

## SYNTHESIS AND STEREOCHEMISTRY OF SOME NEW BIS(1,3-DIOXANE-2-YL)ALKANES

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The synthesis, structural analysis and NMR investigations of some new 1,3-dioxane derivatives obtained by the (a)ketalization reaction of some linear dialdehydes and diphenyl-polymethylene-diketones are reported.

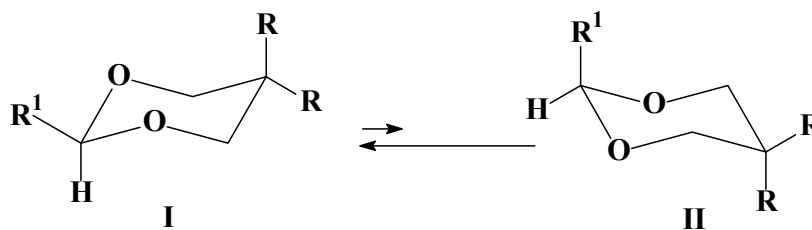
### INTRODUCTION

The conformational analysis of series of 2-substituted-1,3-dioxane derivatives<sup>1-5</sup> has pointed out the equatorial preference of the alkyl and aryl groups located in the acetal part of the 1,3-dioxacyclohexane ring. These compounds exhibit anancomeric structures and the flipping of the heterocycle is strongly shifted towards the conformer with the hydrocarbon substituents in equatorial orientation (structure I, Scheme 1).

The equatorial preference of these groups is due to their high A-values (free conformational enthalpy of the substituent).

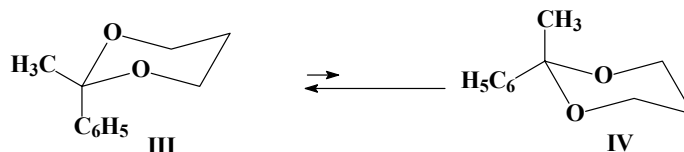
Because the A-values of alkyl groups (e.g.  $A_{Me} = 3.98$  and  $A_{i-Pr} = 4.17$  kcal/mol)<sup>5-9</sup> are higher than the A-values of the aromatic substituents (e.g.  $A_{Ph} = 3.12$  kcal/mol),<sup>5-9</sup> the derivatives bearing

simultaneously alkyl and aryl groups at position 2 of the 1,3-dioxane ring exhibit the aromatic substituent in axial orientation. The thermodynamic measurements revealed that the axial preference of the aryl group is considerably higher than it should be considered by the simple addition of the individual A values of the alkyl and aryl substituents. For instance the conformational equilibrium of 2-methyl-2-phenyl-1,3-dioxane (Scheme 2; III, IV) is shifted towards the conformer with the phenyl group in axial orientation (structure III) and the measured modification of free enthalpy  $\Delta G^{\circ}_{exp}$  is about three times higher than the calculated value  $\Delta G^{\circ}_{calcd}$  ( $\Delta G^{\circ}_{exp} = 2.42$  kcal/mol;  $\Delta G^{\circ}_{calcd} = A_{Me} - A_{Ph} = 0.86$  kcal/mol).<sup>5</sup>



Scheme 1

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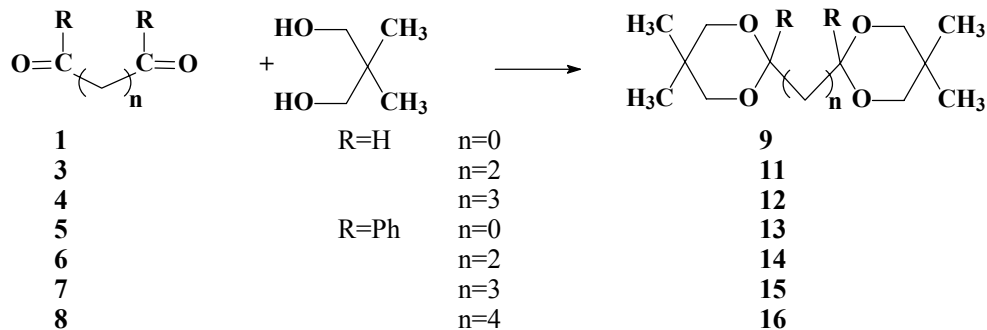
Scheme 2

