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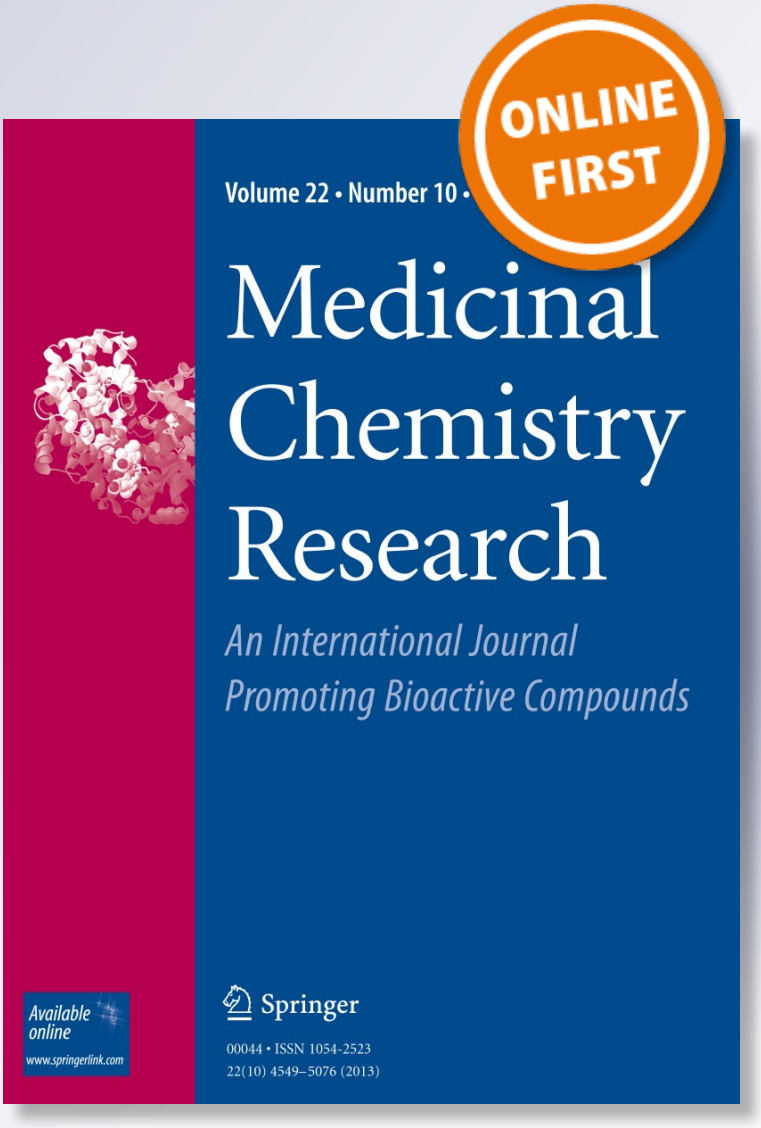


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ORIGINAL RESEARCH

# New 4-thiazolidinones from 5-ethyl pyridine-2-ethanol: their antibacterial, antifungal, and antitubercular activity

**Navin B. Patel · Hemant R. Patel · Faiyazalam M. Shaikh · Dhanji Rajani**

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**Abstract** New thiazolidinones **5a–o** were prepared from Schiff base **4a–o** and thioglycolic acid in the presence of $ZnCl_2$ from 4-[2-(5-ethylpyridin-2-yl)ethoxy]benzaldehyde. The structures of the synthesized compounds were assigned on the basis of elemental analysis, IR, $^{1}H$ NMR $^{13}C$ NMR, and mass spectral data. All the compounds were screened against different strains of bacteria and fungi. Compounds **4e**, **4n**, **4m**, **4o**, **5e**, **5f**, **5j**, and **5m** possessed very good activity against bacterial and fungal species. These active compounds impelled us to study their antitubercular activity. Schiff base **4n** and **4e** showed *M. tuberculosis* MIC value 25 and 62.5 μg/ml, respectively. Thiazolidinone **5m** displayed *M. tuberculosis* MIC at 25 μg/ml, which is better antitubercular activity compared with rifampicin.

**Keywords** Schiff base · Thiazolidinone · Antitubercular · Antibacterial · Antifungal

## Introduction

An extensive research work on thiazolidinones has been done in the past. Synthesis of novel thiazolidinones was opted for the research due to their commercial and medicinal importance. They have been synthesized by the condensation of amino and aldehyde group containing molecules via the intermediacy of a Schiff base, employing conventional methodologies. Schiff base also possessed various biological activities like antimicrobial, antibacterial, antifungal (Ronad *et al*., 2009; Ansari and Lal, 2009; Panneerselvam *et al*., 2009; Bharti *et al*., 2010), antitubercular (Raparti *et al*., 2009; Kamel *et al*., 2010), analgesic, and anti-inflammatory activity (Bhandari *et al*., 2008). The thiazolidione nucleus is well known as wonder nucleus with wide variety of biological activities such as antimicrobial(Vicini *et al*., 2006; Fuloria *et al*., 2009; Mulwad *et al*., 2009; Patel and Rathod, 2007; Patel and Patel, 2007; Patel and Patel, 2009a, b; Shah and Desai, 2007; Petrikaite *et al*., 2007; Sattigeri *et al*., 2005; Kavitha *et al*., 2006; Saeed *et al*., 2007; Sayyed *et al*., 2006), antitubercular, antiviral and anticancer activity(Tatar *et al*., 2008), anti-inflammatory, analgesic, CNS depressant, Muscle relaxant, and antipyretic activity (Kumar *et al*., 2007; Gurupadayya *et al*., 2008; Tarnalli *et al*., 2008; Ottana *et al*., 2005). They also possessed anti-HIV, antiproliferative, AT1 angiotensin-II (AII) receptor antagonists activity (Rawal *et al*., 2007; Kucukguzel *et al*., 2006; Shreenivas *et al*., 2009; Chandrappa *et al*., 2008).

The structure of pioglitazone HCl (Fig. 1) is well known for its anti-diabetic activity because of 1,3-thiazolidine-2,4-dione ring attached to the (4-[2-(5-ethylpyridin-2-yl)ethoxy] benzaldehyde). It is evident from the literature survey that very little work done on thiazolidinones from pioglitazone HCl or its intermediate. In continuation of our work on pyridine and thiazolidinones earlier, we have reported antimicrobial and antitubercular activity (Patel and Patel, 2009a, b, 2010a, b; Patel *et al*., 2010a, b; Patel and Shaikh, 2010).

In chemotherapeutic point of view, the design of new agents by molecular hybridization with multiple potential targets has always been a very attractive strategy for medicinal chemists (Barbosa *et al*., 2011). In a view of these data, we report in this paper the synthesis of

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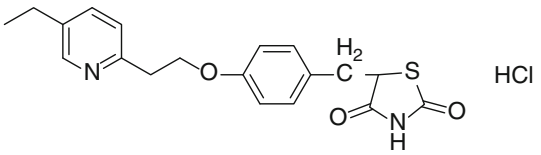


