152.9, 145.1, 137.4, 136.4, 134.4, 133.0, 129.6, 129.4, 129.12 (d, JCF = 8.069 Hz), 128.9, 128.3, 128.2, 127.4, 120.9, 110.7 (d, JCF = 21.274 Hz), 115.1, 114.9, 114.0, 72.3 ppm; IR(KBr): ν = 3283, 3060, 2922, 1656, 1609, 1493, 1367, 1229 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ cale for C14H12F3N2O3H 489.1609, found 489.1607.

2-(4-(4-Methylbenzoyl)-5-p-tolylfurazan-2-yl)-2-phenyl-2,3-dihydroquinazolin-4(1H)-one (3e). Following the general procedure, 100 mg (0.274 mmol, 1.0 equiv.) of 1e, 56 mg (0.412 mmol, 1.5 equiv.) of 2a, 10 mg (10 mol%) of KAuCl4 and 89 mg (0.549 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.5; hexane/ethyl acetate mixture 10/3.5), 103 mg of 3e was obtained in 75% yield as a yellow solid. Mp: 223–225 °C; 1H NMR (500 MHz, CDCl3): δ 7.90 (d, J = 7.4 Hz, 1H), 7.62–7.56 (m, 4H), 7.51 (d, J = 8.0 Hz, 2H), 7.44–7.39 (m, 3H), 7.36–7.30 (m, 1H), 7.14 (d, J = 7.3 Hz, 2H), 7.06 (d, J = 7.1 Hz, 2H), 6.93–6.87 (m, 1H), 6.73 (d, J = 7.9 Hz, 1H), 6.69 (s, 1H), 6.55–6.27 (m, 1H), 5.28–5.02 (br s, 1H), 2.37 (s, 3H), 2.30 (s, 3H) ppm; 13C NMR (100 MHz, CDCl3): δ 190.8, 163.8, 156.2, 152.6, 145.3, 143.8, 140.6, 139.4, 135.0, 134.2, 129.7, 129.5, 129.0, 128.9, 128.6, 128.3, 127.2, 126.9, 126.3, 120.4, 119.6, 115.1, 114.8, 114.0, 72.6, 21.5, 21.2 ppm; IR(KBr): ν = 3357, 3050, 2918, 1653, 1612, 1507, 1372, 1271, 1179 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ cale for C16H16N2O3H 468.1146, found 468.0998.
1H), 7.71–7.67 (m, 2H), 7.63–7.57 (m, 2H), 7.53–7.46 (m, 2H), 7.37–7.23 (m, 9H), 6.94–6.89 (m, 1H), 6.76–6.72 (m, 2H), 6.33 (s, 1H), 5.08 (br s, 1H), 2.39 (s, 3H). ppm; 1H NMR (100 MHz, DMSO-d6 & CDCl3): δ 189.1, 162.5, 153.7, 153.0, 145.2, 140.2, 136.0, 132.0, 131.4, 127.9, 127.8, 127.7, 126.8, 126.7, 126.5, 126.2, 125.8, 122.7, 119.2, 116.4, 113.4, 113.2, 111.6, 70.6, 19.9. ppm; IR(KBr): ʋ = 3335, 3061, 2919, 1668, 1487, 1265, 1148, 756 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺ 190.1205; calculated for C9H7N2O2 190.1206.

2-(4-(Methylbenzyl)-5-(p-tolyl)furane-2-yl)-2-(m-tolyl)-2,3-dihydroquinazolin-4(1H)-one (3). Following the general procedure, 100 mg (0.264 mmol, 1.0 equiv.) of 1a, 54 mg (0.396 mmol, 1.5 equiv.) of 2a, 10 mg (10 mol%) of KauCl4 and 85 mg (0.529 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3.0), 56 mg of 3i was obtained in 41% yield as a light yellow solid. Mp: 204–206 °C; 1H NMR (400 MHz, CDCl3): δ 7.93 (dd, J = 7.8, 1.3 Hz, 1H), 7.60 (d, J = 8.1 Hz, 2H), 7.52 (d, J = 8.1 Hz, 2H), 7.46 (s, 1H), 7.37–7.23 (m, 4H), 7.15 (d, J = 7.9 Hz, 2H), 7.08 (d, J = 8.0 Hz, 2H), 6.94–6.89 (m, 1H), 6.72 (d, J = 7.9 Hz, 1H), 6.68 (s, 1H), 6.24 (s, 1H), 5.40 (br s, 1H), 2.38 (d, J = 1.5 Hz, 6H), 2.31 (s, 3H). ppm; 13C NMR (75 MHz, CDCl3): δ 191.9, 163.9, 136.2, 135.1, 135.0, 130.5, 129.8, 129.0, 128.9, 128.6, 128.4, 127.6, 127.3, 126.4, 123.9, 120.5, 119.8, 115.2, 114.9, 114.1, 72.7, 21.6, 21.5, 21.3. ppm; IR(KBr): ʋ = 3448, 3034, 2919, 1633, 1497, 1215, 1171, 754 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺, calculated for C19H13N2O2 329.1148, found 329.1144.