In previous works<sup>10-16</sup> we revealed the conformational behavior of 1,3-dioxane rings in derivatives bearing two or three 1,3-dioxacyclohexane units (1,3-dioxan-2-yl groups) connected at the same aromatic unit and we observed that the axial or equatorial preference of the aromatic rings are correlated, in these cases too, with the presence or the missing of the alkyl substituent in geminal position with the aromatic unit. We considered of interest to obtain new 1,3-dioxane derivatives bearing two heterocycles at the extremities of an alkyl chain and to investigate the conformational behavior of these compounds in correlation both, with the type and number of substituents located at the positions 2 (2') of the heterocycles and with the length of the polymethylene chain.

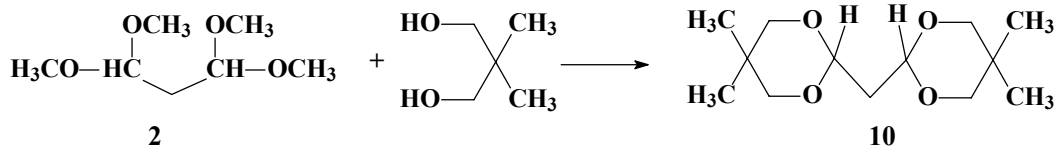
## RESULTS AND DISCUSSION

Bis(1,3-dioxane-2-yl) derivatives **9-16** were obtained by the direct condensation reaction of several linear dialdehydes and diphenylpolymethylene-diketones with neopentylglycol (Scheme 3) or by the transacetalization reaction of the tetramethylacetal of malonic dialdehyde (**2**) with the same diol (Scheme 4).

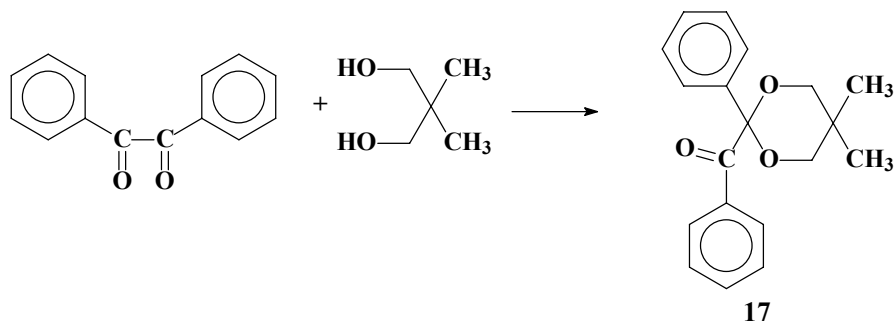
The reactions underwent in fair or good yields (41 – 60 %); compounds **11**, **12** and **14-16** are new ones, while the obtaining of derivatives **9**, **10** and **13** was already reported.<sup>17-19</sup> In the synthesis of **13** the investigation of the crude product showed the formation of important amounts (yield 13%) of monocarbonyl derivative **17** (Scheme 5) and this new compound could be isolated and investigated.



Scheme 3



Scheme 4



Scheme 5

The structural investigations of compounds **9-17** were carried out using NMR spectra ( $^1\text{H}$  and  $^{13}\text{C}$ ) and the determination of the molecular structure of **17** by single crystal X-ray diffractometry.

