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N-Nitrosation of Glycolurils Catalyzed by 1-Hydroxyethylidene-1,1-diphosphonic Acid

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Abstract—A number of *N*-nitroso derivatives of glycolurils were obtained for the first time, using sodium nitrite and 1-hydroxyethylidene-1,1-diphosphonic acid (HEDP) as a green catalyst. The procedure was carried out in an aqueous heterophase medium without the use of aggressive acids.

Keywords: glycoluril, *N*-nitrosoglycoluril, 1-hydroxyethylidene-1,1-diphosphonic acid, catalyst, *N*-nitrosation, hydantoin

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INTRODUCTION

N-Nitrosation reactions are of special interest for organic synthesis and biochemistry [1]. For example, *N*-nitroso compounds are used as drugs (transnitrosation agents [2]) and as intermediates for the synthesis of various organic compounds containing an N–N fragment [2–5].

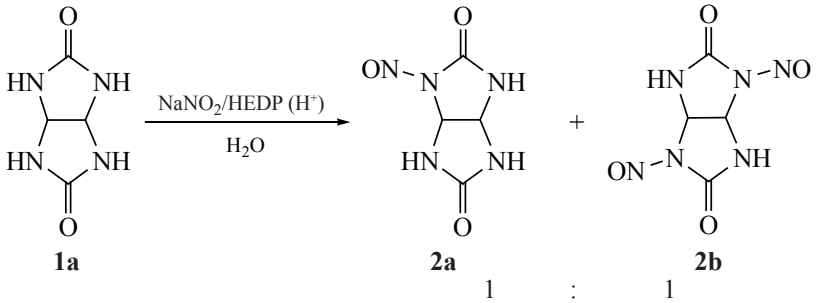
The known *N*-nitroso derivatives of glycoluril amount to as little as mono- (**2a**) and dinitrosated (**2b**) glycolurils which exhibit foaming properties and are used in the production of thermoplastic polymers [6]. The main method for the preparation of *N*-nitroso derivatives **2a** and **2b** involves the reaction of substrate **1a** with sodium nitrite in an aqueous solution of concentrated mineral acid at a low temperature [6, 7]. As known [6], with nitric acid, high yields of **2a** are achieved, while other acids, such as sulfuric, hydrochloric, or glacial acetic, sharply reduce the yield of this derivative. The above *N*-nitrosation conditions are quite corrosive [2, 8].

1-Hydroxyethylidene-1,1-diphosphonic acid (HEDP) are known by its anticorrosive properties and used as a scale inhibitor in water circulation cooling systems of industrial plants and thermal power plants [9]. We earlier found that HEDP is capable of efficiently catalyzing condensation [10, 11] and *N*-acetylation reactions of glycoluryl **1a** [12]. In view of the aforesaid, we set ourselves the task to explore the potential of HEDP as a catalyst in the synthesis of a series of *N*-nitroso derivatives of glycoluryls (Scheme 1).

RESULTS AND DISCUSSION

N-Nitrosation of glycoluryl **1a** was performed according to the procedure in [7], but instead of mineral

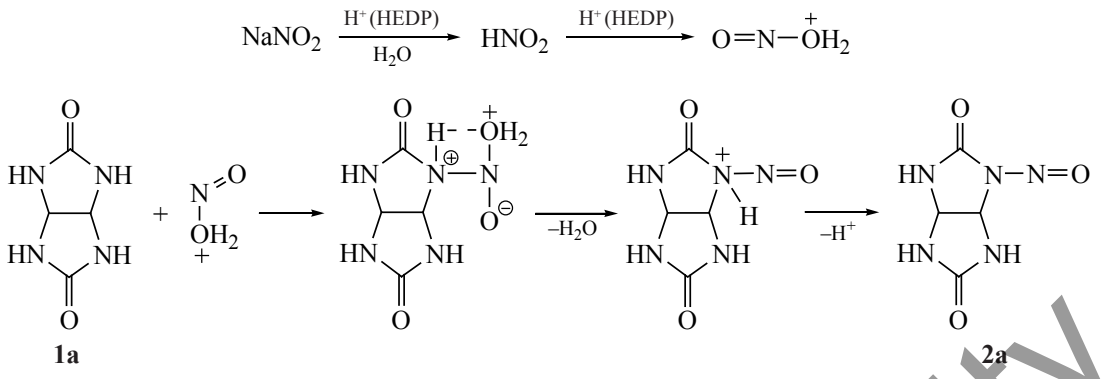
Scheme 1.



1a **2a** **2b**

1 : 1

Scheme 2.



