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Study of the methylation reaction of 2,4-dinitroimidazole and potassium 2,4,5-trinitroimidazol-1-ide with dimethyl sulfate

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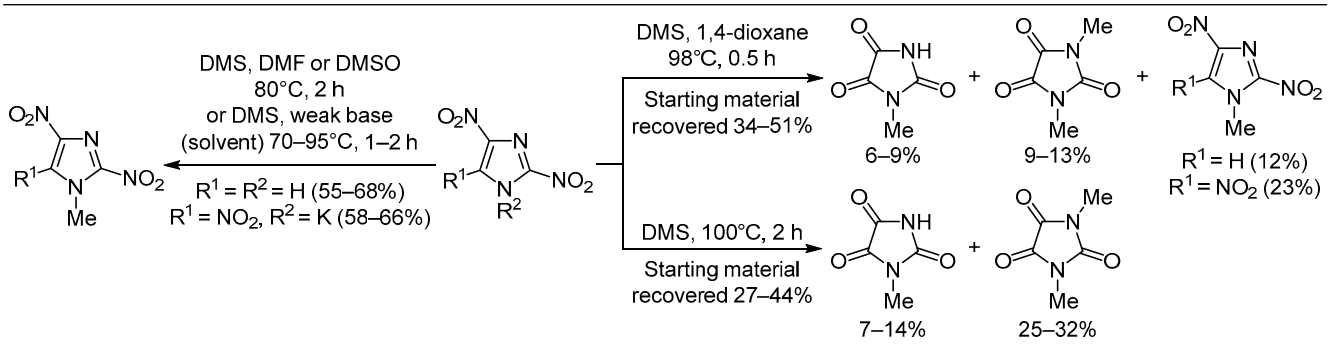


The methylation of 2,4-dinitroimidazole and potassium salt of 2,4,5-trinitroimidazole with dimethyl sulfate was reinvestigated. When the reaction system contained a weak base, the reaction products were 1-methyl-2,4-dinitroimidazole and 1-methyl-2,4,5-trinitroimidazole, respectively. However, in the absence of a base in the reaction system, the products contained oxidation byproducts 1-methylimidazolidine-2,4,5-trione, 1,3-dimethylimidazolidine-2,4,5-trione and recovered starting materials. All products were characterized using FT-IR, NMR, and elemental analysis. The structure of 1-methylimidazolidine-2,4,5-trione and 1,3-dimethylimidazolidine-2,4,5-trione were further confirmed by single crystal X-ray diffraction.

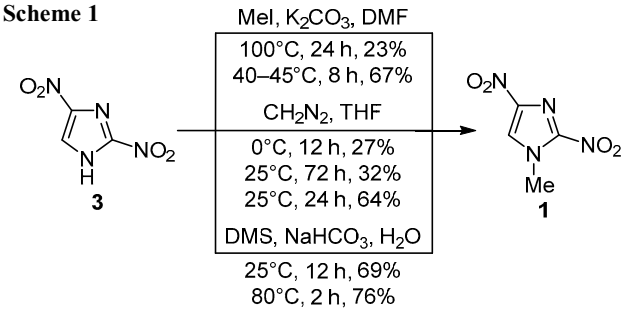
Keywords: 1,3-dimethylimidazolidine-2,4,5-trione, 2,4-dinitroimidazole, 1-methyl-2,4-dinitroimidazole, 1-methylimidazolidine-2,4,5-trione, 1-methyl-2,4,5-trinitroimidazole, potassium 2,4,5-trinitroimidazol-1-ide, methylation reaction.

In the past few decades, polynitroimidazoles and their derivatives have been widely used as antitumor, antibacterial, antifungal, antiviral drugs.[1–4] In recent years, polynitroimidazoles have attracted increased interest as energetic materials, especially high-energy insensitive energetic materials.[5–13] 1-Methyl-2,4-dinitroimidazole (**1**) and 1-methyl-2,4,5-trinitroimidazole (**2**) are two important nitroimidazole compounds, which can be prepared from 2,4-dinitroimidazole (**3**) and potassium 2,4,5-trinitroimidazol-1-ide (**4**), respectively, by reaction with methylation reagents (such as MeI, CH_2N_2, dimethyl sulfate (DMS), Schemes 1 and 2).[10–14] Meanwhile, compound **1** is also an important precursor for the preparation of compound **2**.[10b,c]