The molecular structure of **17** (Figure 1) reveals the axial orientation of the CO-Ph substituent and the peculiar rotameric behavior of the substituents at position 2. In the axial -CO-Ph group a torsion angle of  $\alpha_1 = 17.7^\circ$  between the planes of the carbonyl and aromatic groups could be noted, while the aromatic group of this benzoyl substituent exhibits a dihedral angle of  $\alpha_2 = 72.5^\circ$  with the best plane (described by the exocyclic bonds of positions 2 or 5 of the heterocycle) of the 1,3-dioxane ring, being almost in the orthogonal rotamer (which shows a characteristic dihedral angle of  $90^\circ$ ). The other phenyl group exhibits a rotameric behavior which is intermediary (the dihedral angle with the same best plane is  $\alpha_3 = 52.3^\circ$ ) to the bisectonal ( $\alpha = 0^\circ$ ) and orthogonal ( $\alpha = 90^\circ$ ) rotamers. This peculiar arrangement of the aromatic groups is the result of several intra- and intermolecular CH- $\pi$  and  $\pi$ - $\pi$  stacking interactions (Figure 2). The formation of dimeric units in the lattice (e.g. molecules V and VI; Figure 2) is observed. The association of the molecules in the dimers are explained by the  $\pi$ - $\pi$  stacking between the aromatic parts of the -CO-Ph moieties (they show a **face-to-face** arrangement, the distance between the centroids of the two rings is 3.854 Å) and by the CH- $\pi$  interactions between the *para* proton of the  $\text{C}_6\text{H}_5$  part of the benzoyl group and the equatorial phenyl group at position 2. These two rings show an **edge tilted to face**

arrangement (dihedral angle of  $75.9^\circ$ ) and the distance from the hydrogen atom of the *para* position to the centroid of the opposite aromatic group is 3.409 Å. The dimeric units are connected one to the other by CH- $\pi$  interactions between the 2-Ph group of one of the molecules of the dimer (V in Figure 2) and the aromatic part of the benzoyl group of a molecule belonging to another dimer unit (VII in Figure 2; the second molecule pertaining to this dimer unit is not shown). The distance from the *para* hydrogen atom to the centroid of the aromatic ring of the  $\text{C}_6\text{H}_5\text{CO}$  unit is 3.161 Å, the arrangement of the aromatic rings is **edge tilted to face** and the dihedral angle between the considered aromatic rings is  $75.3^\circ$ . In figure 2, one can also observe the intramolecular CH- $\pi$  interaction with the participation of one of the *ortho* hydrogen atoms of the CO-Ph group and the other equatorial Ph group. The arrangement of the two aromatic groups is **edge tilted to face**, the distance from the *ortho* hydrogen atom to the centroid of the Ph substituent is 3.534 Å and the dihedral angle between the two aromatic groups is  $75.8^\circ$ .

Compound **17** exhibits in solution an anancomeric structure and the conformational equilibrium is shifted towards the conformer with the benzoyl group in axial orientation (VIII, Scheme 6). This conformational preference is due in one hand to the smaller hindrance of the axial -CO-Ph group compared with the phenyl group in the axial 2 position and on the other hand to the anomeric effect of the axial carbonyl substituent.

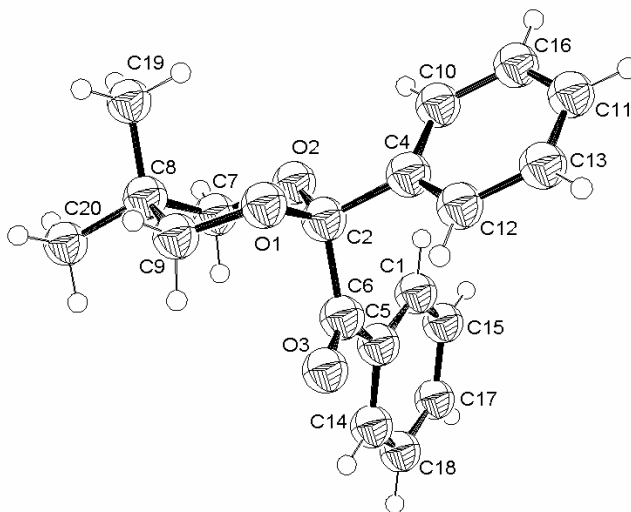


Fig. 1 – ORTEP diagram of compound **17**.