2-(4-(Methylbenzyl)-5-(p-tolyl)furane-2-yl)-2-(m-tolyl)-2,3-dihydroquinazolin-4(1H)-one (3). Following the general procedure, 100 mg (0.264 mmol, 1.0 equiv.) of 1a, 54 mg (0.396 mmol, 1.5 equiv.) of 2a, 10 mg (10 mol%) of KauCl4 and 85 mg (0.529 mmol, 2.0 equiv.) of FeCl3 was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3.0), 56 mg of 3i was obtained in 41% yield as a light yellow solid. Mp: 204–206 °C; 1H NMR (400 MHz, CDCl3): δ 7.93 (dd, J = 7.8, 1.3 Hz, 1H), 7.60 (d, J = 8.1 Hz, 2H), 7.52 (d, J = 8.1 Hz, 2H), 7.46 (s, 1H), 7.37–7.23 (m, 4H), 7.15 (d, J = 7.9 Hz, 2H), 7.08 (d, J = 8.0 Hz, 2H), 6.94–6.89 (m, 1H), 6.72 (d, J = 7.9 Hz, 1H), 6.68 (s, 1H), 6.24 (s, 1H), 5.40 (br s, 1H), 2.38 (d, J = 1.5 Hz, 6H), 2.31 (s, 3H). ppm; 13C NMR (75 MHz, CDCl3): δ 191.9, 163.9, 136.2, 135.1, 135.0, 130.5, 129.8, 129.0, 128.9, 128.6, 128.4, 127.6, 127.3, 126.4, 123.9, 120.5, 119.8, 115.2, 114.9, 114.1, 72.7, 21.6, 21.5, 21.3. ppm; IR(KBr): ʋ = 3448, 3034, 2919, 1633, 1497, 1215, 1171, 754 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺, calculated for C19H13N2O2 329.1148, found 329.1144.
129.8, 129.6, 128.9, 128.8, 128.3, 128.2, 127.4, 126.9, 126.0, 116.7, 114.1, 111.8, 72.7 ppm; IR(KBr): v = 3325, 3046, 2923, 1651, 1605, 1490, 1319, 1144 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ calc'd for C₁₄H₁₂N₂O₂H 273.1100, found 273.1091.

2-(4-Benzoyl-5-phenylfuran-2-yl)-6-iodo-2-phenyl-2,3-dihydroquinazolin-4(1H)-one (3a). Following the general procedure, 100 mg (0.3 mmol, 1.0 equiv.) of 1b, 64 mg (0.428 mmol, 1.5 equiv.) of 2b, 10 mg (0.6 mmol, 10% equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3.0), 90 mg of 3a was obtained in 72% yield as a yellow solid. Mp: 102–104 °C; ¹H NMR (400 MHz, CDCl₃): δ 7.69 (d, J = 7.3 Hz, 2H), 7.66–7.60 (m, 2H), 7.54–7.43 (m, 3H), 7.38–7.32 (m, 2H), 7.29–7.21 (m, 5H), 7.18–7.13 (m, 1H), 6.73 (s, 1H), 6.69 (d, J = 7.4 Hz, 1H), 6.58 (d, J = 7.9 Hz, 1H), 6.25 (s, 1H), 5.01 (br s, 1H), 3.85 (s, 3H), 2.67 (s, 3H) ppm; ¹³C NMR (100 MHz, CDCl₃): δ 191.1, 164.4, 156.1, 153.4, 146.6, 142.2, 137.6, 133.1, 132.9, 129.6, 129.4, 129.2, 128.9, 128.7, 127.4, 126.8, 125.8, 123.0, 120.8, 114.0, 113.7, 71.3, 71.9, 22.1, 21.1 ppm; IR(KBr): ν = 3321, 3037, 2919, 1652, 1601, 1507, 1373, 1269 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ calc'd for C₁₃H₁₂ClIF₃NO₂H 523.1219, found 523.1230.

