A novel multicomponent reaction between amino acids, aromatic aldehydes and \( p \)-toluenesulfonylmethyl isocyanide: an efficient and green one-pot synthesis using nanosilica

Ladan Edjliali\(^a\), Esmail Vessally\(^b\), Zahra Jafari\(^b\) and Mehdi D. Esrafi\(^c\)

\(^a\)Department of Chemistry, Tabriz Branch, Islamic Azad University, Tabriz, Iran; \(^b\)Department of Chemistry, Payame Noor University, Tehran, Iran; \(^c\)Chemistry Department, University of Maragheh, Maragheh, Iran

**ABSTRACT**

The novel diastereomeric sulfonylamide derivatives 4a–h were synthesized by multi-component reactions between (L)-\( \alpha \)-amino acids, 1, aromatic aldehydes, 2 and \( p \)-toluenesulfonylmethyl isocyanide, 3 in water/methanol using nanosilica as a catalyst. The one-pot green reactions were carried out at room temperature with a quantitative yield in water/methanol. The yield of the reactions and products was determined and discussed. The reusability of the catalyst is discussed.

**ARTICLE HISTORY**

Received 28 November 2014
Accepted 2 November 2015

**KEYWORDS**

Ugi reaction; tosyl acyloxy amino carboxamide derivatives; amino acid; aromatic aldehydes; toluenesulfonylmethyl isocyanide; silica nanoparticles

---

**1. Introduction**

Multi-component reactions (MCRs) based on isocyanide has attracted much attention (1–20). The two most important isocyanide-based MCRs are the Passerini three-component reaction to produce \( \alpha \)-acyloxy carboxamides and the Ugi four-component reaction, which yields the \( \alpha \)-acylamino carboxamides (1). The Ugi reaction has an inherent high atom economy as only a molecule of water is lost and chemical yields in general are high (11). Several groups such as \( \beta \)-amino acids have used in the Ugi reaction to prepare \( \beta \)-lactams (21). This approach relies on acyl transfer in the Mumm rearrangement to form the four-membered ring. The reaction proceeds in moderate yield at room temperature in methanol with formaldehyde or a variety of aryl aldehydes.

There are a few reports about the \( \alpha \)-amino acids used in the Ugi reaction (22). The Ugi reaction offers the possibility to synthesize a great number of different compounds in one reaction. These libraries can then be tested with enzymes or living organisms to find new active pharmaceutical substances. As a continuation our recent studies (23–26), we report the green MCR between (L)-\( \alpha \)-amino acids, aromatic aldehydes and \( p \)-toluenesulfonylmethyl isocyanide in water/methanol (Figure 1).

**2. Experimental detail**

Starting materials and solvents were purchased from Merck (Germany) and Fluka (Switzerland) and were used without further purification. The reactions were monitored by TLC and NMR techniques, which indicated that there were no side products. IR spectra were measured on a Perkin-Elmer RXI, FT-IR spectrometer. \(^1\)H and \(^13\)C NMR spectra (CDCl\(_3\)) were recorded on a Bruker-Avance spectrometer at 300.0 and 75.0 MHz, respectively. Elemental analyses were performed by using a Perkin-Elmer 2400(II) CHN/O analyzer. The TLC plates were prepared from Merck silica gel powder.

**2.1. General procedure for the synthesis of 4a–h**

Silica nanoparticles (SNPs) (0.2 g) were added to a mixture of (L)-\( \alpha \)-aminoacid (1 mmol) and aldehyde (1 mmol) which were stirred in 5 mL water–methanol (1:1) at the room temperature for 60 min. To this mixture, isocyanide (1 mmol) at 15°C was rapidly added and the solution was allowed to stand for 24 h at room temperature. The solvent was removed under reduced pressure and the diastereomeric products (4a–h) were obtained. Two diastereomers produced through purification with preparative
thin layer liquid chromatography (PTLC) method by using petroleum ether–diethyl ether (10:1). Yield of the mixture of the diastereomers was determined after PTLC. The diastereomeric ratio was determined by 1H-NMR analysis of unpurified products (Table 1). The characterization data of the compounds are given below.