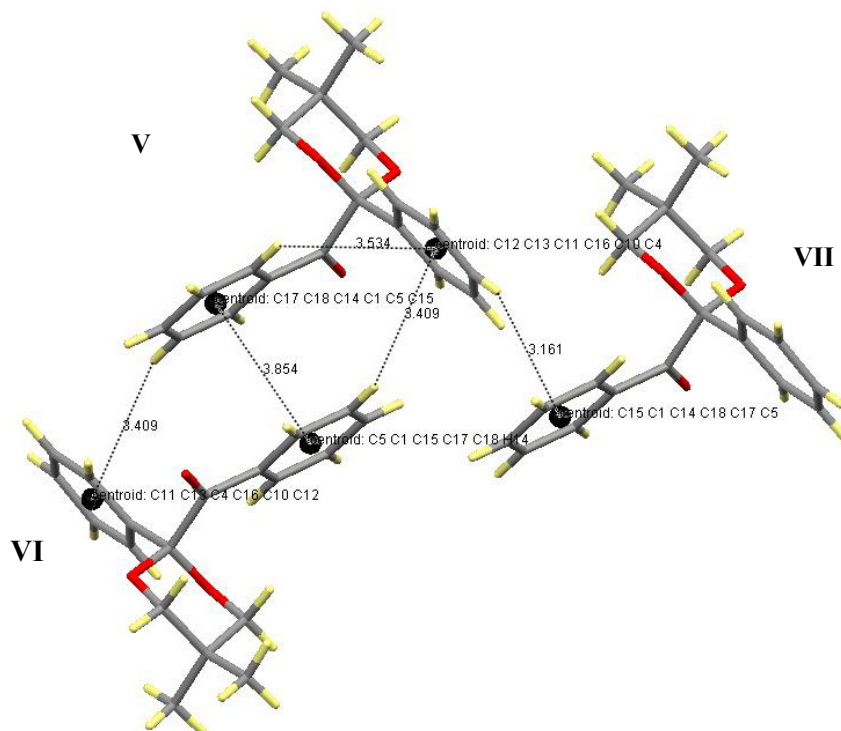
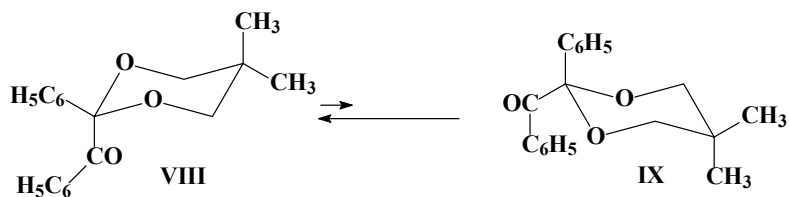
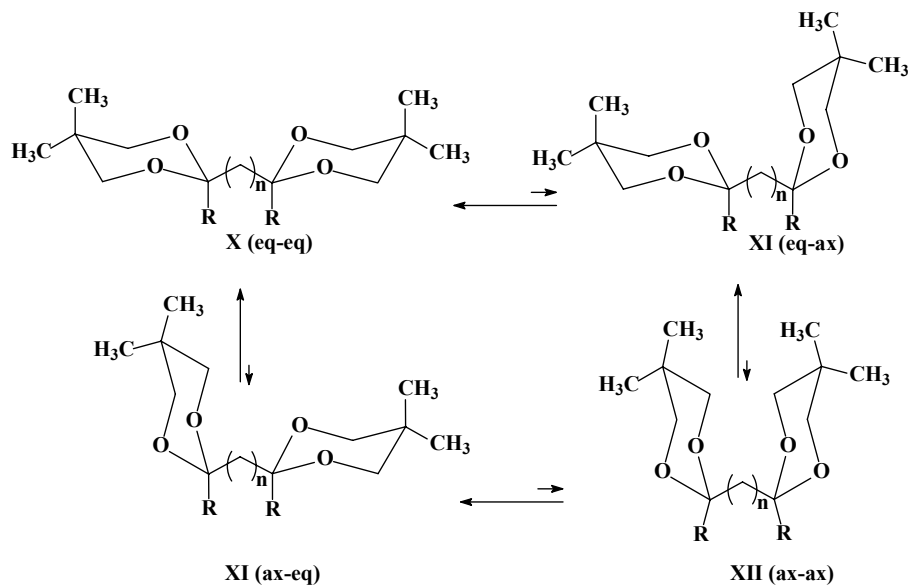