**Fig. 1** Pioglitazone HCl

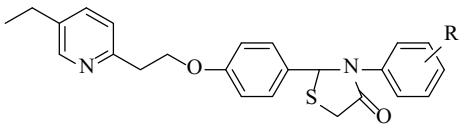


**Fig. 2** Final thiazolidinones

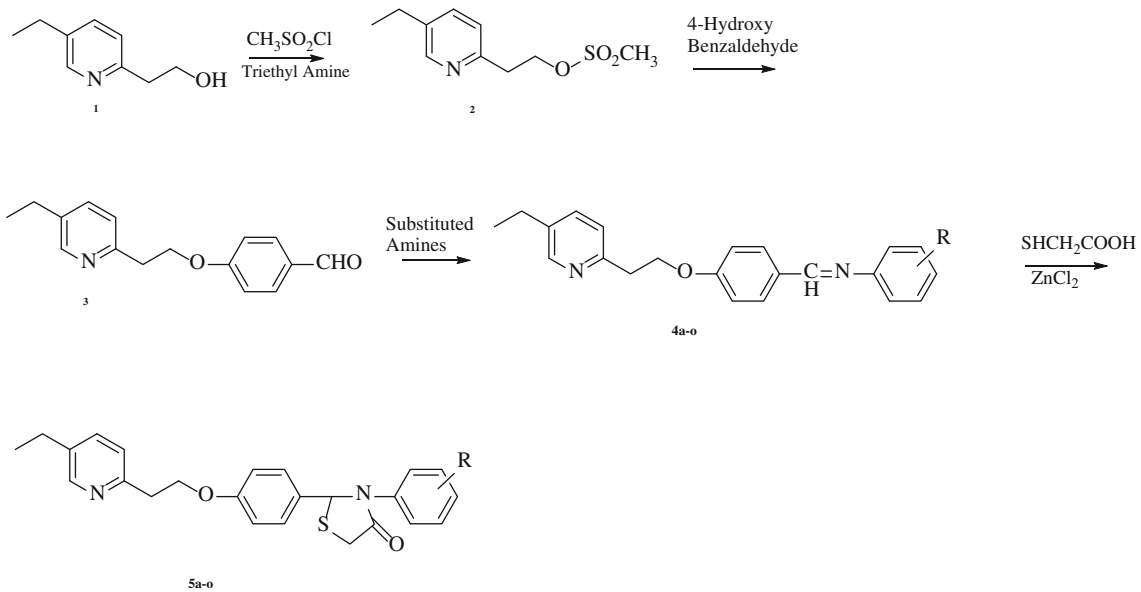
thiazolidinones (Fig. 2), cyclized from Schiff base of 4-[2-(5-ethylpyridin-2-yl)ethoxy]benzaldehyde and studied their antimicrobial and antitubercular activity.

## Results and discussion

### Chemistry

The synthesis of Schiff bases and thiazolidinones was performed shown in (Scheme 1). Schiff bases **4a**–**o** were synthesized by condensing 4-[2-(5-ethylpyridin-2-yl)ethoxy]benzaldehyde with appropriate aromatic amine in absolute alcohol and 2–3 drops of glacial acetic acid at refluxed temperature. The cyclic thiazolidinone **5a**–**o** was synthesized from appropriate Schiff base and thioglycolic acid in the presence of $ZnCl_2$. The purity of the compounds was checked by TLC and elemental analysis. Spectral data (IR, $^1$H NMR, $^{13}$C NMR and Mass) of all the compounds were in full agreement with the proposed structures.

The compounds **4a**–**o** were confirmed by the IR, NMR, and Mass spectra. In the IR spectrum, the band of –CH=N observed at 1,627 $cm^{-1}$, the asymmetric and symmetric band of C–O–C ether linkage observed at 1,227 and 1,046 $cm^{-1}$. A singlet observed at $\delta$ 8.25 attributed to the –CH=N protons and triplet at $\delta$ 4.30 confirmed the –$CH_2$–O–. The higher resonance at $\delta$ 160.8 ppm was attributed to the –CH group present in Schiff base. Molecular ion peak observed at *m*/*z* 345, confirmed its molecular weight of **4o**. Disappearance of –CH=N band at 1,627 $cm^{-1}$ and appearance of 1,760 $cm^{-1}$ of –C=O (thiazolidinone) was observed in IR, signal at $\delta$ 3.20 and $\delta$ 5.30 for the –$CH_2$– and –CH of cyclic thiazolidinone observed in $^1$H NMR, and –C=O carbon appeared at $\delta$ 170.1 was also observed in$^{13}$C NMR and molecular ion peak at *m*/*z* 419 of **5o** confirmed its molecular weight.



**R:** **a.** 4-F **b.** 3,5-F **c.** 4-Cl **d.** 3,4-Cl **e.** 2,4-Cl
**f.** 3,4-F **g.** 2-Br, 4-F **h.** 3-Cl, 4-$CH_3$ **i.** 3,5-Cl **j.** 2-Cl
**k.** 2-F **l.** 2,5-Cl **m.** 2,3-Cl **n.** 3-Cl **o.** 4-$CH_3$

**Scheme 1** Synthetic route for 4-thiazolidinones from 5-ethylpyridine-ethanol **5a**–**o**

## Antimicrobial activity

### Methods

All MTCC cultures were collected from Institute of Microbial Technology, Chandigarh and tested against known drugs ampicillin and griseofulvin. Mueller–Hinton broth was used as nutrient medium to grow and dilute the drug suspension for the test. Inoculum size for test strain was adjusted to $10^8$ CFU (Colony Forming Unit) per milliliter by comparing the turbidity. DMSO was used as diluents to get desired concentration of drugs to test upon standard bacterial strains. Serial dilutions were prepared in primary and secondary screening. The control tube containing no antibiotic was immediately sub cultured (before inoculation) by spreading a loopful evenly over a quarter of plate of medium suitable for the growth of the test organism and put for incubation at 37 °C overnight. The tubes were then incubated overnight. The MICs of compounds were carried out by broth microdilution method as described by Rattan (2000). Antibacterial activity was screened against two gram positive (*Staphylococcus aureus* MTCC 96, *Streptococcus pyogenus* MTCC 443) and two gram negative (*Escherichia coli* MTCC 442, *Pseudomonas aeruginosa* MTCC 2488) bacteria, ampicillin was used as a standard antibacterial agent. Antifungal activity was screened against three fungal species *Candida albicans* MTCC 227, *Aspergillus niger* MTCC 282 and *Aspergillus clavatus* MTCC 1323, griseofulvin was used as a standard antifungal agent.

### Antibacterial activity

The minimum Inhibitory concentrations (MIC) of the tested compounds **4a–o** and **5a–o** are shown in (Table 1). From the screening data, most of the compounds possessed very good antibacterial activity (MIC, 100–250 μg/ml) against *S. aureus*; some of them possessed better activity compared to ampicillin.

Schiff bases **4e** and **4n** containing mono and dichloro substituents possessed pronounced activity (100 μg/ml) against *E. coli.* Compound **4e** displayed better activity (100 μg/ml) against *P. aeruginosa* due to 2,4-dichloro substituent. Compounds **4g**, **4l**, **4m,** and **4o** containing 2-bromo-4-fluoro, 2,5-dichloro, 2,3-dichloro, and 4-methyl substituents exhibited significant activity in the range of (100–200 μg/ml) against *S. aureus* which indicates that these compounds are very effective toward the bacteria. Remaining compounds **4d**, **4e**, **4h**, **4k**, and **4n** having 3,4-dichloro, 2,4-dichloro, 3-chloro, 4-methyl, 2-fluoro, and 3-chloro substituents showed comparable to moderate activity (100–500 μg/ml) against reference drug. The thiazolidinones **5e**, **5m**, **5f**, and **5j** having 2,4-dichloro, 2,3-dichloro, 3,4-difluoro, and 2-chloro substituents showed higher activity (62.5–100 μg/ml) against *E. coli*. Compound **5e** having 2,4-dichloro substituent possessed better activity (100 μg/ml) against *P. aeruginosa.* Compounds **5b**, **5c**, **5d**, **5e**, **5f**, **5i**, **5j**, **5k**, **5l**, **5m**, and **5o** having monochloro, dichloro, fluoro, and methyl substituents displayed better activity (100–250 μg/ml) against *S. aureus.* Compound 5m having 2,3-dichloro substituent is found to be very active (MIC value of 100 μg/ml) against *S. pyogenus.* Remaining compounds possessed moderate-to-weak activity against all four bacterial species.