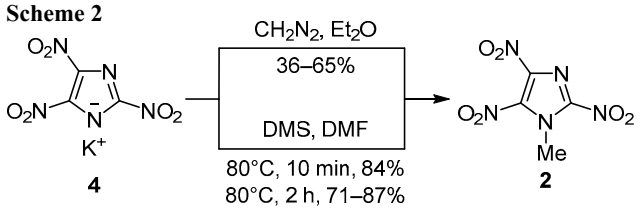
The methylation reaction of compound **3** with MeI,[12] CH_2N_2,[12a,14] or DMS[10c,13,14b] was used to synthesize compound **1** (Scheme 1). The yields of compound **1** were 23–67%, 27–64%, and 69–76%, respectively. The methylation reaction of compound **4** with CH_2N_2[10] or DMS[11] afforded compound **2** as product (Scheme 2), the yields were 36–65% and 71–87%, respectively.

From these data it is clear that the highest yields of compounds **1** and **2** were obtained using DMS as methylation agent. Since the reaction conditions used as well as the yields differ in the published works, we decided

Scheme 1



Scheme 2



to determine the optimal methylation conditions for compounds **3** and **4** in DMS in this study.

In our initial experiment, we conducted methylation of compound **3** with a minimal excess of DMS in 1,4-dioxane according to Sudarsanam,[12a] however, it was surprising that the mixture of reaction products contained the desired compound **1** in only 5% yield, along with 1-methylimidazolidine-2,4,5-trione (**5**) (3%), 1,3-dimethylimidazolidine-2,4,5-trione (**6**) (3%), and much of unreacted starting material (compound **3** (79%), Scheme 3, Table 1, entry 1). The possible mechanism for the transformation of compound **3** into compound **5** is shown in Scheme 4.[15] Furthermore, as the amount of DMS increased, the yield of compound **1** was further increased up to 12% (entries 2–5), but even if the molar ratio of DMS to compound **3** was 20:1, a significant amount of starting material **3** was recovered after heating the reaction mixture for 2.5 h (entry 5). The methylation reaction of compound **3** with DMS without any solvent was also examined (Table 1, entries 6–10), and the products consisted of unreacted starting material **3** (44–73%), compounds **5** (5–7%) and **6** (7–25%), but the methylation product **1** was not isolated. Similarly, as the amount of DMS increased, the yield of compounds **5** and **6** was also increased. Under the same reaction conditions, the product composition did not change with increasing reaction time (Table 1, entries 4, 5, 9, 10).

Our next step was carrying out the reaction in DMF or DMSO or with addition of a weak base to avoid possible acidic conditions in the reaction medium. In this case, the only reaction product was *N*-methyl derivative **1** (Table 1, entries 11–17).

Furthermore, the methylation reaction of compound **4** with DMS was also investigated (Scheme 5). The results were analogous to those obtained with compound **3**. The data for the methylation reaction of compound **4** with DMS are summarized in Table 2.

The structure of compounds **5** and **6** was finally confirmed by a single crystal X-ray study. The single