2-(4-Benzoyl-5-phenylfuran-2-yl)-6-chloro-2-(4-fluorophenyl)-2,3-dihydroquinazolin-4(1H)-one (3b). Following the general procedure, 100 mg (0.328 mmol, 1.0 equiv.) of 1d, 63 mg (0.423 mmol, 1.5 equiv.) of 2b, 10 mg (0.6 mmol, 10% equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3.0), 93 mg of 3b was obtained in 63% yield as a yellow solid. Mp: 130–132 °C; ¹H NMR (400 MHz, CDCl₃): δ 7.85 (d, J = 2.2 Hz, 1H), 7.68 (d, J = 7.4 Hz, 2H), 7.59–7.47 (m, 5H), 7.37–7.22 (m, 6H), 7.15–7.06 (m, 2H), 6.76–6.67 (m, 3H), 5.19 (br s, 1H) ppm; ¹³C NMR (100 MHz, CDCl₃): δ 190.9, 163.3 (d, J_C-F = 250.8 Hz), 162.8, 156.5, 152.4, 143.5, 137.3, 136.0, 134.3, 133.1, 129.6, 129.5, 129.13 (d, J_C-F = 8.1 Hz), 128.8, 128.4, 128.3, 127.9, 127.5, 125.2, 120.9, 116.5, 116.4, 115.8 (d, J_C-F = 22.0 Hz), 114.2, 72.3 ppm; IR(KBr): ν = 3279, 3062, 2923, 1660, 1607, 1486, 1347, 1231 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]+ calc'd for C₁₃H₁₂ClF₂NO₂H 528.1309, found 528.1318.
1144 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺ caged for C₁₁H₁₀BrF₅O₇N₂H 567.0714, found, 567.0732.

2-(4-Benzyl-5-phenylfuran-2-yl)-2-(4-fluorophenyl)-6-ido-2,3-dihydroquinazolin-4(1H)-one (3u). Following the general procedure, 100 mg (0.282 mmol, 1.0 equiv.) of 1d, 111 mg (0.423 mmol, 1.5 equiv.) of 2e, 10 mg (10 mol%) of KAuCl₄ and 91 mg (0.564 mmol, 2.0 equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3), 107 mg of 3u was obtained in 62% yield as a yellow solid. Mp: 194–196 °C; ¹H NMR (400 MHz, CDCl₃): δ 8.20 (d, J = 1.5 Hz, 1H), 7.72–7.67 (m, 2H), 7.62–7.49 (m, 6H), 7.40–7.33 (m, 2H), 7.30–7.24 (m, 4H), 7.16–7.08 (m, 1H), 6.74 (s, 1H), 6.54 (d, J = 8.4 Hz, 1H), 6.49 (s, 1H), 5.13 (br s, 1H) ppm; ¹³C NMR (75 MHz, DMSO-d₆ & CDCl₃): δ 188.8, 160.95, 160.93 (d, J_C=CF = 247.0 Hz), 153.8, 152.1, 144.4, 140.0, 136.0, 135.8, 130.4, 131.4, 128.2, 127.6, 127.5, 126.5 (d, J_C=CF = 110 Hz), 125.9, 119.1, 115.9, 115.3, 113.4 (d, J_C=CF = 22.0 Hz), 111.7, 70.0 ppm; IR(KBr): ν = 3319, 3062, 1650, 1601, 1494, 1319, 1231, 1153 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺ caged for C₂₃H₁₅F₂N₂O₂H 615.0575, found, 615.0602.

5-Methyl-2-(4-(methylbenzyl)-5-p-tolylfuran-2-yl)-2-phenyl-2,3-dihydroquinazolin-4(1H)-one (3v). Following the general procedure, 100 mg (0.274 mmol, 1.0 equiv.) of 1e, 62 mg (0.412 mmol, 1.5 equiv.) of 2b, 10 mg (10 mol%) of KAuCl₄ and 89 mg (0.549 mmol, 2.0 equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.7; hexane/ethyl acetate mixture 10/2.5), 99 mg of 3v was obtained in 70% yield as a yellow solid. Mps: 180–182 °C; ¹H NMR (400 MHz, CDCl₃): δ 7.64–7.52 (m, 6H), 7.46–7.40 (m, 3H), 7.19–7.13 (m, 3H), 7.08 (d, J = 8.0 Hz, 2H), 6.73–6.66 (m, 2H), 6.58 (d, J = 7.9 Hz, 1H), 6.24 (s, 1H), 5.04 (br s, 1H), 2.68 (s, 3H), 2.39 (s, 3H), 2.31 (s, 3H) ppm; ¹³C NMR (100 MHz, CDCl₃): δ 190.8, 164.3, 156.2, 156.6, 146.3, 143.8, 142.3, 140.6, 139.4, 135.2, 133.0, 129.7, 129.6, 129.0, 128.9, 128.7, 127.2, 126.5, 125.3, 126.0, 114.4, 113.7, 113.2, 72.1, 22.2, 21.6, 21.3 ppm; IR(KBr): ν = 3236, 3056, 2922, 1652, 1602, 1499, 1368, 1269 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺ caged for C₂₅H₂₂F₂N₂O₂H 513.2172, found, 513.2177.