2.1.1. Methyl-2-[1-phenyl-2-oxo-2-(tosylmethylamino)ethylamino]-4-methylpentanoate (4a)

Yellow powder, yield 60%. For mixture of two diastereomers: IR (KBr) \(\nu_{\text{max}}/cm^{-1}\): 3292 (NH), 3050 (C–H aromatic), 2949 (C–H aliphatic), 1731 (C=O of ester), 1594 (C=O of amide), 1470 aromatic ring, 1272 (C–O); 1H NMR (300 MHz, CDCl3, major diastereomer): \(\delta\): 0.94 (6H methyl, d, Me2CH–), 1.58 (2H methylene, t, –CHCH2CHN–), 1.78 (1H methine, m, Me2CH–), 2.19 (1H amine), 2.36 (3H methyl, s, benzil), 3.47 (1H methine, s, N–CHCON–), 5.30 (2H methylene, s, CH2–SO2), 7.24–7.56 (9H aromatic, m), 8.05 (NH–amide); 13C NMR (75 MHz, CDCl3, major diastereomer): \(\delta\): 18.2 (2CH3), 18.5 (CH), 30.4 (CH 2), 35.4 (CH3ph), 49.7 (CHCOO), 52.5 (OCH3), 59.9 (CHCON), 62.1 (CH2SO2), 126.5, 127.1, 128.9, 130.1, 130.3, 139.13, 139.34, 142.4 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.2. Methyl-2-[1-(2,5-dimethoxyphenyl)-2-oxo-2-(tosylmethylamino)ethylamino]-4-methylpentanoate (4b)

Yellow powder, yield 68%. For mixture of two diastereomers: IR (KBr) \(\nu_{\text{max}}/cm^{-1}\): 3290 (NH), 3052 (C–H aromatic), 2949 (C–H aliphatic), 1731 (C=O of ester), 1594 (C=O of amide), 1470 aromatic ring, 1272 (C–O); 1H NMR (300 MHz, CDCl3, major diastereomer): \(\delta\): 0.96 (6H methyl, d, Me2CH–), 1.59 (2H methylene, t, –CHCH2CHN–), 1.79 (1H methine, m, Me2CH–), 2.19 (1H amine), 2.38 (3H methyl, s, benzil), 3.48 (1H methine, t, –NCHR–COO), 5.30 (2H methylene, s, CH2–SO2), 6.96–7.59 (9H aromatic, m), 8.05 (NH–amide); 13C NMR (75 MHz, CDCl3, major diastereomer): \(\delta\): 18.5 (2CH3), 18.9 (CH), 31.2 (CH3), 34.7 (CH3ph), 50.3 (CHCOO), 52.8 (OCH3), 60.1 (CHCON), 62.3 (CH2SO2), 115.9, 126.7, 128.9, 131.9, 136.2, 139.34, 142.4, 162.5 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.3. Methyl-2-[1-(4-furorophenyl)-2-oxo-2-(tosylmethylamino)ethylamino]-4-methyl pentanoate (4c)