Scheme 3

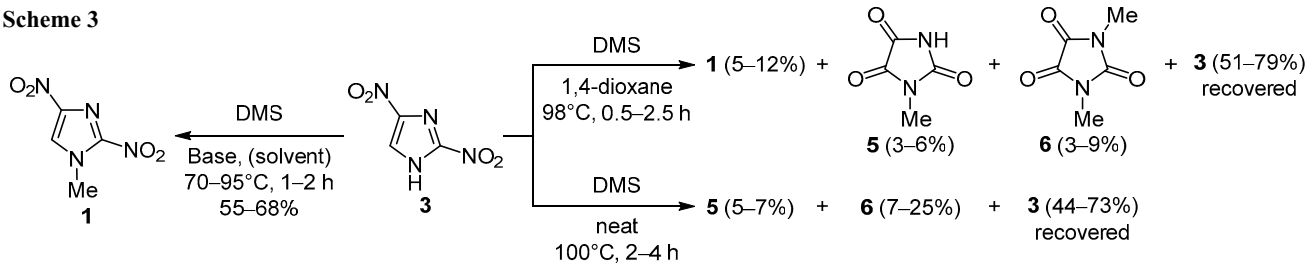
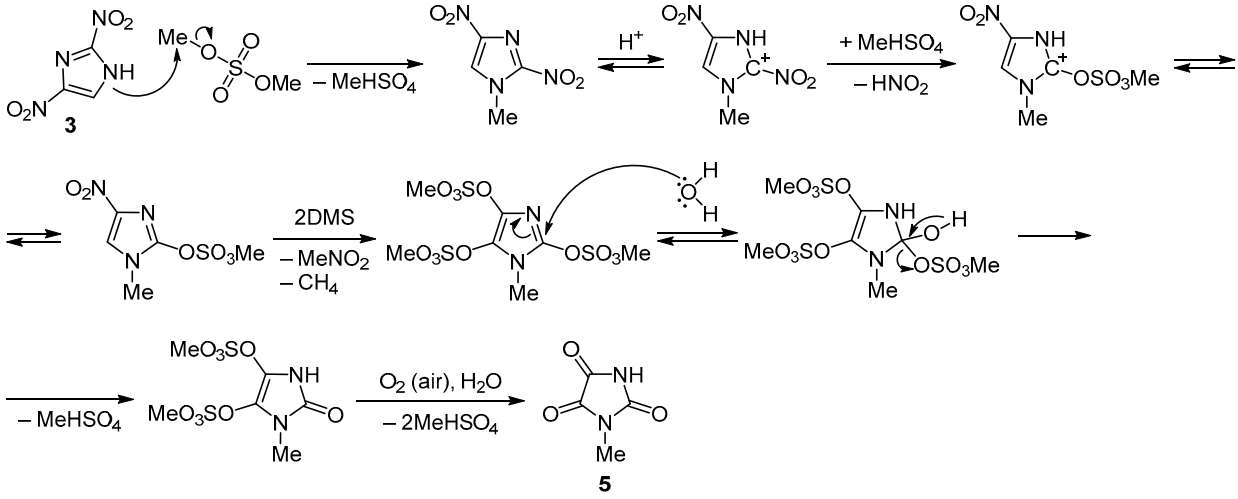


Table 1. Summary of the data for the methylation of 2,4-dinitroimidazole (**3**) with DMS*

Entry	DMS, ml (mol)	Solvent, ml	Base (amount)	Temperature, °C	Time, h	Yield**, %			
						1	**3**	**5**	**6**
1	1.0 (0.01)	1,4-Dioxane, 30	–	98	0.5	5	79	3	3
2	1.9 (0.02)	1,4-Dioxane, 30	–	98	0.5	8	70	3	4
3	9.5 (0.10)	1,4-Dioxane, 30	–	98	0.5	10	63	5	7
4	19.0 (0.20)	1,4-Dioxane, 30	–	98	0.5	12	51	6	9
5	19.0 (0.20)	1,4-Dioxane, 30	–	98	2.5	12	51	6	9
6	1.0 (0.01)	–	–	100	2.0	0	73	5	7
7	1.9 (0.02)	–	–	100	2.0	0	64	6	10
8	9.5 (0.10)	–	–	100	2.0	0	55	7	19
9	19.0 (0.20)	–	–	100	2.0	0	44	7	25
10	19.0 (0.20)	–	–	100	4.0	0	44	7	25
11	1.9 (0.02)	DMF, 10	–	80	2.0	62	0	0	0
12	1.9 (0.02)	DMF, 10	K_2CO_3 (2.76 g)	80	2.0	62	0	0	0
13	1.9 (0.02)	DMSO, 10	–	80	2.0	55	0	0	0
14	1.9 (0.02)	H_2O, 20	NaOH (0.80 g)	70	1.5	66	0	0	0
15	1.9 (0.02)	–	Et_3N (30 ml)	83	1.0	68	0	0	0
16	1.9 (0.02)	EtOH, 30	25–28% aq NH_3 (1.5 ml)	75	2.0	68	0	0	0
17	1.9 (0.02)	1,4-Dioxane, 30	K_2CO_3 (2.76 g)	95	2.0	64	0	0	0

* The amount of starting compound **3** is 10.0 mmol in all cases.
** Yield of the isolated product.

