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SYNTHESIS OF HETEROCYCLIC COMPOUNDS USING BASIC ZEOLITE Csβ*

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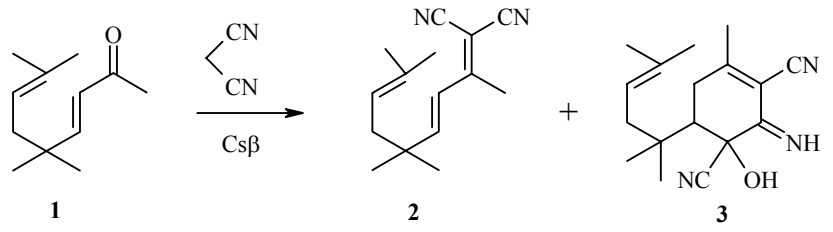
The application of the basic zeolite Csβ as catalyst for the interaction of methyl vinyl ketone (MVK) with 5-methoxybenzimidazole-2-thiol leads to a Michael heteroreaction exclusively at the N-nucleophilic center with the formation of a fairly unusual product of di-addition of MVK to thiol. The reaction of 1,2,4-triazole-3-thiol with MVK in the presence of zeolite Csβ proceeds both at the S- and also at the N-nucleophilic center and leads to the formation of products of mono- and diaddition according to Michael, and also to the product of heterocyclization. On interacting crotonaldehyde with salicylaldehyde in the presence of Csβ 2-methyl-2H-chromene-3-carbaldehyde is formed.

Keywords: methyl vinyl ketone, 5-methoxybenzimidazole-2-thiol, 1,2,4-triazole-3-thiol, basic zeolite, heterogeneous catalysis, Michael addition.

We showed previously that basic zeolite Csβ, containing clusters of cesium oxides in its structure, is a fairly efficient heterogeneous catalyst for reactions of α,β-unsaturated carbonyl compounds of terpenoids with various CH-acids [1, 2]. The use of zeolite Csβ enabled reactions to be carried out without solvent and simplified treatment of reaction mixtures, the catalyst was applied in catalytic amounts and may be regenerated by calcining

Scheme 1



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* Dedicated to Academician of the Russian Academy of Sciences B. A. Trofimov on his 70th jubilee.

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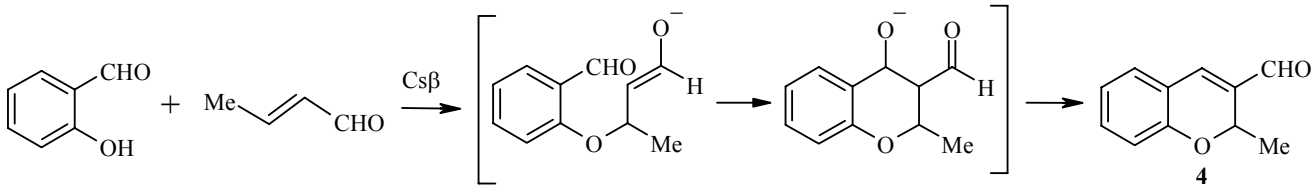
in air without loss of catalytic activity. The application of zeolite Csβ by us led in certain cases to unusual reaction products, for example, on interacting ketone **1** with malononitrile in the presence of zeolite Csβ (Scheme 1) the Knoevenagel reaction product **2** and the fairly complex polyfunctional compound **3** were formed.

It is necessary to record that compound **3** is promising for the study of its biological activity, since substances containing the CN group in the β-position to a nitrogen atom may display antitubercular [3] and antitumor [4] activity.

While continuing to broaden the circle of reactions catalyzed by basic zeolite Csβ, we have studied certain Michael heteroreactions, including interaction with O-, N-, and S-nucleophiles in the present work. As in the previous case all reactions were carried out without solvent, at room temperature, and zeolite Csβ was used in catalytic amounts (12-20 wt. %).

The interaction between salicylaldehyde and crotonaldehyde in the presence of zeolite Csβ during 5 days leads to the formation of a 2H-chromene derivative, *viz*. compound **4** in 50% yield (Scheme 2).