Yellow powder, yield 57%. For mixture of two diastereomers: IR (KBr) \(\nu_{\text{max}}/cm^{-1}\): 3291 (NH), 3052 (C–H aromatic), 2950 (C–H aliphatic), 1730 (C=O of ester), 1596 (C=O of amide), 1471 aromatic ring, 1272 (C–O); 1H NMR (300 MHz, CDCl3, major diastereomer): \(\delta\): 0.96 (6H methyl, d, Me2CH–), 1.59 (2H methylene, t, –CHCH2CHN–), 1.79 (1H methine, m, Me2CH–), 2.19 (1H amine), 2.38 (3H methyl, s, benzil), 3.48 (1H methine, t, –NCHR–COO), 5.30 (2H methylene, s, –COO–CH3), 4.88 (1H methine, s, N–CHCON–), 1.19 (1H amine, 2.16 (1H amine), 2.36 (3H methyl, s, benzil), 3.47 (1H methine, t, –NCHR–COO), 3.52 (3H methyl, s, –COO–CH3), 3.65 (3H methyl, s, phOCH3), 4.01 (3H methyl, s, phOCH3), 4.86 (1H methine, s, N–CHCON–), 5.30 (2H methylene, s, CH2–SO2), 7.24–7.56 (9H aromatic, m), 8.05 (sec-amine); 13C NMR (75 MHz, CDCl3, major diastereomer): \(\delta\): 18.1 (2CH3), 18.3 (CH), 30.2 (CH2), 35.3 (CH3ph), 49.6 (CHCOO), 52.3 (OCH3), 56.5 (phOCH3), 57.3 (phOCH3), 59.8 (CHCON), 62.0 (CH2SO2), 109.8, 114.0, 115.1, 126.5, 128.4, 128.9, 139.34, 142.4, 149.9, 155.1 (C-aromatic), 170.5 (COO), 172.1 (CON).
Table 1. Yields of the reactions for the synthesis of tosylmethylamino derivatives 4a–h.

| Entry | 1 | 2 | 3 | Yield (%)<sup>a</sup> | Diastereomeric ratio (d.r.)<sup>b</sup> |
|-------|---|---|---|----------------------|----------------------------------------|
| 4a    | 10<sup>c</sup> | 36<sup>d</sup> | 60<sup>e</sup> | 60:40                |
| 4b    | 12<sup>c</sup> | 39<sup>d</sup> | 68<sup>e</sup> | 62:38                |
| 4c    | _<sup>c</sup> | 25<sup>d</sup> | 57<sup>e</sup> | 59:41                |
| 4d    | _<sup>c</sup> | 26<sup>d</sup> | 60<sup>e</sup> | 60:40                |
| 4e    | 15<sup>c</sup> | 39<sup>d</sup> | 68<sup>e</sup> | 65:35                |
| 4f    | 18<sup>c</sup> | 36<sup>d</sup> | 65<sup>e</sup> | 60:40                |
| 4g    | _<sup>c</sup> | 26<sup>d</sup> | 55<sup>e</sup> | 59:41                |
| 4h    | _<sup>c</sup> | 30<sup>d</sup> | 42<sup>e</sup> | 58:42                |

<sup>a</sup> Yield of the mixture of diastereomer after PTLC.
<sup>b</sup> Diastereomeric ratio was determined by 1H-NMR analysis of unpurified products.
<sup>c</sup> Without any catalyst.
<sup>d</sup> Silica as a catalyst.
<sup>e</sup> SNPs as a catalyst.
2.1.4. Methyl-2-[1-(4-chlorophenyl-2-oxo-2-(tosylmethylamino)ethylamino)-4-methyl pentanoate (4d)

Yellow powder, yield 60%. For a mixture of two diastereomers: IR (KBr) umax/cm⁻¹: 3291(NH), 3052(C–H aromatic), 2950 (C–H aliphatic), 1730(C= O of ester), 1596 (C= O of amide), 1471 aromatic ring, 1272 (C–O); ¹H NMR (300 MHz, CDCl₃, major diastereomer): δ 0.94 (6H methyl, d, Me₂CH–), 1.58 (2H methylene, t, –CHCH₂CHN–), 1.78 (1H methine, m, Me₂CH–), 2.17 (1H amine), 2.38 (3H methyl, s, benzil), 3.46(1H methine, t, –NCHR–COO), 3.52(3H methyl, s, –COO–CH₃), 4.86 (1H methine, s, –N–CHCON–), 5.30(2H methylene, s, 1alpha–N CO–C, CH₂SO₂), 7.31–7.56(8H aromatic, m), 8.05 (NH-amide); ¹³C NMR (75 MHz, CDCl₃, major diastereomer): δ: 18.2 (2CH₃), 18.4 (CH), 30.3 (CH₂), 35.4(CH₃ph), 49.6 (CHCOO), 52.3 (OCH₃), 59.7 (CHCON), 62.0 (CH₂SO₂), 126.5, 127.5, 128.9, 132.5, 134.9, 137.4, 139.34, 142.4 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.5. Methyl 2-(1-(2,4-dimethoxyphenyl)-2-oxo-2-(tosylmethylamino)ethylamino)-4-methyl pentanoate (4e)