Scheme 4



Scheme 5

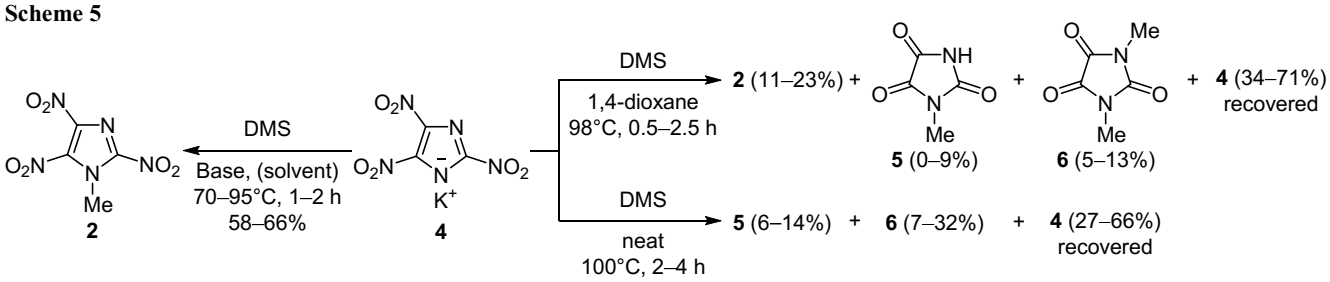


Table 2. Summary of the data for the methylation of potassium 2,4,5-trinitroimidazol-1-ide (**4**) with DMS

Entry	DMS, ml (mol)	Solvent, ml	Base (amount)	Temperature, °C	Time, h	Yield**, %			
						2	**4**	**5**	**6**
1	1.0 (0.01)	1,4-Dioxane, 30	–	98	0.5	11	71	4	5
2	1.9 (0.02)	1,4-Dioxane, 30	–	98	0.5	16	58	8	10
3	9.5 (0.10)	1,4-Dioxane, 30	–	98	0.5	21	42	8	11
4	19.0 (0.20)	1,4-Dioxane, 30	–	98	0.5	23	34	9	13
5	19.0 (0.20)	1,4-Dioxane, 30	–	98	2.5	23	34	0	13
6	1.0 (0.01)	–	–	100	2.0	0	66	6	7
7	1.9 (0.02)	–	–	100	2.0	0	53	9	11
8	9.5 (0.10)	–	–	100	2.0	0	40	12	25
9	19.0 (0.20)	–	–	100	2.0	0	27	14	32
10	19.0 (0.20)	–	–	100	4.0	0	27	14	32
11	1.9 (0.02)	DMF, 10	–	80	2.0	58	0	0	0
12	1.9 (0.02)	DMF, 10	K_2CO_3 (2.76 g)	80	2.0	60	0	0	0
13	1.9 (0.02)	DMSO, 10	–	80	2.0	58	0	0	0
14	1.9 (0.02)	H_2O, 20	NaOH (0.80 g)	70	1.5	63	0	0	0
15	1.9 (0.02)	–	Et_3N (30 ml)	83	1.0	64	0	0	0
16	1.9 (0.02)	EtOH, 30	25–28% aq NH_3 (1.5 ml)	75	2.0	66	0	0	0
17	1.9 (0.02)	1,4-Dioxane, 30	K_2CO_3 (2.76 g)	95	2.0	60	0	0	0

* The amount of starting compound **4** is 10.0 mmol in all cases.
** Yield of the isolated product.

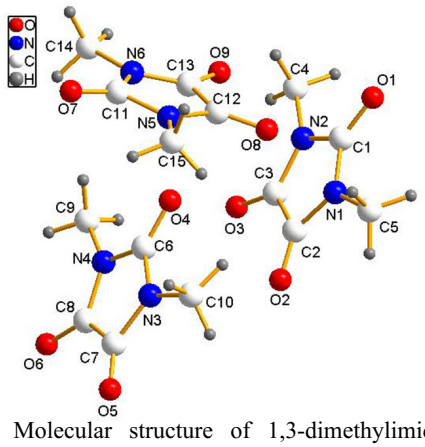


Figure 1. Molecular structure of 1,3-dimethylimidazolidine-2,4,5-trione (**6**) according to X-ray diffraction analysis.

crystal data and structure image of compound **5** are shown in Tables 1S–5S and Figure 1S (Supplementary information file), respectively, which are identical to that reported in our previous article.[16]

Compound **6** crystallizes in the orthorhombic crystal system with space group $Pna2_1$, and each elementary cell has twelve molecules. The main crystallographic data are listed in Table 6S (Supplementary information file). As shown in Figure 1, the asymmetric unit of compound **6** consists of three molecules, and each of the imidazolidine rings is almost planar with the dihedral angles 0.904, 1.362, and 2.037°, respectively (Table 7S, Supplementary information file). The keto groups and the carbon atoms of the methyl groups are almost coplanar with the imidazolidine ring; for the respective torsion angles (see Table 8S, Supplementary information file). However, hydrogen atoms of the methyl groups are rotated out of the plane.