Scheme 2

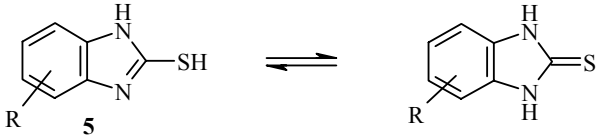


This compound was obtained previously in [5]. Various catalysts have been applied for the synthesis of chromenes and their derivatives, including complexes of palladium and ruthenium [6-8], secondary amines [9], cyclic diamines [10-12], and zeolites [13]. It is known that the interaction of salicylaldehyde derivatives with various α,β-unsaturated carbonyl compounds in the presence of basic catalysts proceeds as a tandem Michael oxareaction and aldol condensation [10, 14] (Scheme 2). Preferably the formation of compound **4** proceeds by an analogous mechanism. It is necessary to record that that compounds containing a 2H-chromene framework possess fairly varied biological activity [15, 16].

At the present time even more studies are appearing devoted to the use of basic zeolites for catalyzing addition reactions of various types to unsaturated compounds [17-19]. For the first time we have studied the reaction of methyl vinyl ketone (MVK) with thiols containing simultaneously S- and N-nucleophilic centers in the presence of zeolite Csβ.

For the synthesis of compounds displaying antiulcer activity 5-methoxybenzimidazole-2-thiol (**5**) is used in many cases [20-22]. It is known that azolethiols may exist in two tautomeric forms (Scheme 3) [23-26] and may interact with unsaturated compounds, depending on the conditions, or at a nitrogen atom [27], or at a sulfur atom [24, 25]. Usually in the presence of basic catalysts addition takes place initially at the sulfur atom, possessing a greater nucleophilicity than the nitrogen atom [24, 25].

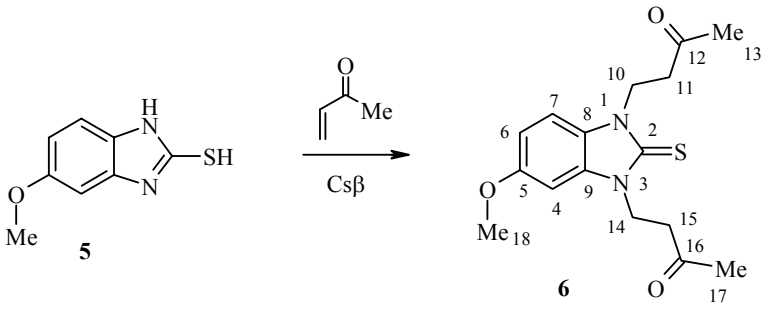
Scheme 3



We carried out the reaction of compound **5** with MVK on basic zeolite Csβ (Scheme 4) during 60 h. Compound **6** was isolated from the reaction mixture as the sole product in 50% yield on reacted thiol **5** (Scheme 4), in which the MVK molecule is added to benzimidazolethiol **5** exclusively at the nitrogen atom. Conversion of compound **5** was 43%. A structural analog of compound **6**, containing no methoxyl group, was obtained previously in [28], in which the authors used sodium methylate as reaction catalyst.

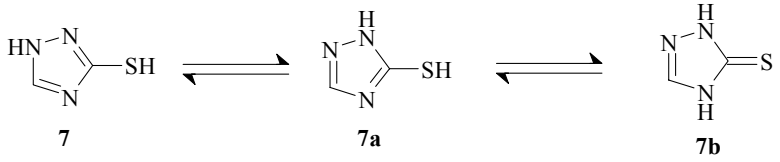
The absence from the reaction mixture of products of the addition of MVK at the sulfur atom may be explained by the fact that compound **5** in the presence of basic zeolite Csβ interacts with MVK exclusively as the thione (Scheme 3). It is interesting that, in spite of the presence in the reaction mixture of unreacted initial compound **5**, we did not detect in the reaction mixture products corresponding to the addition of only one molecule of MVK to benzimidazolethiol **5**.