Fig. 2 – Representation of the lattice and of the stacking interactions of compound 17 (Mercury program).



Scheme 6



Scheme 7

The bis(1,3-dioxan-2yl) derivatives exhibit as well anancomeric structures and in both cases (Scheme 7; compounds obtained from dialdehydes **9-12** or from diketones **13-16**) the polymethylene chains prefer the equatorial positions. These compounds can show three isomers (conformers X-XII) but the conformational equilibria are shifted towards the conformer (X) in which the alkylidene substituent exhibits the equatorial orientation for both 1,3-dioxane rings. The conformational behavior of the molecules is not significantly influenced by the distance between

the heterocycles (chain's length). In the derivatives in which the two heterocycles are directly connected one to the other ( $n=0$ ), the heterocycles exhibit equatorial orientations.

As a consequence of the similar conformational behavior of the two heterocycles the NMR spectra (Table 1) exhibit only one set of signal and due to the anancomericity of the structures these spectra display different signals for the protons of the axial and equatorial positions of the 1,3-dioxane rings and for the protons or carbon atoms of the axial and equatorial methyl groups at positions 5 (5').

Table 1

<sup>1</sup>H RMN data for compounds **9-17** ( $\delta$  values in ppm, coupling constant <sup>2</sup> $J_{ax-eq}$  in Hz) in CDCl<sub>3</sub> or C<sub>6</sub>D<sub>6</sub>\*

Compound	$\delta\text{CH}_{3eq}$	$\delta\text{CH}_{3ax}$	$\Delta\delta_{ax-eq}$	$\delta\text{H}_{4(6)ax}$	$\delta\text{H}_{4(6)eq}$	$\Delta\delta\text{H}_{4(6)eq-ax}$	<sup>2</sup> $J_{ax-eq}$
<b>9*</b>	0.21	1.17	0.96	3.13	3.46	0.33	10.5
<b>10</b>	0.71	1.18	0.47	3.43	3.59	0.16	10.5
<b>11</b>	0.71	1.18	0.47	3.41	3.60	0.19	10.8
<b>12</b>	0.70	1.17	0.47	3.39	3.73	0.18	10.7
<b>13</b>	0.26	0.40	0.14	3.20	3.40	0.20	11.0
<b>14</b>	0.54	1.22	0.68	3.23	3.33	0.10	10.7
<b>14*</b>	0.08	1.13	1.05	3.20	3.34	0.14	10.9
<b>15</b>	0.54	1.21	0.67	3.17	3.30	0.13	10.7
<b>15*</b>	0.14	1.25	1.11	3.23	3.37	0.14	10.7
<b>16</b>	0.54	1.20	0.66	3.32	3.40	0.08	10.7
<b>17</b>	0.79	1.23	0.44	3.63	3.73	0.10	11.0

The analysis of the NMR data points out the shielding through space of the protons of the equatorial methyl group at position 5 and of the protons of positions 4 and 6 in compounds **13-16** as a result of the influence of the axial orthogonal phenyl ring at position 2 (similar situations were observed on other derivatives with axial aromatic groups at position 2).<sup>20</sup> The <sup>13</sup>C-NMR spectra exhibit different signals for the axial and equatorial methyl groups at position 5. If the spectra of compounds **14-16** are compared with the spectra of **9-12** one can observe the shielding effect produced by axial orthogonal phenyl group ( $\Delta\delta$  of 4-5 ppm) on the carbon atoms of positions 4 and 6.