In summary, we have studied the methylation reactions of 2,4-dinitroimidazole with DMS in 1,4-dioxane, which produced 1-methyl-2,4-dinitroimidazole in low yield along with 1-methylimidazolidine-2,4,5-trione and 1,3-dimethylimidazolidine-2,4,5-trione as oxidation byproducts, as well as unreacted starting material. When 2,4-dinitroimidazole reacted with DMS without solvent, no methylation took place, and only the oxidation byproducts were formed. The reaction of 2,4-dinitroimidazole with DMS in DMF, DMSO, low concentration sodium hydroxide solution or weak base allowed to obtain 1-methyl-2,4-dinitroimidazole in moderate to good yield without side products. Similar results were observed for the methylation of potassium 2,4,5-trinitroimidazol-1-ide with DMS. The structure of products of these reactions indicate that 2,4-dinitro-1-methylimidazole and 1-methyl-2,4,5-trinitroimidazole are susceptible to nucleophilic substitution to form methylimidazolidine-2,4,5-triones.

Experimental

IR spectra were recorded on a Bruker Model Vertex 80 FTS spectrometer in KBr pellets. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Model Avance spectrometer (400 and 100 MHz, respectively) in Me_2CO-d_6 or DMSO-d_6. Chemical shift values (δ) are reported in ppm relative to Me_2CO-d_6 or DMSO-d_6 as internal standard (2.05 or 2.50, and 29.8 or 39.5 ppm for ^{1}H and ^{13}C nuclei, respectively). Elemental analyses were taken on an Elementar Vario EL elemental analyzer. Melting points were collected using a Buchi Melting Point M-565 apparatus and are uncorrected.

Dimethyl sulfate (98.5%), DMF (99.5%), DMSO (99.0%), 1,4-dioxane (99.0%, H_2O 0.1%), 25–28% aqueous NH_3, Et_3N (99.0%), EtOH (99.7%), NaOH (96.0%), K_2CO_3 (99.0%), KCl (99.5%), Et_2O (99.5%), and CH_2Cl_2 (99.5%) were provided by Xi Long Science Company. Reagent grade (Type 3, 300–400 mesh) silica gel was used, which was purchased from Qingdao Haiyang Chemical Co., Ltd. 2,4-Dinitroimidazole (**3**)[17] and potassium 2,4,5-trinitroimidazol-1-ide (**4**)[10a] were prepared according to the published procedure by the North University of China.

Caution! Polynitroimidazoles are considered dangerous and proper precaution should be taken in their handling and storage!

Methylation reaction of 2,4-dinitroimidazole (3) or potassium 2,4,5-trinitroimidazol-1-ide (4) (General method). 2,4-Dinitroimidazole (**3**) (1.58 g, 10.0 mmol) or potassium 2,4,5-trinitroimidazol-1-ide (**4**) (2.41 g, 10.0 mmol) was added to the appropriate solvent with stirring. After the addition was complete, the mixture was stirred for additional 15 min. DMS (19 ml, 0.2 mol) was then added dropwise carefully at room temperature. Finally, if needed, the requisite amount of a base was added. After completion of the addition, the mixture was heated gradually to the required temperature, stirred for 0.5–4 h, cooled to room temperature, and poured into distilled H_2O (50 ml). The mixture was extracted with CH_2Cl_2 or Et_2O (5×30 ml). In the case of potassium 2,4,5-trinitroimidazol-1-ide (**4**), saturated K_2CO_3 and KCl solutions were subsequently added to the combined Et_2O extracts to obtain a yellow precipitate of the starting material **4**, which was filtered off and dried. The combined organic extracts were evaporated under reduced pressure. All compounds were purified by column chromatography on silica gel (petroleum ether – EtOAc, 5:1).

1-Methyl-2,4-dinitroimidazole (1). Yellow powder, mp 141–143°C (mp 144–146°C[12a]). IR spectrum, ν, cm^{-1}: 1327 (NO_2), 1372 (N–CH_3), 1502 (C=C), 1554 (NO_2), 3128 (=C–H), 3152 (=C–H). (IR spectrum, ν, cm^{-1}: 1138 (C–N), 1325 (NO_2), 1461 (–CH_3), 1498 (C=C), 1515 (C=C), 1556 (NO_2), 2899 (–CH_3), 3152 (=C–H).[12b]) ^{1}H NMR spectrum (Me_2CO-d_6), δ, ppm: 4.28 (3H, s, CH_3); 8.52 (1H, s, CH). ^{13}C NMR spectrum (Me_2CO-d_6), δ, ppm: 39.0; 126.9; 143.0; 143.6. Found, %: C 27.89; H 2.35; N 32.57. $C_4H_4N_4O_4$. Calculated, %: C 27.92; H 2.34; N 32.56.