1a **2a**

acid we used HEDP in a double excess to substrate **1a** (Scheme 1). The NMR spectra of the isolated products showed the presence of a mixture of *N*-nitrosylation products **2a** and **2b**: *N*-mononitrosoglycoluryl (**2a**) and *N*-dinitrosoglycoluryl (**2b**) in a 1 : 1 ratio as evidenced by the integral intensities of the corresponding signals). As known [13], the ratio of reactive species in nitrosation reactions depends in the acidity of the medium and the nature of the mineral acid. In the reaction in a weaker acid (in our case, HEDP), the likely diazotating species is nitrosacidium cation $H_2NO_2^+$. The probable mechanism of *N*-nitrosation of glycoluryl **1a** is presented in Scheme 2.

The classical *N*-nitrosation uses an excess of mineral acid, which is necessary to dissolve glycoluryl **1a**, because the low solubility of the latter prevents efficient reaction. One of the advantages of the HEDP catalyst is that it is able to enhance the solubility of glycoluryl **1a** [12], selectively destroying the crystal packing and thereby making substrate **1a** readily available for reaction at the nitrogen atoms. However, the excess of HEDP leads to the formation of by-product hydantoin **3** (Scheme 3) [10–12], and this explains why we use as little as 2 equiv of HEDP.

The *N*-nitrosation reaction of glycoluryl **1a** was carried out at 0–5°C, and no heat release was observed as sodium nitrite was added in the reaction mixture in the presence of HEDP; on the contrary, after $NaNO_2$ had dissolved in water, the temperature of the reaction mixture decreased by an average of 2°C.

The main advantage of the proposed method is that the synthesis of the target product is performed in softer and better controlled reaction medium, because it excludes the use of aggressive acids and organic solvents, despite the comparable reaction time and the relatively low yield of disubstituted *N*-nitrosoglycoluryl **2b** (14% vs. 64% in the classical synthesis [7]).

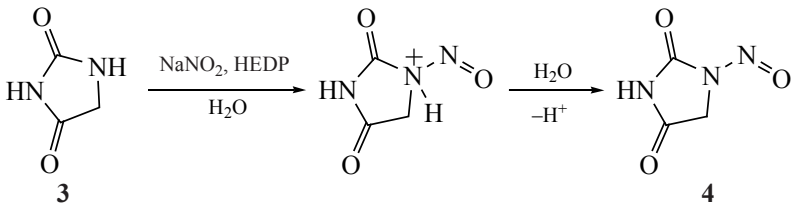
The NMR analysis of the reaction filtrate (Scheme 1) (see Supplementary Information) showed that the reaction mixture contained, along with HEDP, hydantoin **3** [10–12] and *N*-nitrosohydantoin **4** (Scheme 3).

The *N*-nitrosation of hydantoins has almost never been reported, despite the fact that nitrosohydantoins are cyclic analogs of *N*-nitrosoureas and are of considerable interest due to their biological activity [8].

To expand the range of *N*-nitrosated derivatives of glycoluryl **1a**, we carried out *N*-nitrosation of a number of glycoluryls **1b–1f** in the presence of HEDP as an acid catalyst (Scheme 4).

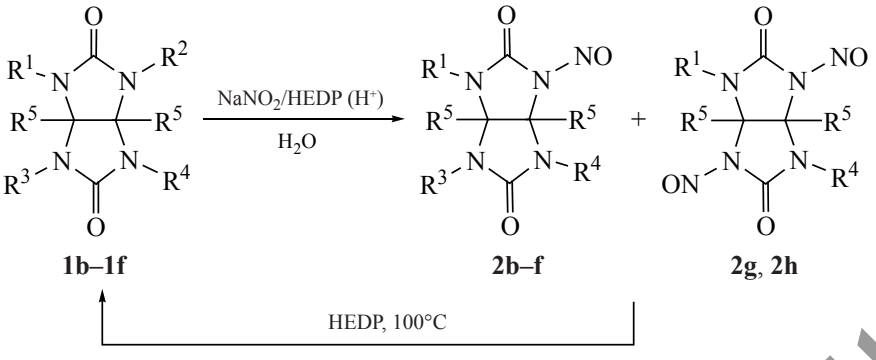
The *N*-nitrosation of 2,6-dimethylglycoluryl (**1d**) resulted in the isolation of mononitrosated (**2e**, 60%) and dinitrosated (**2h**, 31%) derivatives as light yellow solids.

Scheme 3.



3 **4**

Scheme 4.