**Table 1** Antibacterial activity of **4a–o** and **5a–o**

| Compounds | Minimal bactericidal concentration (μg/ml) | | | |
|---|---|---|---|---|
| | Gram negative | | Gram positive | |
| | *E. coli* | *P. aeruginosa* | *S. aureus* | *S. pyogenus* |
| **4a** | 250 | 500 | 500 | 500 |
| **4b** | 500 | 500 | 1,000 | 1,000 |
| **4c** | 1,000 | 1,000 | 1,000 | 1,000 |
| **4d** | 200 | 250 | 250 | 250 |
| **4e** | 100 | 100 | 250 | 500 |
| **4f** | 250 | 250 | 500 | 500 |
| **4g** | 250 | 250 | 200 | 250 |
| **4h** | 500 | 500 | 250 | 500 |
| **4i** | 1,000 | 1,000 | 500 | 500 |
| **4j** | 500 | 500 | 500 | 500 |
| **4k** | 250 | 200 | 250 | 500 |
| **4l** | 500 | 250 | 150 | 200 |
| **4m** | 250 | 200 | 100 | 250 |
| **4n** | 100 | 150 | 250 | 250 |
| **4o** | 150 | 200 | 100 | 250 |
| **5a** | 250 | 250 | 500 | 500 |
| **5b** | 200 | 150 | 200 | 500 |
| **5c** | 250 | 500 | 250 | 500 |
| **5d** | 500 | 500 | 250 | 250 |
| **5e** | 62.5 | 100 | 200 | 250 |
| **5f** | 100 | 150 | 150 | 200 |
| **5g** | 250 | 500 | 500 | 500 |
| **5h** | 250 | 500 | 500 | 500 |
| **5i** | 500 | 500 | 150 | 200 |
| **5j** | 100 | 200 | 250 | 250 |
| **5k** | 150 | 250 | 250 | 250 |
| **5l** | 250 | 250 | 200 | 200 |
| **5m** | 62.5 | 200 | 100 | 100 |
| **5n** | 500 | 500 | 500 | 500 |
| **5o** | 500 | 500 | 250 | 250 |
| Ampicillin | 100 | 100 | 250 | 100 |

### Antifungal activity

MIC of compounds **4a–o** and **5a–o** are summarized in (Table 2). Most of the compounds possessed very good

**Table 2** Antifungal activity of **4a–o** and **5a–o**

| Compounds | Minimal fungicidal concentration (μg/ml) | | |
|---|---|---|---|
| | *C. albicans* | *A. niger* | *A. clavatus* |
| **4a** | 500 | >1,000 | >1,000 |
| **4b** | >1,000 | 1,000 | 1,000 |
| **4c** | >1,000 | >1,000 | >1,000 |
| **4d** | 200 | 500 | 500 |
| **4e** | 250 | 500 | 500 |
| **4f** | 500 | >1,000 | >1,000 |
| **4g** | 500 | 500 | 1,000 |
| **4h** | 500 | 1,000 | 1,000 |
| **4i** | >1,000 | >1,000 | >1,000 |
| **4j** | 1,000 | 1,000 | 1,000 |
| **4k** | 500 | 500 | 500 |
| **4l** | 500 | 1,000 | 1,000 |
| **4m** | 500 | 1,000 | >1,000 |
| **4n** | 1,000 | 1,000 | 1,000 |
| **4o** | >1,000 | >1,000 | >1,000 |
| **5a** | 1,000 | 500 | 500 |
| **5b** | 1,000 | 1,000 | 1,000 |
| **5c** | 500 | 1,000 | 1,000 |
| **5d** | 1,000 | 500 | 500 |
| **5e** | 250 | 250 | 500 |
| **5f** | 1,000 | >1,000 | >1,000 |
| **5g** | 500 | 500 | 500 |
| **5h** | 1,000 | 1,000 | >1,000 |
| **5i** | 500 | 1,000 | 1,000 |
| **5j** | 1,000 | 1,000 | >1,000 |
| **5k** | 100 | 250 | 250 |
| **5l** | 1,000 | 1,000 | 1,000 |
| **5m** | 500 | 500 | 500 |
| **5n** | 1,000 | 1,000 | >1,000 |
| **5o** | 1000 | 500 | 500 |
| Nystatin | 100 | 100 | 100 |
| Greseofulvin | 500 | 100 | 100 |

antifungal activity against *C. albicans*; their MIC values were in the range between 100 and 500 μg/ml.

Schiff base **4a**, **4d**, **4e**, **4f**, **4g**, **4h**, **4k**, **4l**, and **4m** possessed better activity of (200–500 μg/ml) against *C. albicans* which is due to 4-fluoro, 3,4-dichloro, 2,4-dichloro, 3,4-difluoro, 2-bromo-4-fluoro, 3-chloro-4-methyl, 2,5-chloro, and 2,3-dichloro substituents, whereas remaining Schiff bases possessed weak activity against *A. niger* and *A. clavatus*. Thiazolidinones **5c**, **5e**, **5g**, **5i**, and **5m** containing 4-chloro, 2,4-dichloro, 2-bromo-4-fluoro, 3,5-dichloro, and 2,3-dichloro substituents possessed good activity of (250–500 μg/ml) against *C. albicans.* Compound **5k** having 2-fluoro substituent showed significant activity (100 μg/ml) against nysatin, whereas remaining compounds possessed weak activity against *A. niger* and *A. clavatus*.

**Table 3** Antimycobacterial activity of **4a–o** and **5a–o**

| Compounds | MIC values (μg/ml) of *M. tuberculosis* $H_{37}Rv$ | % Inhibition |
|---|---|---|
| **4e** | 62.5 | 98 |
| **4n** | 25 | 95 |
| **4m** | 500 | 97 |
| **4o** | 125 | 95 |
| **5e** | >1,000 | 95 |
| **5f** | 250 | 96 |
| **5j** | 100 | 98 |
| **5m** | 25 | 99 |
| Rifampicin | 40 | 99 |

## Antitubercular activity

The encouraging results from the antibacterial studies impelled us to go for preliminary screening against *M. tuberculosis*; the results are summarized in (Table 3). In this screening, 1,000, 500, and 250 μg/ml concentrations of the compounds were taken. The active compounds found in this screening were further tested in a secondary screening against *M. tuberculosis* $H_{37}Rv$ in L. J. Medium (conventional method). The compounds found active in primary screening were similarly diluted to obtain 200, 100, 50, 25, 12.5, 6.250, and 3.50 μg/ml concentrations. The antitubercular activity data were compared with rifampicin at a 40 μg/ml concentration. Schiff base, **4n** having 3-chloro group showed *M. tuberculosis* MIC value 25 μg/ml which is very active as rifampicin which have standard concentration 40 μg/ml. Compounds **4e** containing 2,4-dichloro substituent showed *M. tuberculosis* MIC value in the range between 62.5 μg/ml. Thaizolidinone **5m** having 2,3-dichloro substituent displayed MIC value 25 μg/ml which indicate better activity. The highest dilution showing at least 95–99 % inhibition growth is taken as MIC. Remaining compounds showed moderate-to-weak activity against $H_{37}Rv$ strain.

## Conclusions

Compounds 5e and 5m possessed excellent activity of 62.5 μg/ml comparable to ampicillin. Compound **5k** showed remarkable activity of 100 μg/ml comparable to nystatin. Compound bearing fluoro, chloro, and disubstituted chloro are more effective to inhibit the both bacterial and fungal species. Compound **4n** and **5m** having chloro and dichloro substituents exhibited *M. tuberculosis* MIC

value 25 μg/ml comparable with rifampicin. From this observations, we can conclude that halogen groups can impart a positive effect for biological activity i.e., activity increasing effect. Present work will be useful for understanding antimicrobial and antitubercular activity of hybrids of thiazolidinones and pyridine.