Scheme 4



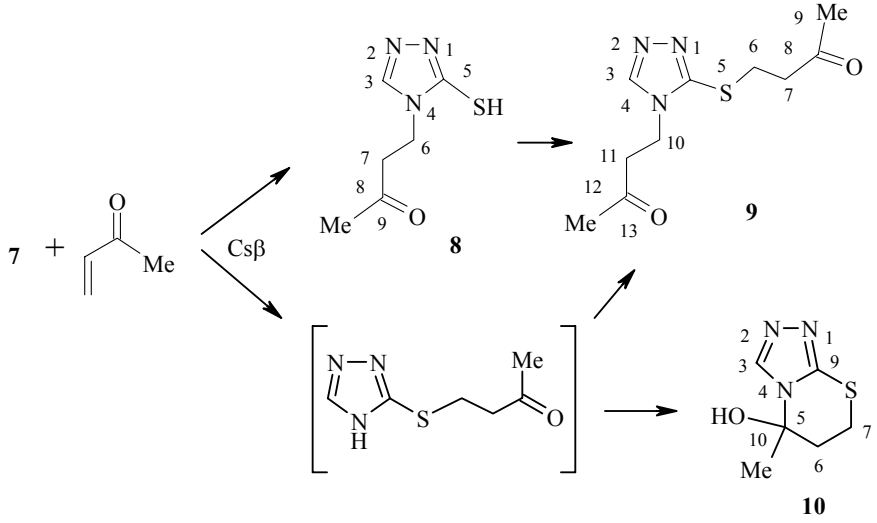
One further S,N-nucleophile, with which we carried out reaction of methyl vinyl ketone in the presence of basic zeolite Csβ, was 1,2,4-triazole-3-thiol (**7**). Compound **7** was used for the synthesis of substances possessing high pharmacological activity [29]. It is known [23, 26, 30], that thiol **7** may be found in three tautomeric forms (Scheme 5).

Scheme 5



The interaction of thiol **7** with MVK on zeolite Csβ during 60 h, unlike compound **5**, leads to the formation of products of addition of methyl vinyl ketone both at the nitrogen atom and at the sulfur atom, compounds **8** and **9** in yields of 3 and 11% respectively, and also, as the main product, of bicyclic compound **10** in 39% yield (Scheme 6). The formation of compound **8** was fairly unexpected since it is known from the

Scheme 6



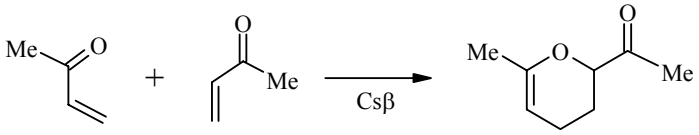
literature that base catalyzed nucleophilic addition of 1,2,4-triazole-3-thiol (**7**) and its derivatives at a carbon–nitrogen double bond proceeds at the nitrogen atom only in the presence at the double bond of a strong electron acceptor, such as a fluorine atom [31], in other cases addition occurs at the sulfur atom [32].

The first step of forming heterocyclic compound **10** is the Michael heteroreaction involving the sulfur of compound **7** at the double bond of MVK with subsequent heterocyclization, somewhat unexpected for such systems. We found no similar reactions in the literature for 1,2,4-triazole-3-thiol or its derivatives. Most likely in the presence of zeolite Csβ a fairly large proportion of thiol **7** may be found in tautomeric form **7a**, which leads to the formation of the unusual products **8** and **10**.

We attempted to carry out the interaction of thiols **5** and **7** in the presence of zeolite Csβ with a more complex α,β-unsaturated compound than MVK, *viz.* with the common monoterpenoid (-)-carvone. However even carrying out the reaction at enhanced temperature (60°C) did not lead to the formation of any products.

We note that, although storing MVK in the presence of zeolite Csβ leads to the formation of the Diels-Alder reaction product, compound **11** (Scheme 7), in low yield (17%), compound **11** was not detected by us in the reaction mixtures formed when interacting MVK with thiols **5** and **7**.

Scheme 7



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No data on the preparation of compounds **6, 8-10** were discovered by us in the literature, their structures were established with the aid of ^{1}H and ^{13}C NMR and high resolution mass spectroscopy.