5-Methyl-2-(4-(methylbenzyl)-5-p-tolylfuran-2-yl)-2-p-tolyl-2,3-dihydroquinazolin-4(1H)-one (3w). Following the general procedure, 100 mg (0.264 mmol, 1.0 equiv.) of 1f, 60 mg (0.369 mmol, 1.5 equiv.) of 2b, 10 mg (10 mol%) of KAuCl₄ and 85 mg (0.529 mmol, 2.0 equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.7; hexane/ethyl acetate mixture 10/2.5), 95 mg of 3w was obtained in 68% yield as a yellow solid. Mps: 226–228 °C; ¹H NMR (300 MHz, CDCl₃): δ 7.88 (d, J = 7.7 Hz, 1H), 7.50–7.30 (m, 6H), 6.87 (t, J = 7.7 Hz, 1H), 6.71 (d, J = 7.9 Hz, 1H), 6.50 (d, J = 3.3 Hz, 1H), 6.27 (s, 1H), 4.99 (br s, 1H), 3.75–3.63 (m, 6H), 2.45 (d, J = 1.9 Hz, 3H) ppm; ¹³C NMR (75 MHz, DMSO-d₆ & CDCl₃): δ 162.2, 158.8, 158.4, 153.0 (d, J_C=CF = 14.8 Hz), 144.9, 140.4, 131.9, 127.0, 126.5, 125.7, 125.5, 116.1, 113.2, 112.9, 110.2 (d, J_C=CF = 11.5 Hz), 106.0, 103.2, 70.2, 50.6 (d, J_C=CF = 4.9 Hz), 11.9 ppm; ¹⁹F NMR (162 MHz, CDCl₃ & DMSO-d₆): δ 21.287 (m); IR(KBr): ν = 3443, 3252, 2953, 1659, 1521, 1208, 830 cm⁻¹; HRMS (ESI-TOF) m/z: [M + H]⁺ caged for C₂₃H₁₆F₂N₂O₂F₃H 413.1266, found, 413.1266.

N’-(4-Benzyl-5-phenylfuran-2-yl)(phenyl)methylene)-2-(prop-2-yn-1-yl)benzohydrazide (5). Following the general procedure, 100 mg (0.297 mmol, 1.0 equiv.) of 1a, 85 mg (0.446 mmol, 1.5 equiv.) of 7, 10 mg (10 mol%) of KAuCl₄ and 96 mg (0.595 mmol, 2.0 equiv.) of FeCl₃ was used and the reaction time was 12 h. After flash column chromatography on silica gel (eluted with Rf: 0.6; hexane/ethyl acetate mixture 10/3.0), 97 mg of 8 was obtained in 62% yield as a yellow solid. Mps: 73–75 °C; ¹H NMR (400 MHz, CDCl₃): δ 10.81 (s, 1H), 8.34 (dd, J = 7.8 Hz, 1.7 Hz, 1H), 7.83–7.78 (m, 2H), 7.75–7.70 (m, 2H), 7.69–7.62 (m, 3H), 7.55–7.41 (m, 4H), 7.39–7.33 (m, 2H), 7.30–7.26 (m, 3H), 7.18–7.11 (m, 1H), 7.01 (d, J = 8.3 Hz, 1H), 6.72 (s, 1H), 4.18 (d, J = 2.2 Hz, 2H), 2.50 (t, J = 2.2 Hz, 1H) ppm; ¹³C NMR (100 MHz, CDCl₃): δ 191.4, 161.2, 157.1, 154.9, 149.8, 145.0, 137.4, 133.2, 133.0, 131.4, 130.2, 129.69, 129.62, 129.4, 128.9, 128.6, 128.3, 128.2, 127.9, 122.5, 122.4, 120.8, 116.9,
Conflicts of interest

There are no conflicts to declare.

Acknowledgements

We thank Department of Science and Technology (DST) India grant no: DST-SB/EMEQ-257/2014 and CSIR, New Delhi for financial support. We are thankful to Director Dr S. Chandrasekhar CSIR-ICT for his support. We thank Dr S. Suresh for his suggestions on this work. We acknowledge our colleagues from CSIR-ICT Dr K. Srinivas and Dr Rajesh for their encouragement. V. K thanks to UGC-SRF, K. S thanks to Dr Ch. Raji Reddy and CSIR grant HCP-0011. P. B. K and C. E. R thanks to DST for Inspire fellowship and also thanks to AcSIR, manuscript communication number:ICT/pubs/2019/176.

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22 See the ESI for X-ray crystallographic data for compound 3a and 5.

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