## Experimental

Laboratory Chemicals were supplied by Rankem India Ltd. and Ficher Scientific Ltd. Melting points were determined by the open tube capillary method and are uncorrected. The purity of the compounds was determined by thin layer chromatography (TLC) plates (silica gel G) in the solvent system toluene:ethyl acetate (7.5:2.5). The spots were observed by exposure to iodine vapors or by UV light. The IR spectra were obtained on a Perkin–Elmer 1720 FT-IR spectrometer (KBr pellets). The $^{1}$HNMR and $^{13}$CNMR spectra were recorded on a Bruker Avance II 400 spectrometer using TMS as the internal standard in $CDCl_3$. Elemental analysis of the newly synthesized compounds were carried out on Carlo Erba 1108 analyzer. The mass spectra were recorded on micromass Q-T of micro (TOF MS ES$^{+}$).

Procedure for the synthesis of 4-[2-(5-ethylpyridin-2-yl)ethoxy]benzaldehyde (**3**)

4-[2-(5-Ethylpyridin-2-yl)ethoxy]benzaldehyde (3) was synthesized by the method described in the literature (Gaonkar *et al*., 2006).

General preparation of the compounds (**4a**–**o**)

A mixture of 4-[2-(5-ethylpyridin-2-yl)ethoxy]benzaldehyde (0.01 mol) and aromatic amine (0.01 mol) was taken in absolute ethanol and few drops of glacial acetic acid were added. Then, the mixture was refluxed for 6–8 h on water bath. The completion of reaction was monitored by TLC (toluene:ethyl acetate, 7.5:2.5). The excess solvent was distilled off, and then remaining residue poured into ice cold water. The separated solid was filtered, washed, and recrystalized from ethanol to give product **4a**–**o** (Fig. 3).

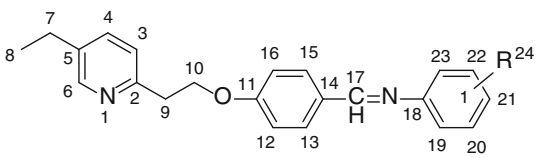



**Fig. 3** Schiff base **4a**–**o**

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-4-fluorobenzenamine (**4a**)*

This compound was obtained as off white solid, yield: 70 %, M.P. 85–87 °C, $R_f$: 0.45; IR (KBr): 3055 (Ar–H), 2956, 2844 (–$CH_2$–), 1628 (–CH=N), 1227, 1045 (C–O–C), 972 (C–F); $^{1}$H NMR ($CDCl_3$): $\delta$ 1.12 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.50 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.17 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.32 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.00–7.87 (m, 8H, Ar–H), 7.34–8.34 (m, 3H, Pyridine-H), 8.27 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.4 ($C_8$), 32.5 ($C_7$), 37.5 ($C_9$), 67.6 ($C_{10}$), 115.2–155.4 ($C_{11}$–$C_{16}$), 116.5–161.4 ($C_{18}$–$C_{23}$), 123.3–157.3 ($C_2$–$C_6$), 160.6 ($C_{17}$); MS (*m/z*): 348 ($M^+$); Anal. Calcd. for $C_{22}H_{21}N_2OF$: C, 75.84; H, 6.08; N, 8.04; Found C, 75.82; H, 6.09; N, 8.05.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-3,5-difluorobenzenamine (**4b**)*

This compound was obtained as off white solid, yield: 72 %, M.P. 100–103 °C, $R_f$: 0.43; IR (KBr): 3058 (Ar–H), 2958, 2845 (–$CH_2$–), 1627 (–CH=N), 1228, 1046 (C–O–C), 974 (C–F); $^{1}$H NMR ($CDCl_3$): $\delta$ 1.13 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.52 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.15 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.33 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 6.72–7.85 (m, 7H, Ar–H), 7.34–8.36 (m, 3H, Pyridine-H), 8.26 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.7 ($C_8$), 32.6 ($C_7$), 37.5 ($C_9$), 67.3 ($C_{10}$), 115.2–155.4 ($C_{11}$–$C_{16}$),123.7–151.3 ($C_{18}$–$C_{23}$), 123.2–157.6 ($C_2$–$C_6$), 160.7 ($C_{17}$); MS (*m/z*): 366 ($M^+$); Anal. Calcd. for $C_{22}H_{20}N_2OF_2$: C, 72.12; H, 5.50; N, 7.65; Found C, 72.13; H, 5.51; N, 7.62.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-4-chlorobenzenamine (**4c**)*

This compound was obtained as off yellow solid, yield: 80 %, M.P. 113–115 °C, $R_f$: 0.47; IR (KBr): 3056 (Ar–H), 2960, 2844 (–$CH_2$–), 1626 (–CH=N), 1227, 1044 (C–O–C), 745 (C–Cl); $^{1}$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.54 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.16 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.30 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.05–7.87 (m, 8H, Ar–H), 7.35–8.35 (m, 3H, Pyridine-H), 8.28 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.7 ($C_8$), 32.6 ($C_7$), 37.5 ($C_9$), 67.3 ($C_{10}$), 115.2–155.4 ($C_{11}$–$C_{16}$),123.7–151.3 ($C_{18}$–$C_{23}$), 123.2–157.6 ($C_2$–$C_6$), 160.7 ($C_{17}$); MS (*m/z*): 364 ($M^+$), 366 (M+2); Anal. Calcd. for $C_{22}H_{21}N_2OCl$: C, 72.42; H, 5.80; N, 7.68; Found C, 72.41; H, 5.79; N, 7.66.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)- 3,4-dichlorobenzenamine (**4d**)*

This compound was obtained as off yellow liquid, yield: 78 %, M.P. semi solid, $R_f$: 0.43; IR (KBr): 3057 (Ar–H),

2957, 2845 (–$CH_2$–), 1629 (–CH=N), 1228, 1046 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.14 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.56 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.13 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.33 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 7.09–7.86 (m, 7H, Ar–H), 7.33–8.35 (m, 3H, Pyridine-H), 8.25 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.6 ($C_8$), 32.5 ($C_7$), 37.7 ($C_9$), 67.4 ($C_{10}$), 115.4–155.6 ($C_{11}$–$C_{16}$),121.8–152.6 ($C_{18}$–$C_{23}$), 123.4–157.4 ($C_2$–$C_6$), 160.5 ($C_{17}$); MS (*m/z*): 398 ($M^+$), 400 (M+2), 402 (M+4); Anal. Calcd. for $C_{22}H_{20}N_2OCl_2$: C, 66.17; H, 5.05; N, 7.02; Found C, 66.15; H, 5.04; N, 7.00.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-2,4-dichlorobenzenamine (**4e**)*

This compound was obtained as off yellow liquid, yield: 75 %, M.P. semi solid, $R_f$: 0.50; IR (KBr): 3056 (Ar–H), 2957, 2847 (–$CH_2$–), 1624 (–CH=N), 1227, 1043 (C–O–C), 745 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.13 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.54 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.15 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.35 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 6.70–7.88 (m, 7H, Ar–H), 7.34–8.35 (m, 3H, Pyridine-H), 8.26 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.4 ($C_8$), 32.6 ($C_7$), 37.5 ($C_9$), 67.5 ($C_{10}$), 115.3–155.7 ($C_{11}$–$C_{16}$),125.1–141.3 ($C_{18}$–$C_{23}$), 123.5–157.3 ($C_2$–$C_6$), 160.4 ($C_{17}$); MS (*m/z*): 398 ($M^+$), 400 (M+2), 402 (M+4); Anal. Calcd. for $C_{22}H_{20}N_2OCl_2$: C, 66.17; H, 5.05; N, 7.02; Found C, 66.16; H, 5.06; N, 7.01.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-3,4-difluorobenzenamine (**4f**)*