EXPERIMENTAL

The ^{1}H and ^{13}C NMR spectra of the synthesized compounds were recorded on a Bruker DRX-500 instrument (500 and 126 MHz respectively), solvent was DMSO-d_6 or a $CDCl_3$–CCl_4 mixture (~1:1 by vol.), internal standard was DMSO (δ_H 2.50, δ_C 39.50 ppm) or chloroform (δ_H 7.24, δ_C 76.90 ppm). The structure of compounds was established with the aid of ^{1}H NMR spectra, including analysis of coupling constants of protons in double resonance ^{1}H–^{1}H spectra, and also analysis of ^{13}C NMR spectra recorded in the *J*-modulation mode, with extra resonance proton suppression, and two-dimensional ^{13}C–^{1}H spectra with heteronuclear correlation on the direct constants (COSY, $^{1}J_{C,H}$ = 135 Hz). Elemental composition was determined by data of mass spectra recorded on a Thermo Scientific DFS spectrometer in full scanning mode in the range 0-500 *m/z*, ionization by electron impact 70 eV with direct insertion of sample. Commercially available preparations used in experiments were methyl vinyl ketone (Merck), 5-methoxybenzimidazole-2-thiol (**5**) (Alfa Aesar), 1,2,4-triazole-3-thiol (**7**) (Merck), (*R*)-(-)-carvone (Aldrich), salicylaldehyde, and crotonaldehyde (Aldrich).

Zeolite Csβ was used as catalyst, the preparation and properties of which are described in [1].

Solvents were dried by passing through a column of calcined aluminum oxide. Separation of reaction products was carried out by column chromatography on silica gel (Merck 60-200 μ), eluent was diethyl ether in hexane, from 0 to 100%. Combination of fractions was carried out on the basis of TLC data (Sorbfil plates, silica gel STKh-1VE, size 8-12 μm, eluent hexane–ethyl acetate, 3:1).

Interaction of Salicylaldehyde and Crotonaldehyde in the Presence of Zeolite Csβ. Salicylaldehyde (0.259 g, 2.12 mmol) was added to zeolite Csβ (0.1 g). Crotonaldehyde (0.152 g, 2.17 mmol) was then added to the mixture. The reaction mixture was maintained at room temperature for 5 days, then extracted from the

catalyst with diethyl ether (20 ml). The catalyst was filtered off, and the solvent distilled. After separation by column chromatography salicylaldehyde (0.006 g, conversion 98%) and 2-methyl-2H-chromene-3-carbaldehyde (**4**) (0.182 g, 50%) were isolated.

^{1}H NMR spectrum ($CDCl_3$–CCl_4), δ, ppm (*J*, Hz): 1.35 (3H, d, $J_{11,2}$ = 6.6, 11-CH_3); 5.38 (1H, q, $J_{2,11}$ = 6.6, H-2); 6.83 (1H, br. d, $J_{9,8}$ = 8.3, H-9); 6.90 (1H, ddd, $J_{7,6}$ = 7.5, $J_{7,8}$ = 7.5, $J_{7,9}$ = 1.0, H-7); 7.13 (1H, br. s, H-4); 7.15 (1H, dd, $J_{6,7}$ = 7.5, $J_{6,8}$ = 1.7, H-6); 7.26 (1H, ddd, $J_{8,9}$ = 8.3, $J_{8,7}$ = 7.5, $J_{8,6}$ = 1.7, H-8); 9.51 (1H, s, H-12). ^{13}C NMR spectrum ($CDCl_3$–CCl_4), δ, ppm: 69.74 (d, C-2); 136.27 (s, C-3); 139.80 (d, C-4); 119.83 (s, C-5); 129.00 (d, C-6); 121.44 (d, C-7); 133.27 (d, C-8); 117.33 (d, C-9); 154.51 (s, C-10); 19.93 (q, C-11); 189.38 (d, C-12). Found, *m/z* 174.0673 $[M]^+$. $C_{11}H_{10}O_2$. Calculated: M = 174.0675.