Yellow powder, yield 69%. For a mixture of two diastereomers: IR (KBr) umax/cm⁻¹: 3290(NH), 3050(C–H aromatic), 2949 (C–H aliphatic), 1730(C=O of ester), 1592(C= O of amide), 1470 aromatic ring, 1271 (C–O); ¹H NMR (300 MHz, CDCl₃, major diastereomer): δ 0.94 (6H methyl, d, Me₂CH–), 1.57 (2H methylene, t, –CHCH₂CHN–), 1.77 (1H methine, m, Me₂CH–), 2.16 (1H amine), 2.38 (3H methyl, s, benzil), 3.45(1H methine, t, –NCHR–COO), 3.50 (3H methyl, s, –COO–CH₃), 4.85 (1H methine, s, –NCHCON–), 5.30(2H methylene, s, CH₂–SO₂), 7.31–7.71(8H aromatic, m), 8.05(sec-amine), ¹³C NMR (75 MHz, CDCl₃, major diastereomer): δ: 18.0 (2CH₃), 18.3 (CH), 30.4 (CH₂), 35.1(CH₃ph), 49.5 (CHCOO), 52.3 (OCH₃), 59.5 (CHCON), 62.0 (CH₂SO₂), 123.4, 126.5, 128.9, 129.3, 131.2, 138.6, 139.34, 142.4 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.6. Methyl-2-[1-(4-bromophenyl-2-oxo-2-(tosylmethylamino)ethylamino)-4-methyl pentanoate (4f)

Yellow powder, yield 65%. For a mixture of two diastereomers: IR (KBr) umax/cm⁻¹: 3290(NH), 3050(C–H aromatic), 2949 (C–H aliphatic), 1730(C=O of ester), 1592(C= O of amide), 1470 aromatic ring, 1271 (C–O); ¹H NMR (300 MHz, CDCl₃, major diastereomer): δ 0.94 (6H methyl, d, Me₂CH–), 1.57 (2H methylene, t, –CHCH₂CHN–), 1.77 (1H methine, m, Me₂CH–), 2.16 (1H amine), 2.38 (3H methyl, s, benzil), 3.45(1H methine, t, –NCHR–COO), 3.50 (3H methyl, s, –COO–CH₃), 4.85 (1H methine, s, –NCHCON–), 5.30(2H methylene, s, CH₂–SO₂), 7.31–7.71(8H aromatic, m), 8.05(sec-amine), ¹³C NMR (75 MHz, CDCl₃, major diastereomer): δ: 18.0 (2CH₃), 18.3 (CH), 30.4 (CH₂), 35.1(CH₃ph), 49.5 (CHCOO), 52.3 (OCH₃), 59.5 (CHCON), 62.0 (CH₂SO₂), 123.4, 126.5, 128.9, 129.3, 131.2, 138.6, 139.34, 142.4 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.7. Methyl-2-[1-(4-nitrophenyl-2-oxo-2-(tosylmethylamino)ethylamino)-4-methylpentanoate (4g)