## EXPERIMENTAL

**General remarks:** <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded at room temperature using C<sub>6</sub>D<sub>6</sub> (CDCl<sub>3</sub>) as solvents in 5 mm

tubes on a Varian Gemini 300 NMR spectrometer equipped with a dual <sup>13</sup>C-<sup>1</sup>H (multinuclear) head operating at 300 MHz for protons and 75 MHz for carbon atoms at "Babes-Bolyai" University Cluj-Napoca, Roumania. Melting points were measured with an APOTEC melting point apparatus and are uncorrected. Elemental analyses were obtained at the University of Medicine and Pharmaceutics, Cluj-Napoca. Their results agreed favorably with the calculated values. Thin-layer chromatography was performed on Merck silica gel 60 F 254. Silica gel Merck (40-63  $\mu\text{m}$ ) was used for flash chromatography.

### General procedure for the synthesis of compounds

**Compounds 9, 11-17:** To a solution of 10 mmol of the corresponding dicarbonyl derivative in 50 ml of toluene were added 22 mmol of neopentylglycol and 0.05 g of p-toluenesulphonic acid as catalyst. The mixture was refluxed and the water resulted in the reaction was removed using a Dean-Stark trap. When the theoretical water was separated, after cooling at room temperature, the catalyst was neutralized (under stirring) with CH<sub>3</sub>COONa powder in excess (0.5 g). The reaction mixture was washed twice with 20 ml water. The organic layer was dried (with Na<sub>2</sub>SO<sub>4</sub>), then toluene was

removed and the 1,3-dioxane derivatives were purified by crystallization.

**Compound 10:** To a solution of 10 mmol of malonaldehyde-bis(dimethylacetal) in 50 ml of toluene were added 22 mmol of neopentylglycol and 0.05 g of *p*-toluenesulphonic acid as catalyst. The mixture was heated on a glycerin bath under magnetic stirring at 80–90°C and the resulted methanol was distilled off. When the theoretical methanol was removed, the reaction mixture was stirred for further 15 min. After cooling at room temperature, the catalyst was neutralized (under stirring for 30 min) with CH<sub>3</sub>COONa powder in excess (0.5 g). The reaction mixture was washed twice with 20 ml water. The organic layer was dried (with Na<sub>2</sub>SO<sub>4</sub>), then toluene was removed and the 1,3-dioxane derivative was purified by crystallization.

*1,2-bis(5,5-dimethyl-1,3-dioxane-2-yl)-ethane (11)*

Yield 41%; m.p.=74–74.5°C (from ethanol). Found: C 64.86; H 10.05; C<sub>14</sub>H<sub>26</sub>O<sub>4</sub> requires: C 65.09; H 10.14.

<sup>1</sup>H-NMR (CDCl<sub>3</sub>, δ, ppm, *J*, Hz): 0.71 [s, 6H, 5'(5'')-CH<sub>3</sub>eq]; 1.18 [s, 6H, 5'(5'')-CH<sub>3</sub>ax]; 1.76–1.78 (m, 4H, 1,2-H<sub>2</sub>); 3.41 [d, 4H, 10.8; 4'(4''), 6'(6'')-H<sub>ax</sub>]; 3.60 [d, 4H, 10.8; 4'(4''), 6'(6'')-H<sub>eq</sub>]; 4.51 [m, 2H, 2'(2'')-H<sub>ax</sub>].