Interaction of 5-Methoxybenzimidazole-2-thiol (5) with Methyl Vinyl Ketone in the Presence of Zeolite Csβ. 5-Methoxybenzimidazole-2-thiol (**5**) (0.309 g, 1.72 mmol) in methanol (10 ml) was added to basic zeolite Csβ (0.05 g). The solvent was distilled off. MVK (0.1 g, 1.43 mmol) was added to the mixture. The reaction mixture was maintained at room temperature for 60 h, then extracted from the catalyst with ethyl acetate (25 ml). The catalyst was filtered off and the solvent evaporated. The reaction mixture was dissolved in chloroform (20 ml) and left at 0°C for 1 day. Crystalline compound **5** (0.168 g) precipitated from the solution, and the crystals were filtered off. The solvent was distilled from the filtrate, the solid residue (0.127 g) contained a mixture of compound **5** and 4-[5-methoxy-3-(3-oxobutyl)-2-thioxo-2,3-dihydro-1H-benzimidazol-1-yl]butan-2-one (**6**) in a ratio of 1:9.4. Conversion of the initial compound **5** was 43%, yield of thione **6** on reacted thiol **5** was 50%.

^{1}H NMR spectrum ($CDCl_3$–CCl_4), δ, ppm (*J*, Hz): 2.14 and 2.15 (6H, s, 13-CH_3, 17-CH_3); 3.03 and 3.07 (4H, t, *J* = 6.7, -C<u>H</u>$_2$COCH$_3$, H-11,15); 3.84 (3H, s, 18-CH_3); 4.43 and 4.44 (4H, t, *J* = 6.7, -C<u>H</u>$_2$CH$_2$COCH$_3$, H-10,14); 6.77 (1H, dd, $J_{6,7}$ = 8.8, $J_{6,4}$ = 2.3, H-6); 6.88 (1H, d, $J_{4,6}$ = 2.3, H-4); 7.20 (1H, d, $J_{7,6}$ = 8.8, H-7). ^{13}C NMR spectrum ($CDCl_3$–CCl_4), δ, ppm: 168.52 (s, C-2); 94.92 (d, C-4); 156.85 (s, C-5); 110.31 (d, C-6); 109.97 (d, C-7); 126.07 (s, C-8); 132.62 (s, C-9); 39.09 and 39.22 (t, C-10,14); 41.16 and 41.24 (t, C-11,15); 205.43 and 205.53 (s, C-12,16); 30.12 (q, C-13,17); 55.84 (q, C-18). Found: *m/z* 320.1185 $[M]^+$. $C_{16}H_{20}N_2O_3S$. Calculated: M = 320.1189.

Interaction of Methyl Vinyl Ketone with 1,2,4-Triazole-3-thiol (7) in the Presence of Zeolite Csβ. Compound **7** (0.12 g, 1.19 mmol) was added to zeolite Csβ (0.05 g), diethyl ether (20 ml) was added to the mixture, and the suspension was stirred for 30 min. The solvent was distilled. MVK (0.08 g, 1.14 mmol) was added to the reaction mixture. The mixture was maintained at room temperature for 60 h, and extracted from the catalyst with ethyl acetate (20 ml). The catalyst was filtered off, and the solvent evaporated. After separation by column chromatography a mixture (0.08 g) containing the initial thiol **7** (0.004 g, conversion 97%) and 5-methyl-6,7-dihydro-5H-[1,2,4-triazolo[3,4-*b*][1,3]thiazin-5-ol (**10**) (0.076 g, yield on reacted MVK was 39%), and also a mixture (0.036 g) containing 4-(3-mercapto-4H-1,2,4-triazol-4-yl)butan-2-one (**8**) (0.005 g) (yield on reacted MVK 3%) and 4-([3-[(3-oxobutyl)thio]-4H-1,2,4-triazol-4-yl]butan-2-one (**9**) (yield on reacted MVK was 11%).