Yellow powder, yield 55%. For a mixture of two diastereomers: IR (KBr) umax/cm⁻¹: 3288(NH), 3052(C–H aromatic), 2950 (C–H aliphatic), 1732(C=O of ester), 1598 (C=O of amide), 1471 aromatic ring, 1273 (C–O); ¹H NMR (300 MHz, CDCl₃, major diastereomer): δ 0.95 (6H methyl, d, Me₂CH–), 1.59 (2H methylene, t, –CHCH₂CHN–), 1.78 (1H methine, m, Me₂CH–), 2.16 (1H amine), 2.39 (3H methyl, s, benzil), 3.47(1H methine, t, –NCHR–COO), 3.52 (3H methyl, s, –COO–CH₃), 4.88 (1H methine, s, –NCHCON–), 5.32(2H methylene, s, CH₂–SO₂), 7.31–8.05(8H aromatic, m), 8.06(NH-amide); ¹³C NMR (75 MHz, CDCl₃, major diastereomer): δ: 18.3 (2CH₃), 18.6 (CH), 30.4 (CH₂), 35.4(CH₃ph), 49.7 (CHCOO), 52.5 (OCH₃), 60.2 (CHCON), 62.1 (CH₂SO₂), 121.6, 126.5, 128.9, 130.4, 139.34, 142.4, 143.7, 147.9 (C-aromatic), 170.5 (COO), 172.1 (CON).

2.1.8. Methyl 3-[3-(4,5-dihydro-1H-imidazol-5-yl)-2-(2-oxo-1-(2-imidazoline)-4-yl)-2-(tosyl methylamino)ethylamino)propanoate (4h)

White powder, yield 42%, H NMR (300 MHz, CDCl₃, major diastereomer): δ 1.9 (2H methylene, t CH₂-Imidazoline), 1.99(1H amine), 2.35 (3H methyl, s, benzil), 3.25(1H methine, t, –NCHR–COO), 3.35 (1H methine, m, HNCH– in the imidazoline ring), 3.67(3H methyl –OCH₂CH₂), 3.90(2H, t, methylene = NCH₂– in the imidazoline ring), 4.90(1H methine, s, N–CHCON–), 5.10(2H methylene, CH₂–SO₂), 5.7(1H, s, –NCH= N), 7.31–8.60(8H aromatic, m), 7.80(1H, amide); ¹³C NMR (75 MHz, CDCl₃, major diastereomer): δ 36.5 (CH₃), 34.4(CH₃ph), 46.7 (CHCOO), 52.0 (OCH₃), 55.7(–NCH₂–), 57.8(HNCH–), 59.2 (CHCON), 62.2 (CH₂SO₂), 121.8, 126.5, 129.0, 139.30, 142.1, 144.5, 149.8 (C-aromatic), 144.9 (–NCH= N), 172.1 (CON), 173.5 (COO).
3. Results and discussion

As a continuation of our studies, the aim of this work was to develop simple, one-pot, green and multi-component reactions between α-amino acids, 1, aromatic aldehydes 2 and p-toluenesulfonylmethyl isocyanide 3 in the presence of the SNPs at room temperature, leading to tosylmethylamino derivatives 4 (Figure 1 and Table 1). The reaction occurs smoothly in the presence of the SNP at ambient temperature in water/methanol to produce the tosylmethylamino derivatives 4 in good yields (Table 1). The diastereomeric products were obtained after purification with a quantitative yield in one pot.

Figure 2. X-ray diffraction pattern of the synthesized of SNPs.

Figure 3. SEM of the synthesized of SNPs.

Figure 4. Proposed mechanism for the formation of tosylmethylamino derivatives 4a–h in water–methanol.
the absence of SNP, the reactions were not carried out to obtain the compounds \textbf{4a–h} or obtain with low yields. We also used silica particles as catalyst which increase slightly the yield of reactions (Table 1).

The SNPs were prepared by thermal decomposition of rice hulls \cite{27}. The results from X-ray diffraction (XRD) showed that the sample was SNP as indicated by broadened peaks around $2\theta = 22^\circ$ (Figure 2). The scanning electron microscopy (SEM) was used to investigate the morphology and grain size of the SNP (Figure 3) \cite{28,29}.