This compound was obtained as yellow solid, yield: 68 %, M.P. 120–122 °C, $R_f$: 0.51; IR (KBr): 3059 (Ar–H), 2954, 2845 (–$CH_2$–), 1626 (–CH=N), 1226, 1045 (C–O–C), 973 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.16 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.53 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.13 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.36 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 6.89–7.85 (m, 7H, Ar–H), 7.34–8.37 (m, 3H, Pyridine-H), 8.25 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.7 ($C_8$), 32.4 ($C_7$), 37.6 ($C_9$), 67.6 ($C_{10}$), 115.0–155.5 ($C_{11}$–$C_{16}$),111.2–150.3 ($C_{18}$–$C_{23}$), 123.1–157.4 ($C_2$–$C_6$), 160.8 ($C_{17}$); MS (*m/z*): 366 ($M^+$); Anal. Calcd. for $C_{22}H_{20}N_2OF_2$: C, 72.12; H, 5.50; N, 7.65; Found C, 72.11; H, 5.51; N, 7.64.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)2-bromo-4-fluorobenzenamine (**4g**)*

This compound was obtained as off brown liquid, yield: 59 %, M.P. semi solid, $R_f$: 0.46; IR (KBr): 3056 (Ar–H), 2955, 2844 (–$CH_2$–), 1627 (–CH=N), 1225, 1046 (C–O–C), 972 (C–F), 857 (C–Br); $^1$H NMR ($CDCl_3$): $\delta$ 1.14 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.57 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.16 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.34 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 6.89–7.86 (m, 7H, Ar–H), 7.30–8.38 (m, 3H, Pyridine-H), 8.27 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.8 ($C_8$), 32.5 ($C_7$), 37.4 ($C_9$), 67.7 ($C_{10}$), 115.2–155.3 ($C_{11}$–$C_{16}$),112.6–163.5 ($C_{18}$–$C_{23}$), 123.2–157.6 ($C_2$–$C_6$), 160.7 ($C_{17}$); MS (*m/z*): 426 ($M^+$), 428 (M+2); Anal. Calcd. for $C_{22}H_{20}N_2OFBr$: C, 61.84; H, 4.72; N, 6.56; Found C, 61.80; H, 4.71; N, 6.53.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)3-chloro-4-methyl benzenamine (**4h**)*

This compound was obtained as off brown solid, yield: 62 %, M.P. 135–137 °C, $R_f$: 0.48; IR (KBr): 3058 (Ar–H), 2958, 2845 (–$CH_2$–), 1628 (–CH=N), 1228, 1046 (C–O–C), 744 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.18 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.35 (s, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.58 (q, $J$ = 7.0 Hz, 2H, –$CH_2$), 3.15 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.33 (t, 2H, –$CH_2$–O), 7.00–7.82 (m, 7H, Ar–H), 7.35–8.36 (m, 3H, Pyridine-H), 8.24 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.6 ($C_8$), 20.1 ($C_{24}$), 32.6 ($C_7$), 37.6 ($C_9$), 67.5 ($C_{10}$), 115.8–155.4 ($C_{11}$–$C_{16}$),120.0–151.5 ($C_{18}$–$C_{23}$), 123.4–157.8 ($C_2$–$C_6$), 160.3 ($C_{17}$); MS (*m/z*): 378 ($M^+$), 380 (M+2); Anal. Calcd. for $C_{23}H_{23}N_2OCl$: C, 72.91; H, 6.12; N, 7.39; Found C, 72.90; H, 6.10; N, 7.36.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)- 3,5-dichlorobenzenamine (**4i**)*

This compound was obtained as off yellow solid, yield: 77 %, M.P. 110–112 °C, $R_f$: 0.43; IR (KBr): 3056 (Ar–H), 2956, 2843 (–$CH_2$–), 1627 (–CH=N), 1224, 1045 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.17 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.58 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.15 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.33 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 7.05–7.84 (m, 7H, Ar–H), 7.35–8.36 (m, 3H, Pyridine-H), 8.24 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.6 ($C_8$), 32.6 ($C_7$), 37.6 ($C_9$), 67.5 ($C_{10}$), 115.8–155.4 ($C_{11}$–$C_{16}$), 120.5–156.1 ($C_{18}$–$C_{23}$), 123.4–157.6 ($C_2$–$C_6$), 160.5 ($C_{17}$); MS (*m/z*): 398 ($M^+$), 400 (M+2), 402 (M+4); Anal. Calcd. for $C_{22}H_{20}N_2OCl_2$: C, 66.17; H, 5.05; N, 7.02; Found C, 66.18; H, 5.04; N, 7.04.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-2-chlorobenzenamine (**4j**)*

This compound was obtained as brown liquid, yield: 78 %, M.P. semi solid, $R_f$: 0.44; IR (KBr): 3057 (Ar–H), 2957, 2846 (–$CH_2$–), 1627 (–CH=N), 1229, 1047 (C–O–C), 745 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J$ = 8.0 Hz, 3H, –$CH_3$), 2.56 (q, $J$ = 8.0 Hz, 2H, –$CH_2$), 3.14 (t, $J$ = 7.0 Hz, 2H, –$CH_2$), 4.34 (t, $J$ = 7.0 Hz, 2H, –$CH_2$–O), 7.08–7.82 (m, 8H, Ar–H), 7.34–8.37 (m, 3H, Pyridine-H), 8.22 (s, 1H,

–CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.4 ($C_8$),), 32.5 ($C_7$), 37.7 ($C_9$), 67.6 ($C_{10}$), 115.5–155.3 ($C_{11}$–$C_{16}$),123.–143.4 ($C_{18}$–$C_{23}$), 123.5–157.3 ($C_2$–$C_6$), 160.2 ($C_{17}$); MS (*m/z*): 364 ($M^+$), 366 (M+2); Anal. Calcd. for $C_{22}H_{21}N_2OCl$: C, 72.42; H, 5.80; N, 7.68; Found C, 72.44; H, 5.81; N, 7.67.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-2-fluorobenzenamine (**4k**)*

This compound was obtained as brown liquid, yield, 69 %, M.P. semi solid, $R_f$: 0.47; IR (KBr): 3056 (Ar–H), 2958, 2845 (–$CH_2$–), 1627 (–CH=N), 1227, 1048 (C–O–C), 975 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.16 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.57 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.16 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.36 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.00–7.85 (m, 8H, Ar–H), 7.35–8.38 (m, 3H, Pyridine-H), 8.24 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.1 ($C_8$), 32.5 ($C_7$), 37.2 ($C_9$), 67.5 ($C_{10}$), 115.4–155.4 ($C_{11}$–$C_{16}$),116.8–153.2 ($C_{18}$–$C_{23}$), 123.4–157.4 ($C_2$–$C_6$), 160.3 ($C_{17}$); MS (*m/z*): 348 ($M^+$); Anal. Calcd. for $C_{22}H_{21}N_2OF$: C, 75.84; H, 6.08; N, 8.04; Found C, 75.82; H, 6.06; N, 8.03.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-2,5-dichlorobenzenamine (**4l**)*

This compound was obtained as brown liquid, yield: 75 %, M.P. semi solid, $R_f$: 0.42; IR (KBr): 3059 (Ar–H), 2955, 2845 (–$CH_2$–), 1629 (–CH=N), 1225, 1049 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.19 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.53 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.17 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.37 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.06–7.88 (m, 7H, Ar–H), 7.33–8.36 (m, 3H, Pyridine-H), 8.26 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.3 ($C_8$), 32.6 ($C_7$), 37.4 ($C_9$), 67.6 ($C_{10}$), 115.5–155.6 ($C_{11}$–$C_{16}$),124.0–144.6 ($C_{18}$–$C_{23}$), 123.5–157.7 ($C_2$–$C_6$), 160.7 ($C_{17}$); MS (*m/z*): 398 ($M^+$), 400 (M+2), 402 (M+4); Anal. Calcd. for $C_{22}H_{20}N_2OCl_2$: C, 66.17; H, 5.05; N, 7.02; Found C, 66.15; H, 5.03; N, 7.04.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-2,3-dichlorobenzenamine (**4m**)*