<sup>13</sup>C-NMR (CDCl<sub>3</sub>, δ, ppm): 21.9 [5'(5'')-CH<sub>3</sub>eq]; 23.0 [5'(5'')-CH<sub>3</sub>ax]; 29.1 (C-1,2); 30.2 [C-5'(5'')]; 77.2 [C-4'(4''), 6'(6'')]; 101.7 [C-2'(2'')].

*1,3-bis(5,5-dimethyl-1,3-dioxane-2-yl)-propan (12)*

Yield 60%; m.p.=69.5–70.5°C (from ethanol). Found: C 66.26; H 10.21; C<sub>15</sub>H<sub>28</sub>O<sub>4</sub> requires: C 66.14; H 10.36.

<sup>1</sup>H-NMR (CDCl<sub>3</sub>, δ, ppm, *J*, Hz): 0.70 [s, 6H, 5'(5'')-CH<sub>3</sub>eq]; 1.17 [s, 6H, 5'(5'')-CH<sub>3</sub>ax]; 1.54 (m, 2H, 2-H<sub>2</sub>), 1.64 (m, 4H, 1,3-H<sub>2</sub>), 3.39 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>ax</sub>]; 3.57 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>eq</sub>]; 4.39 [m, 2H, 2'(2'')-H<sub>ax</sub>].

<sup>13</sup>C-NMR (CDCl<sub>3</sub>, δ, ppm): 18.7 (C-2); 21.9 [5'(5'')-CH<sub>3</sub>eq]; 23.0 [5'(5'')-CH<sub>3</sub>ax]; 30.2 [C-5'(5'')]; 34.6 (C-1,3); 76.6 [C-4'(4''), 6'(6'')]; 102.1 [C-2'(2'')].

*1,2-bis(2-phenyl-5,5-dimethyl-1,3-dioxane-2-yl)-ethane (14)*

Yield 53%; m.p.=199–200°C (from ethanol) Found: C 76.22; H 8.41; C<sub>26</sub>H<sub>34</sub>O<sub>4</sub> requires: C 76.09; H 8.29.

<sup>1</sup>H-NMR (CDCl<sub>3</sub>, δ, ppm, *J*, Hz): 0.54 [s, 6H, 5'(5'')-CH<sub>3</sub>eq]; 1.22 [s, 6H, 5'(5'')-CH<sub>3</sub>ax]; 1.97 [s, 4H, 1,2-H<sub>2</sub>]; 3.23 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>ax</sub>]; 3.33 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>eq</sub>]; 7.27–7.79 (overlapped signals, 10H, aromatic protons).

<sup>13</sup>C-NMR (C<sub>6</sub>D<sub>6</sub>, δ, ppm): 21.6 [5'(5'')-CH<sub>3</sub>eq]; 23.3 [5'(5'')-CH<sub>3</sub>ax]; 30.0 [C-5'(5'')]; 39.1 (C-1,2); 71.6 [C-4'(4''), 6'(6'')]; 101.4 [C-2'(2'')]; 127.0–129.0 (aromatic carbon atoms).

*1,3-bis(2-phenyl-5,5-dimethyl-1,3-dioxane-2-yl)-propane (15)*

Yield 42%; m.p.=108–112°C (from hexane). Found: C 76.28; H 8.53; C<sub>27</sub>H<sub>36</sub>O<sub>4</sub> requires: C 76.41; H 8.49.

<sup>1</sup>H-NMR (C<sub>6</sub>D<sub>6</sub>, δ, ppm, *J*, Hz): 0.14 [s, 6H, 5'(5'')-CH<sub>3</sub>eq]; 1.25 [s, 6H, 5'(5'')-CH<sub>3</sub>ax]; 1.93 (m, 4H, 2-H<sub>2</sub>); 2.08 (m, 2H, 1,3-H<sub>2</sub>); 3.23 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>ax</sub>]; 3.37 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>eq</sub>]; 7.10–7.50 (overlapped signals, 10H, aromatic protons).