The diastereomeric structures of the products were deduced from elemental analyses IR, $^1$H NMR and $^{13}$C NMR spectra. For example, the $^1$H-NMR spectrum of a major diastereomer in \textbf{4a} consists of a doublet for $2\text{CH}_3$ of Me$_2$CH– ($\delta = 0.94$ ppm), and a triplet for 2H methylene of –CHCH$_2$CHN– ($\delta = 1.58$ ppm), a multiplet for 1H methine of Me$_2$CH– ($\delta = 1.78$), a singlet for NH ($\delta = 2.19$ ppm), a singlet for 3H methyl of benizil ($\delta = 2.36$), a triplet for 1H methane of NCHR–COO ($\delta = 3.47$), a singlet for 3H methyl of –COO–CH$_3$ ($\delta = 3.52$), a singlet for 1H methane of –NCHR–COO ($\delta = 4.86$), a singlet for 2H methylene of CH$_2$–SO$_2$ ($\delta = 5.30$). The aryl groups exhibited characteristic signals in the aromatic region of the spectrum.

The $^1$H-decoupled $^{13}$C-NMR spectrum of a major diastereomer in \textbf{4a} showed 18 distinct signals; partial assignment of these signals is given in the experimental section. The $^1$H- and $^{13}$C-NMR spectra of compounds \textbf{4b–h} were similar to those of \textbf{4a}, except for the aromatic moiety, and the alkyl groups, which exhibited characteristic signals with appropriate chemical shifts.

As indicated in Table 1, the reactions proceeded efficiently with leucine \textbf{1} while lysine is not a suitable starting material in this reaction.

Although we have not established the mechanism of the reaction in an experimental manner, a plausible reaction sequence that accounts for the formation of \textbf{4} is shown in Figure 4. Thus, the condensation of amino acid \textbf{1} and aldehyde \textbf{2} gives an iminium ion intermediate \textbf{5}, which is then attacked by the isocyanide \textbf{3} in the presence of SNP to afford intermediate \textbf{6}. The cyclization of the ionic intermediate \textbf{6} leads to the intermediate \textbf{7} that could then lead to the formation of the final product \textbf{4}.

### 3.1. Catalyst recovery

The recovered catalyst from the experiment was washed with acetone (3x5 mL). Then, it was dried and used in the synthesis of sulfonylamide derivatives \textbf{4a–h}. Then, the catalyst was recycled for five times. The separated catalyst was used several times with a slightly decreased activity.

### 4. Conclusion

The novel diastereomeric sulfonylamide derivatives \textbf{4a–h} were synthesized by multi-component reactions between $\alpha$-amino acids, \textbf{1}, aromatic aldehydes, \textbf{2} and $p$-toluenesulfonylmethyl isocyanide, \textbf{3} in water/methanol using nanosilica as a catalyst. The yield of these reactions was increased using SNPs as a catalyst. The electron-donating groups attached at the para position of aromatic aldehydes increased the yield of the reactions. The separated catalyst was used several times with a slightly decreased activity.

### Disclosure statement

No potential conflict of interest was reported by the authors.

### Funding

The authors are thankful to Tabriz Branch, Islamic Azad University for the financial support of this research work. This work was also supported by the ‘Sandoogh Hemayate as Pajuoheshgharane Keshvare’ Iran.