This compound was obtained as brown liquid, yield: 71 %, M.P. semi solid, $R_f$: 0.43; IR (KBr): 3056 (Ar–H), 2957, 2845 (–$CH_2$–), 1627 (–CH=N), 1227, 1046 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.20 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.58 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.15 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.35 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.07–7.83 (m, 7H, Ar–H), 7.32–8.36 (m, 3H, Pyridine-H), 8.28 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.4 ($C_8$), 32.4 ($C_7$), 37.7 ($C_9$), 67.5 ($C_{10}$), 115.2–155.4 ($C_{11}$–$C_{16}$),121.8–144.6 ($C_{18}$–$C_{23}$), 123.2–157.4 ($C_2$–$C_6$), 160.5 ($C_{17}$); MS (*m/z*): 398 ($M^+$), 400 (M+2), 402 (M+4); Anal. Calcd. for $C_{22}H_{20}N_2OCl_2$: C, 66.17; H, 5.05; N, 7.02; Found C, 66.16; H, 5.04; N, 7.03.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-3-chlorobenzenamine (**4n**)*

This compound was obtained as brown liquid, yield: 76 %, M.P. semi solid, $R_f$: 0.51; IR (KBr): 3056 (Ar–H), 2956, 2846 (–$CH_2$–), 1626 (–CH=N), 1226, 1046 (C–O–C), 744 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.24 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.52 (q, $J = 8.0$ Hz, 2H, –$CH_2$), 3.15 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.32 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 7.03–8.23 (m, 8H, Ar–H), 7.33–8.34 (m, 3H, Pyridine-H), 8.27 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.7 ($C_8$), 32.5 ($C_7$), 37.6 ($C_9$), 67.6 ($C_{10}$), 115.3–155.7 ($C_{11}$–$C_{16}$),120.4–154.6 ($C_{18}$–$C_{23}$), 123.5–157.5 ($C_2$–$C_6$), 160.6 ($C_{17}$); MS (*m/z*): 364 ($M^+$), 366 (M+2); Anal. Calcd. for $C_{22}H_{21}N_2OCl$: C, 72.42; H, 5.80; N, 7.68; Found C, 72.43; H, 5.82; N, 7.69.

*N-(4-(2-(5-Ethylpyridin-2-yl)ethoxy)benzylidene)-4-methylbenzenamine (**4o**)*

This compound was obtained as off white solid, yield: 70 %, M.P. 80–83 °C, $R_f$: 0.52; IR (KBr): 3056 (Ar–H), 2957, 2845 (–$CH_2$–), 1627 (–CH=N), 1227, 1046 (C–O–C); $^1$H NMR ($CDCl_3$): $\delta$ 1.13 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.24 (s, $J = 8.0$ Hz, 3H, –$CH_3$), 2.52 (q, $J = 7.0$ Hz, 2H, –$CH_2$), 3.17 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.29 (t, 2H, –$CH_2$–O), 6.85–7.69 (m, 8H, Ar–H), 7.34–8.30 (m, 3H, Pyridine-H), 8.25 (s, 1H, –CH=N–); $^{13}$C NMR ($CDCl_3$): $\delta$ 15.4 ($C_8$), 25.7 ($C_{24}$), 32.4 ($C_7$), 37.4 ($C_9$), 67.4 ($C_{10}$), 115.2–155.3 ($C_{11}$–$C_{16}$),120.8–148.9 ($C_{18}$–$C_{23}$), 123.4–158.9 ($C_2$–$C_6$), 161.4 ($C_{17}$); MS (*m/z*): 345 ($M^+$); Anal. Calcd. for $C_{23}H_{24}N_2O$: C, 80.20; H, 7.02; N, 8.13; Found C, 80.22; H, 7.03; N, 8.11.

General preparation of the compounds (**5a–o**)

A mixture of compound **4a–o** (0.01 mol) and thioglycolic acid (0.02 mol) in the presence of $ZnCl_2$ and solvent 1,4-dioxane was refluxed for 12–16 h. The completion of reaction was monitored by TLC (toluene:ethyl acetate, 7.5:2.5) and reaction mass was dumped in ice cold water. The resulting product was treated with 5 % $NaHCO_3$ solution to remove unreacted traces of thioglycolic acid.

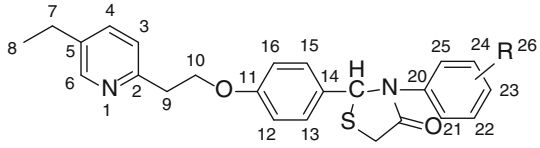



**Fig. 4** Thiazolidinones **5a–o**

The separated solid was washed with water, dried and recrystallized from ethanol to give product **5a**–**o** (Fig. 4).

*3-(4-Fluorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5a**)*

This compound was obtained as off yellow solid, yield: 65 %, M.P. 110–112 °C, $R_f$: 0.65; IR (KBr): 3031 (Ar–H), 2955, 2834 (–$CH_2$–), 1715 (–C=O of thiazolidinone), 1223, 1044 (C–O–C), 975 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.17 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.54 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.22 (s, 2H, –$CH_2$) 3.53 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.20 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.32 (s, 1H, –CH), 6.65–7.83 (m, 8H, Ar–H), 7.34–8.34 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.5 ($C_8$), 32.3 ($C_7$), 33.4 ($C_{18}$), 36.5 ($C_9$), 65.6 ($C_{17}$), 68.4 ($C_{10}$), 114.5–156.8 ($C_{11}$–$C_{16}$), 115.7–158.5 ($C_{20}$–$C_{25}$), 123.4–157.5 ($C_2$–$C_6$), 170.4 ($C_{19}$); MS (*m/z*): 422 ($M^+$); Anal. Calcd. for $C_{24}H_{23}N_2O_2FS$: C, 68.22; H, 5.49; N, 6.63; Found C, 68.21; H, 5.51; N, 6.64.

*3-(3,5-Difluorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5b**)*

This compound was obtained as yellow solid, yield: 67 %, M.P. 95–97 °C, $R_f$: 0.69; IR (KBr): 3034 (Ar–H), 2950, 2832 (–$CH_2$–), 1717 (–C=O of thiazolidinone), 1220, 1041 (C–O–C), 973 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.14 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.52 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.21 (s, 2H, –$CH_2$) 3.52 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.23 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.34 (s, 1H, –CH), 6.58–7.84 (m, 7H, Ar–H), 7.35–8.32 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.2 ($C_8$), 32.4 ($C_7$), 33.6 ($C_{18}$), 36.6 ($C_9$), 65.3 ($C_{17}$), 68.5 ($C_{10}$), 114.2–156.3 ($C_{11}$–$C_{16}$), 100.3–164.7 ($C_{20}$–$C_{25}$), 123.6–157.8 ($C_2$–$C_6$), 170.6 ($C_{19}$); MS (*m/z*): 440 ($M^+$); Anal. Calcd. for $C_{24}H_{22}N_2O_2F_2S$: C, 65.44; H, 5.03; N, 6.36; Found C, 65.42; H, 5.02; N, 6.37.

*3-(4-Chlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5c**)*

This compound was obtained as off brown solid, yield: 69 %, M.P. 125–128 °C, $R_f$: 0.66; IR (KBr): 3038 (Ar–H), 2952, 2834 (–$CH_2$–), 1719 (–C=O of thiazolidinone), 1222, 1043 (C–O–C), 743 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.18 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.54 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.24 (s, 2H, –$CH_2$) 3.53 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.35 (s, 1H, –CH), 6.65–7.84 (m, 8H, Ar–H), 7.38–8.37 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.3 ($C_8$), 32.5 ($C_7$), 33.3 ($C_{18}$), 36.5 ($C_9$), 65.6 ($C_{17}$), 68.7 ($C_{10}$), 114.5–156.6 ($C_{11}$–$C_{16}$), 123.0–139.7 ($C_{20}$–$C_{25}$), 123.5–157.9 ($C_2$–$C_6$), 170.4 ($C_{19}$); MS (*m/z*): 438 ($M^+$), 440 (M+2); Anal. Calcd. for $C_{24}H_{23}N_2O_2ClS$: C, 65.67; H, 5.28; N, 6.38; Found C, 65.65; H, 5.29; N, 6.36.