1-Methyl-2,4,5-trinitroimidazole (2). Yellow powder, mp 81–82°C (mp 81–82°C[16]). IR spectrum, ν, cm^{-1}: 1329 (NO_2), 1361 (N–CH_3), 1503 (C=C), 1549 (NO_2), 2899 (–CH_3). (IR spectrum, ν, cm^{-1}: 1329 (NO_2), 1361 (N–CH_3), 1503 (C=C), 1549 (NO_2), 2899 (–CH_3).[16]) ^{1}H NMR spectrum (Me_2CO-d_6), δ, ppm: 4.37 (3H, s, CH_3). ^{13}C NMR spectrum (Me_2CO-d_6), δ, ppm: 24.5; 154.7; 158.8; 159.0. Found, %: C 22.29; H 1.44; N 32.17. $C_4H_3N_5O_6$. Calculated, %: C 22.13; H 1.39; N 32.26.

2,4-Dinitroimidazole (3). Recovered from reaction mixture, pale-yellow powder, mp 264–266°C (mp 265–274°C[17]).

Potassium 2,4,5-trinitroimidazol-1-ide (4). Recovered from reaction mixture, yellow powder, mp 236–238°C (mp 234°C[10a]).

1-Methylimidazolidine-2,4,5-trione (5). Yield 0.07 g (6%), white powder, mp 146–148°C (mp 146–148°C[16]). IR spectrum, ν, cm^{-1}: 1330 (C–NH), 1455 (–CH_3), 1719 (C=O), 1741 (C=O), 1795 (C=O), 2815 (–CH_3), 3228 (N–H). (IR spectrum, ν, cm^{-1}: 1330 (C–NH), 1455 (–CH_3), 1719 (C=O), 1741 (C=O), 1795 (C=O), 2815 (–CH_3), 3228 (N–H).[16]) ^{1}H NMR spectrum (Me_2CO-d_6), δ, ppm: 3.06 (3H, s, CH_3); 10.77 (1H, br. s, NH). ^{13}C NMR spectrum (Me_2CO-d_6), δ, ppm: 24.5; 154.8; 158.8; 159.1. Found, %: C 37.42; H 3.19; N 21.95. $C_4H_4N_2O_3$. Calculated, %: C 37.51; H 3.15; N 21.87.

1,3-Dimethylimidazolidine-2,4,5-trione (6). Yield 0.13 g (9%), white powder, mp 152–154°C (mp 154°C[18]). IR spectrum, ν, cm^{-1}: 1279 (O=C–N–), 1390 (N–CH_3), 1465 (N–CH_3), 1706 (C=O), 1731 (C=O), 1768 (C=O), 2930 (–CH_3), 2958 (–CH_3). (IR spectrum, ν, cm^{-1}: 1710 (C=O), 1735 (C=O), 1770 (C=O), 2980 (CH).[18]) ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.98 (6H, s, CH_3). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 24.3; 154.8; 157.8. Found, %: C 42.29; H 4.25; N 19.69. $C_5H_6N_2O_3$. Calculated, %: C 42.26; H 4.26; N 19.71.

X-ray structural investigation of 1-methylimidazolidine-2,4,5-trione (5) and 1,3-dimethylimidazolidine-2,4,5-trione (6). Single crystals of 1-methylimidazolidine-2,4,5-trione and 1,3-dimethylimidazolidine-2,4,5-trione were obtained in CH_2Cl_2 solution by solvent evaporation at ambient temperature. X-ray diffraction data were collected on a Bruker D8 Venture Photon 100 CMOS detector equipped with a graphite monochromator, MoKα radiation (λ 0.07107 nm). The structures were solved by the direct methods (SHELXL-97 software)[19] and refined by the full-matrix-block least-squares method on F^2 with anisotropic thermal parameters for all non-hydrogen atoms (OLEX2 software).[20] The hydrogen atoms were added according to the theoretical models. Crystallographic data for 1-methylimidazolidine-2,4,5-trione (**5**) and 1,3-dimethylimidazolidine-2,4,5-trione (**6**) have been deposited at the Cambridge Crystallographic Data Center (deposits CCDC 1966870 and 1966866, respectively).

Supplementary information file containing ^{1}H and ^{13}C NMR spectra of all synthesized compounds and X-ray data of compounds **5**, **6** is available at the journal website at http://link.springer.com/journal/10593.

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