### References

\begin{enumerate}
    \item Ugi, I.; Lohberger, S.; Karl, R. In \textit{Comprehensive Organic Synthesis}. Trost, B.M., Fleming, I., Eds.; Pergamon Press: Oxford, \textbf{1991}, Vol. 2, pp 1083.
    \item Sapi, J.; Laronze, J.-Y. Arkivoc \textbf{2004}, 7, 208–222.
    \item Zhu, J.; Bienayme H. \textit{Multicomponent Reactions}; Wiley-VCH: Weinheim, \textbf{2005}.
    \item Ugi, I. Angew. Chem. Int. Ed. \textbf{1962}, 1, 8–21.
    \item Hazeri, N.; Maghsoodlou, M.T.; Habibi-Khorassani, S.M.; Ziyaadini, M.; Marandi, G.; Khandan-Barani, K.; Bijanzadeh, H.R. Arkivoc \textbf{2007}, 8, 34–40.
    \item Dömling, A.; Beck, B.; Herdtweck, E.; Antuch, W.; Oefner, C.; Yehia, N.; and Gracia Marques, A. Arkivoc \textbf{2007}, 17, 99–109.
    \item Dömling, A.; Ugi, I. Angew. Chem. Int. Ed. \textbf{2000}, 39 (18), 3168–3210.
    \item Ugi, I.; Werner, B.; Dömling, A. Molecules \textbf{2003}, 8 (1), 53–66.
    \item Ugi, I. Pure Appl. Chem. \textbf{2001}, 73, 187–191.
    \item Dömling, A. Chem Rev. \textbf{2006}, 106 (1), 17–89.
    \item Ugi, I.; Steinbrückner, C. ChemBer. \textbf{1961}, 94, 2802–2814.
    \item Soulodozi, A.; Ramazani, A. Tetrahedron Lett. \textbf{2007}, 48 (9), 1549–1551.
    \item Soulodozi, A.; Ramazani, A.; Boulimani, N.; Welter, R. Tetrahedron Lett. \textbf{2007}, 48 (14), 2617–2620.
    \item Moliner, F.D.; Hulme, C. Tetrahedron Lett. \textbf{2012}, 53 (43), 5787–5790.
    \item Moliner, F.D.; Hulme, C. Org. Lett. \textbf{2012}, 14 (5), 1354–1357.
    \item Xu, Z.; Moliner, D.F.; Cappelli, A.P.; Hulme, C. Angew. Chem. Int. Ed. \textbf{2012}, 51 (32), 8037–8040.
    \item Ayaz, M.; Martinez-Ariza, G.; Hulme, C. Synlett \textbf{2014}, 25 (12), 1680–1684.
    \item Ramazani, A.; Khoobi, M.; Torkaman, A.; Nasrabadi, F.Z.; Forootanfar, H.; Shakibaie, M.; Jafari, M.; Ameri, A.
\end{enumerate}
Emami, S.; Faramarzi, M.A.; Foroumadi, A.; Shafiee, A. Eur. J. Med. Chem. 2014, 78, 151–156.

(19) Ramazani, A.; Rezaei, A. Org. Lett. 2010, 12 (12), 2852–2855.

(20) Taran, J.; Ramazani, A.; Joo, S.W.; Ślepokura, K.; Lis, T. Helvetica Chim. Acta 2014, 97 (8), 1088–1096.

(21) Gedey, S.; Van der Eycken, J.; Fülöp, F. Org. Lett. 2002, 4 (11), 1967–1969.

(22) Mandai, H.; Irie, S.; Mitsudo, K.; Suga, S. Molecules 2011, 16 (12), 8815–8832.

(23) Vessally, E.; Ramazani, A.; Shabrendi, H.; Ghadimi, R.; Rouhani, M. J. Chem. 2013. doi.org/10.1155/2013/761982.

(24) Vessally, E.; Ramazani, A.; Yaaghibi, E. Monatsh Chem. 2011, 142 (11), 1143–1147.

(25) Vessally, E.; Fereyduni, E.; Shabrendi, H.; Esrafil, M.D. Spectrochim. Acta A 2013, 116, 65–73.

(26) Asadi, Z.; Asnaashari, M.B.; Vessally, E.; Esrafil, M.D. Spectrochim. Acta A 2015, 140, 585–599.

(27) Souzaa, M.F.D.; Batistaa, P.S.; Regiania, I.; Liboriob, J.B.L.; Souzac, D.P.F.D. Mater. Res. 2000, 3, 25–30.

(28) Ramazani, A.; Mahyari, A. Helv. Chim. Acta 2010, 93 (11), 2203–2209.

(29) Ramazani, A.; Mahyari, A.; Lashgari, H.; Ślepokura, K.; Lis, T. Helv. Chim. Acta 2011, 94 (4), 611–622.