4-(3-Mercapto-4H-1,2,4-triazol-4-yl)butan-2-one (8). ^{1}H NMR spectrum ($CDCl_3$–CCl_4), δ, ppm (*J*, Hz): 2.13 (3H, s, 9-CH_3); 3.00 (2H, t, $J_{7,6}$ = 6.0, –C<u>H</u>$_2$COCH$_3$, H-7); 4.36 (2H, t, $J_{6,7}$ = 6.0, –C<u>H</u>$_2$CH$_2$COCH$_3$, H-6); 8.06 (1H, s, H-3). The closeness of the chemical shifts of the H-6 proton signal in compound **8** and the signals of the H-10 and H-14 protons of thione **6** indicate the addition of MVK to the nitrogen atom and not sulfur in thione **8**. Found: *m/z* 171.0462 $[M]^+$. $C_6H_9N_3OS$. Calculated: M = 171.0461.

4-[3-[(3-Oxobutyl)thio]-4H-1,2,4-triazol-4-yl]butan-2-one (9). ^{1}H NMR spectrum ($CDCl_3$–CCl_4), δ, ppm (*J*, Hz): 2.14 and 2.15 (6H, s, 9-CH_3, 13-CH_3); 2.90 (2H, t, $J_{7,6}$ = 7.0, H-7); 2.99 (2H, t, $J_{11,10}$ = 6.0, –C<u>H</u>$_2$COCH$_3$, H-11); 3.23 (2H, t, $J_{6,7}$ = 7.0, -C<u>H</u>$_2$CH$_2$COCH$_3$, H-6); 4.33 (2H, t, $J_{10,11}$ = 7.0, -C<u>H</u>$_2$CH$_2$COCH$_3$, H-10); 7.98 (1H, s, H-3). ^{13}C NMR spectrum ($CDCl_3$–CCl_4), δ, ppm: 144.71 (d, C-3); 160.65 (s, C-5); 25.50 (t, C-6); 43.76 and 43.88 (t, C-7,11); 204.38 and 205.85 (s, C-8,12); 29.90 and 29.93 (q, C-9,13); 42.24 (t, C-10). Found: *m/z* 241.088 $[M]^+$. $C_{10}H_{15}N_3O_2S$. Calculated: M = 241.088.

5-Methyl-6,7-dihydro-5H-[1,2,4]triazolo[3,4-*b*][1,3]thiazin-5-ol (10). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm (*J*, Hz): 1.76 (3H, s, 10-CH_3); 2.18 (1H, ddd, $J_{6a,6e}$ = 14.5, $J_{6a,7a}$ = 12.5, $J_{6a,7e}$ = 3.0, H-6a); 2.41 (1H, ddd, $J_{6e,6a}$ = 14.5, $J_{6e,7e}$ = 5.4, $J_{6e,7a}$ = 2.5, H-6e); 3.09 (1H, ddd, $J_{7e,7a}$ = 13.0, $J_{7e,6e}$ = 5.4, $J_{7e,6a}$ = 3.0, H-7e); 3.38 (1H, ddd, $J_{7a,7e}$ = 13.0, $J_{7a,6a}$ = 12.5, $J_{7a,6e}$ = 2.5, H-7a); 6.93 (1H, br. s, -OH); 7.87 (1H, s, H-3). ^{13}C NMR spectrum (DMSO-D_6), δ, ppm :149.90 (d, C-3); 83.14 (s, C-5); 37.35 (t, C-6); 21.05 (t, C-7); 147.16 (s, C-9); 27.65 (q, C-10). Found: *m/z* 171.0460 $[M]^+$. $C_6H_9N_3OS$. Calculated: M = 171.0461.

2-Acetyl-6-methyl-3,4-dihydro-2H-pyran (11). MVK (0.219 g, 3.13 mmol) was added to zeolite Csβ (0.047 g). The reaction mixture was maintained at room temperature for 20 h, then extracted from the catalyst with diethyl ether (15 ml). The catalyst was filtered off and the solvent distilled. 2-Acetyl-6-methyl-3,4-dihydro-2H-pyran (**11**) (0.073 g, 17%) was obtained. The ^{1}H NMR spectrum of compound **11** coincided with that described in [33].

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