*3-(3,4-Dichlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5d**)*

This compound was obtained as brown solid, yield: 68 %, M.P. 140–142 °C, $R_f$: 0.65; IR (KBr): 3040 (Ar–H), 2951, 2833 (–$CH_2$–), 1714 (–C=O of thiazolidinone), 1226, 1046 (C–O–C), 745 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.56 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.23 (s, 2H, –$CH_2$) 3.56 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.27 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.31 (s, 1H, –CH), 6.60–7.80 (m, 7H, Ar–H), 7.37–8.36 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.6 ($C_8$), 32.7 ($C_7$), 33.4 ($C_{18}$), 36.3 ($C_9$), 65.2 ($C_{17}$), 68.5 ($C_{10}$), 114.1–156.2 ($C_{11}$–$C_{16}$), 121.0–141.2 ($C_{20}$–$C_{25}$), 123.3–157.7 ($C_2$–$C_6$), 170.2 ($C_{19}$); MS (*m/z*): 472 ($M^+$), 474 (M+2), 476 (M+4); Anal. Calcd. for $C_{24}H_{22}N_2O_2Cl_2S$: C, 60.89; H, 4.68; N, 5.92; Found C, 60.88; H, 4.69; N, 5.93.

*3-(2,4-Dichlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5e**)*

This compound was obtained as brown solid, yield: 66 %, M.P. 145–146 °C, $R_f$: 0.68; IR (KBr): 3038 (Ar–H), 2952, 2834 (–$CH_2$–), 1716 (–C=O of thiazolidinone), 1225, 1045 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.17 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.54 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.24 (s, 2H, –$CH_2$) 3.55 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.32 (s, 1H, –CH), 6.62–7.86 (m, 7H, Ar–H), 7.36–8.37 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.8 ($C_8$), 32.4 ($C_7$), 33.6 ($C_{18}$), 36.4 ($C_9$), 65.3 ($C_{17}$), 68.4 ($C_{10}$), 114.3–156.4 ($C_{11}$–$C_{16}$), 124.4–138.2 ($C_{20}$–$C_{25}$), 123.7–157.8 ($C_2$–$C_6$), 170.3 ($C_{19}$); MS (*m/z*): 472 ($M^+$), 474 (M+2), 476 (M+4); Anal. Calcd. for $C_{24}H_{22}N_2O_2Cl_2S$: C, 60.89; H, 4.68; N, 5.92; Found C, 60.87; H, 4.70; N, 5.93.

*3-(3,4-Difluorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5f**)*

This compound was obtained as off yellow solid, yield: 70 %, M.P. 170–173 °C, $R_f$: 0.67; IR (KBr): 3033 (Ar–H), 2953, 2835 (–$CH_2$–), 1713 (–C=O of thiazolidinone), 1223, 1046 (C–O–C), 977 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.19 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.55 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.26 (s, 2H, –$CH_2$) 3.56 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.24 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.34 (s, 1H, –CH), 6.67–7.74 (m, 7H, Ar–H), 7.38–8.38 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.5 ($C_8$), 32.6 ($C_7$), 33.3 ($C_{18}$), 36.7 ($C_9$), 65.6 ($C_{17}$), 68.5 ($C_{10}$), 114.3–156.7 ($C_{11}$–$C_{16}$), 111.7–149.7 ($C_{20}$–$C_{25}$), 123.6–157.5 ($C_2$–$C_6$), 170.6 ($C_{19}$);

MS (*m/z*): 440 ($M^+$); Anal. Calcd. for $C_{24}H_{22}N_2O_2F_2S$: C, 65.44; H, 5.03; N, 6.36; Found C, 65.45; H, 5.01; N, 6.37.

*3-(2-Bromo-4-fluorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5g**)*

This compound was obtained as off brown solid, yield: 56 %, M.P. 186–187 °C, $R_f$: 0.67; IR (KBr): 3037 (Ar–H), 2955, 2832 (–$CH_2$–), 1712 (–C=O of thiazolidinone), 1228, 1046 (C–O–C), 855 (C–Br) 975 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.56 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.25 (s, 2H, –$CH_2$) 3.57 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.22 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.33 (s, 1H, –CH), 6.66–7.78 (m, 7H, Ar–H), 7.36–8.36 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.4 ($C_8$), 32.5 ($C_7$), 33.5 ($C_{18}$), 36.8 ($C_9$), 65.2 ($C_{17}$), 68.3($C_{10}$), 114.5–156.4 ($C_{11}$–$C_{16}$), 114.7–160.7 ($C_{20}$–$C_{25}$), 123.5–157.4 ($C_2$–$C_6$), 170.7 ($C_{19}$); MS (*m/z*): 502 ($M^+$), 504 (M+2); Anal. Calcd for $C_{24}H_{22}N_2O_2FSBr$: C, 57.49; H, 4.42; N, 5.59; Found C, 57.52; H, 4.44; N, 5.61.

*3-(3-Chloro-4-methylphenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5h**)*

This compound was obtained as dark brown solid, yield: 61 %, M.P. 191–193 °C, $R_f$: 0.62; IR (KBr): 3034 (Ar–H), 2954, 2836 (–$CH_2$–), 1715 (–C=O of thiazolidinone), 1220, 1043 (C–O–C), 747 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.13 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.20 (s, 3H, –$CH_3$), 2.53 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.23 (s, 2H, –$CH_2$) 3.52 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.27 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.32 (s, 1H, –CH), 6.62–7.80 (m, 8H, Ar–H), 7.35–8.34 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.5 ($C_8$), 25.7 ($C_{26}$), 32.6 ($C_7$), 33.7 ($C_{18}$), 36.6 ($C_9$), 65.2 ($C_{17}$), 68.3 ($C_{10}$), 114.4–156.5 ($C_{11}$–$C_{16}$), 119.6–140.1 ($C_{20}$–$C_{25}$), 123.4–157.4 ($C_2$–$C_6$), 170.3 ($C_{19}$); MS (*m/z*): 452 ($M^+$), 454 (M+2); Anal. Calcd for $C_{25}H_{25}N_2O_2ClS$: C, 66.28; H, 5.56; N, 6.18; Found C, 66.26; H, 5.57; N, 6.20.

*3-(3,5-Dichlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5i**)*

This compound was obtained as off brown solid, yield: 68 %, M.P. 155–157 °C, $R_f$: 0.65; IR (KBr): 3036 (Ar–H), 2957, 2834 (–$CH_2$–), 1714 (–C=O of thiazolidinone), 1223, 1045 (C–O–C), 746 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.16 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.58 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.24 (s, 2H, –$CH_2$) 3.54 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.24 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.32 (s, 1H, –CH), 6.64–7.87 (m, 7H, Ar–H), 7.37–8.37 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.6 ($C_8$), 32.4 ($C_7$), 33.6 ($C_{18}$), 36.3 ($C_9$), 65.5 ($C_{17}$), 68.0 ($C_{10}$), 114.0–156.2 ($C_{11}$–$C_{16}$), 120.1–135.9 ($C_{20}$–$C_{25}$), 123.0–157.5 ($C_2$–$C_6$), 170.3 ($C_{19}$); MS (*m/z*): 472 ($M^+$), 474 (M+2), 476 (M+4); Anal. Calcd. for $C_{24}H_{22}N_2O_2Cl_2S$: C, 60.89; H, 4.68; N, 5.92; Found C, 60.88; H, 4.69; N, 5.93.