<sup>13</sup>C-NMR (C<sub>6</sub>D<sub>6</sub>, δ, ppm): 17.5 (C-2); 21.70 [5'(5'')-CH<sub>3</sub>eq]; 23.2 [5'(5'')-CH<sub>3</sub>ax]; 30.0 [C-5'(5'')]; 45.3 (C-1,3); 71.6 [C-4'(4''), 6'(6'')]; 102.1 [C-2'(2'')]; 127.7; 127.8; 128.1; 128.4; 128.7; 141.2 (aromatic carbon atoms).

*1,4-bis(2-phenyl-5,5-dimethyl-1,3-dioxane-2-yl)-butane (16)*

Yield 47%; m.p.=163–165°C (from dichloromethane). Found: C 76.84; H 8.85; C<sub>28</sub>H<sub>38</sub>O<sub>4</sub> requires: C 76.71; H 8.67.

<sup>1</sup>H-NMR (CDCl<sub>3</sub>, δ, ppm, *J*, Hz): 0.54 [s, 6H, 5'(5'')-CH<sub>3</sub>eq]; 1.20 [s, 6H, 5'(5'')-CH<sub>3</sub>ax]; 1.27 (m, 4H, 1,4-H<sub>2</sub>); 1.70 (m, 4H,

2,3-H<sub>2</sub>); 3.32 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>ax</sub>]; 3.40 [d, 4H, 10.7; 4'(4''), 6'(6'')-H<sub>eq</sub>]; 7.33 (overlapped signals, 10H, aromatic protons).

<sup>13</sup>C-NMR (CDCl<sub>3</sub>, δ, ppm): 21.9 [5'(5'')-CH<sub>3</sub>eq]; 23.0 [5'(5'')-CH<sub>3</sub>ax]; 30.1 [C-5'(5'')]; 44.6 (C-1,2,3,4); 71.5 [C-4'(4''), 6'(6'')]; 101.6 [C-2'(2'')]; 127.4; 127.5; 128.3; 139.9 (aromatic carbon atoms).

*2-phenyl-2-benzyloxy-5,5-dimethyl-1,3-dioxane (17)*

Yield 13%; m.p.=135°C (from ethanol). Found: C 76.83; H 6.88; C<sub>19</sub>H<sub>20</sub>O<sub>3</sub> requires: C 77.00; H 6.80.

<sup>1</sup>H-NMR (CDCl<sub>3</sub>, δ, ppm, *J*, Hz): 0.78 (s, 3H, 5-CH<sub>3</sub>eq); 1.23 (s, 3H, 5-CH<sub>3</sub>ax); 3.63 (d, 2H, 11.0; 4, 6-H<sub>ax</sub>); 3.73 (d, 2H, 11.0; 4, 6-H<sub>eq</sub>); 7.31–8.10 (overlapped signals, 10H, aromatic protons).

<sup>13</sup>C-NMR (CDCl<sub>3</sub>, δ, ppm): 22.1 (5-CH<sub>3</sub>); 30.1 (C-5); 73.5 (C-4,6); 101.9 (C-2); 126.1; 128.2; 128.7; 129.1; 130.2; 133.1; 134.5; 138.0 aromatic carbon atoms); 197.0 (C=O).

## CONCLUSIONS

The NMR investigations of compounds bearing two 1,3-dioxane units connected *via* a polymethylene chain reveal the similar conformational behavior of the two heterocycles. The compounds are anancomeric and the alkyldene chains or the 1,3-dioxan-2-yl group (n=0), in the compounds obtained from α-dicarbonyl compounds, prefer the equatorial orientation. The investigations of the mono 1,3-dioxane derivative of benzil show the axial orientation of the benzoyl group in solid state and in solution, while the X-ray diffractometry solid state structure determination reveals for this compound the peculiar rotameric behavior of the aromatic rings and the influence of the CH-π and π-π interactions on the packing of the molecules in the lattice.

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