*3-(2-Chlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5j**)*

This compound was obtained as brown solid, yield: 72 %, M.P. 130–132 °C, $R_f$: 0.69; IR (KBr): 3034 (Ar–H), 2955, 2835 (–$CH_2$–), 1717 (–C=O of thiazolidinone), 1225, 1046 (C–O–C), 748 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.17 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.57 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.23 (s, 2H, –$CH_2$) 3.56 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.34 (s, 1H, –CH), 6.58–7.81 (m, 8H, Ar–H), 7.36–8.39 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.2 ($C_8$), 32.5 ($C_7$), 33.7 ($C_{18}$), 36.6 ($C_9$), 65.4 ($C_{17}$), 68.4 ($C_{10}$), 114.5–156.6 ($C_{11}$–$C_{16}$), 123.0–140.2 ($C_{20}$–$C_{25}$), 123.4–157.1 ($C_2$–$C_6$), 170.4 ($C_{19}$); MS (*m/z*): 438 ($M^+$), 440 (M+2); Anal. Calcd. for $C_{24}H_{23}N_2O_2ClS$: C, 65.67; H, 5.28; N, 6.38; Found C, 65.65; H, 5.26; N, 6.39.

*3-(2-Fluorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5k**)*

This compound was obtained as yellow liquid, yield: 75 %, M.P. semi solid, $R_f$: 0.63; IR (KBr): 3041 (Ar–H), 2950, 2831 (–$CH_2$–), 1711 (–C=O of thiazolidinone), 1226, 1042 (C–O–C), 972 (C–F); $^1$H NMR ($CDCl_3$): $\delta$ 1.20 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.53 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.25 (s, 2H, –$CH_2$) 3.54 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.28 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.33 (s, 1H, –CH), 6.63–7.88 (m, 8H, Ar–H), 7.38–8.38 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.6 ($C_8$), 32.8 ($C_7$), 33.4 ($C_{18}$), 36.5 ($C_9$), 65.7 ($C_{17}$), 68.5 ($C_{10}$), 114.8–156.4 ($C_{11}$–$C_{16}$), 115.7–127.8 ($C_{20}$–$C_{25}$), 123.3–157.5 ($C_2$–$C_6$), 170.5 ($C_{19}$); MS (*m/z*): 422 ($M^+$); Anal. Calcd. for $C_{24}H_{23}N_2O_2FS$: C, 68.22, H, 5.49, N, 6.63; found C, 68.24, H, 5.52, N, 6.65.

*3-(2,5-Dichlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5l**)*

This compound was obtained as brown liquid, yield: 63 %, M.P. semi solid, $R_f$: 0.64; IR (KBr): 3032 (Ar–H), 2954, 2836 (–$CH_2$–), 1713 (–C=O of thiazolidinone), 1225, 1043 (C–O–C), 743 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.21 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.51 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.23 (s, 2H, –$CH_2$) 3.55 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.34 (s, 1H, –CH), 6.60–7.90 (m, 7H, Ar–H), 7.35–8.36 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.3 ($C_8$), 32.2 ($C_7$), 33.5 ($C_{18}$), 36.6 ($C_9$), 65.8 ($C_{17}$), 68.1 ($C_{10}$), 114.4–156.3 ($C_{11}$–$C_{16}$), 123.4–141.6 ($C_{20}$–$C_{25}$), 123.6–157.4 ($C_2$–$C_6$), 170.4 ($C_{19}$);

MS (*m/z*): 472 ($M^+$), 474 (M+2), 476 (M+4); Anal. Calcd. for $C_{24}H_{22}N_2O_2Cl_2S$: C, 60.89; H, 4.68; N, 5.92; Found C, 60.90; H, 4.67; N, 5.90.

*3-(2,3-Dichlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5m**)*

This compound was obtained as brown liquid, yield: 65 %, M.P. semi solid, $R_f$: 0.66; IR (KBr): 3036 (Ar–H), 2958, 2838 (–$CH_2$–), 1718 (–C=O of thiazolidinone), 1223, 1045 (C–O–C), 747 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.54 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.24 (s, 2H, –$CH_2$) 3.52 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.23 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.35 (s, 1H, –CH), 6.56–7.79 (m, 7H, Ar–H), 7.36–8.37 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.6 ($C_8$), 32.5 ($C_7$), 33.6 ($C_{18}$), 36.4 ($C_9$), 65.5 ($C_{17}$), 68.3 ($C_{10}$), 114.3–156.7 ($C_{11}$–$C_{16}$), 121.1–141.6 ($C_{20}$–$C_{25}$), 123.2–157.3 ($C_2$–$C_6$), 170.6 ($C_{19}$); MS (*m/z*): 472 ($M^+$), 474 (M+2), 476 (M+4); Anal. Calcd. for $C_{24}H_{22}N_2O_2Cl_2S$: C, 60.89; H, 4.68; N, 5.92; Found C, 60.87; H, 4.67; N, 5.93.

*3-(3-Chlorophenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5n**)*

This compound was obtained as brown limpid, yield: 74 %, M.P. semi solid, $R_f$: 0.63; IR (KBr): 3035 (Ar–H), 2955, 2836 (–$CH_2$–), 1715 (–C=O of thiazolidinone), 1224, 1047 (C–O–C), 744 (C–Cl); $^1$H NMR ($CDCl_3$): $\delta$ 1.17 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.56 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.25 (s, 2H, –$CH_2$) 3.55 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.34 (s, 1H, –CH), 6.67–7.81 (m, 8H, Ar–H), 7.37–8.34 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.7 ($C_8$), 32.4 ($C_7$), 33.3 ($C_{18}$), 36.8 ($C_9$), 65.7 ($C_{17}$), 68.4 ($C_{10}$), 114.4–156.6 ($C_{11}$–$C_{16}$), 119.7–143.1 ($C_{20}$–$C_{25}$), 123.4–157.6 ($C_2$–$C_6$), 170.3 ($C_{19}$); MS (*m/z*): 438 ($M^+$), 440 (M+2); Anal. Calcd. for $C_{24}H_{23}N_2O_2ClS$: C, 65.67; H, 5.28; N, 6.38; Found C, 65.66; H, 5.29; N, 6.37.

*3-(4-Methylphenyl)-2-(4-(2-(5-ethylpyridin-2-yl)ethoxy)phenyl)thiazolidin-4-one (**5o**)*

This compound was obtained as yellow solid, yield: 67 %, M.P. 99–101 °C, $R_f$: 0.68; IR (KBr): 3033 (Ar–H), 2958, 2838 (–$CH_2$–), 1710 (–C=O of thiazolidinone), 1225, 1042 (C–O–C); $^1$H NMR ($CDCl_3$): $\delta$ 1.15 (t, $J = 8.0$ Hz, 3H, –$CH_3$), 2.16 (s, 3H, –$CH_3$), 2.51 (q, $J = 8.0$ Hz, 2H, –$CH_2$),3.20 (s, 2H, –$CH_2$) 3.50 (t, $J = 7.0$ Hz, 2H, –$CH_2$), 4.25 (t, $J = 7.0$ Hz, 2H, –$CH_2$–O), 5.30 (s, 1H, –CH), 6.85–7.70 (m, 8H, Ar–H), 7.33–8.31 (m, 3H, Pyridine-H); $^{13}$C NMR ($CDCl_3$): $\delta$ 20.9 ($C_8$), 25.6 ($C_{26}$), 32.4 ($C_7$), 33.5 ($C_{18}$), 36.4 ($C_9$), 65.4 ($C_{17}$), 68.1 ($C_{10}$), 114.6–156.7 ($C_{11}$–$C_{16}$), 120.6–137.2 ($C_{20}$–$C_{25}$), 123.1–157.2 ($C_2$–$C_6$), 170.1 ($C_{19}$); MS (*m/z*): 419 ($M^+$); Anal. Calcd. for $C_{25}H_{26}N_2O_2S$: C, 71.74; H, 6.26; N, 6.69; Found C, 71.76; H, 6.25; N, 6.70.

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