1b–1f **2b–f** **2g**, **2h**

1b, $R^1 = R^2 = R^3 = R^4 = H$, $R^5 = CH_3$
1c, $R^1 = R^2 = R^3 = R^4 = H$, $R^5 = Ph$
1d, $R^1 = R^4 = H$, $R^2 = R^3 = CH_3$, $R^5 = H$
1f, $R^1 = R^3 = H$, $R^2 = R^4 = CH_3$, $R^5 = H$

2c, $R^1 = R^3 = R^4 = H$, $R^5 = CH_3$
2d, $R^1 = R^3 = R^4 = H$, $R^5 = Ph$
2e, $R^1 = R^4 = CH_3$, $R^3 = H$, $R^5 = H$
2f, $R^1 = R^3 = CH_3$, $R^4 = H$, $R^5 = H$
2g, $R^1 = R^4 = H$, $R^5 = CH_3$
2h, $R^1 = R^4 = CH_3$, $R^3 = H$, $R^5 = H$

The action of sodium nitrite in the presence of HEDP on 2,8-dimethylglycoluryl (**1f**) gave no dinitroso derivative, and the yield of mononitrosated 2,8-dimethylglycoluryl **2f** was 69%.

It was found that *N*-mononitrosated dimethylglycoluryls **2e** and **2f** were soluble in the reaction water, which made it possible to analyze these compounds by mass spectrometry (see Supplementary Information), and *N*-dinitroso-substituted dimethylglycoluryl **2h** precipitated immediately after the end of the reaction.

It was found that the *m/z* values of the fragment ions of compounds **2e** and **2f** absolutely coincide with each other, and the absence of differences in the mass spectra of compounds **2e** and **2f** is probably due to the similarity of the fragmentation pathways of their parent dimethylglycoluryls **1d** and **1f** [14].

It is noteworthy that the decomposition temperatures of products **2e**, **2f**, and **2h** are quite close to each other and fall in the temperature range 191–193°C. Thus, having reached 191°C, compounds **2e**, **2f**, and **2h** begin to decompose with gas release, which is obviously due to the heterolytic N–N bond cleavage.

Having the developed H-bond network [15], 1,5-dimethylglycoluryl (**1b**) is poorly soluble in water and to dissolve it, requires heating under reflux together with HEDP for 30 min. After the dissolution of compound **1b**, the resulting solution is rapidly cooled to 0°C, and then sodium nitrite is added. As a result of the reaction, mononitrosated (**2c**, 63%) and dinitrosated (**2g**, 36%) 1,5-dimethylglycoluryls are formed.

1,5-Diphenylglycoluryl (**1c**) does not enter into the *N*-nitrosation reaction in an aqueous medium, which is primarily due to its low solubility, because this compound, due to its increased lipophilicity, floates on the surface of the liquid. To increase the wettability of compound **1c**, one part of ethanol was added to the reaction mixture. This approach made it possible to overcome the problem and obtain *N*-monosubstituted product **2d** (greenish powder) in a yield of 10%. It should be noted that in the presence of mineral acid (HCl) we did not obtain *N*-nitrosated 1,5-diphenylglycoluryl **2d**.

In most cases, *N*-nitroso derivatives are unstable [16], and the rate of their decomposition increases and at elevated temperatures (100°C), especially in the presence of acids. We found that heating nitrosoglycoluryls **2a–2h** with 1 equiv of HEDP led to hydrolysis of starting glycoluryls **1a–1f** with N–N bond cleavage.

The absence of hydantoins in the reaction mixtures of glycoluryls **1b–1f** is obviously explained by the fact that *N*- and *C*-substitutions in compounds **1b–1f** make them less prone to destruction [10].

EXPERIMENTAL

The IR spectra of compounds **2a–2h** were recorded on a Thermo Fisher Scientific Nicolet 6700 IR spectrometer in the attenuated total internal reflectance

in the range 400–4000 cm^{-1}. The ^1H and ^{13}C NMR spectra were recorded on a Bruker AVANCE III HD spectrometer at 400 and 100 MHz, respectively, in DMSO-d_6 (compounds **2a–2h**) or H_2O–D_2O (compounds **3** and **4**, see Supplementary Information), internal standard TMS. Compounds **2**e and **2f** were identified by HPLC–MS on an Agilent Technologies 6550 iFunnel Q-TOF LC/MS high-resolution instrument. Analysis conditions: atmospheric pressure chemical ionization (APCI), positive ion registration mode, ionization source temperature 350°C, curtain gas flow rate 4 mL/min, gas flow rate in the ionization source 36 mL/min. Scan range *m/z* 105–400, detection frequency 5 Hz; mobile phase: water–acetonitrile (1 : 1), detection time 1.04 min. The elemental analyses of compounds **2d** and **2h** were obtained on a EuroVector Euro EA3000 CHNS-O analyzer. The melting points were measured on a Büchi M560 automatic melting point apparatus.

2-Nitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2a) and 2,6-dinitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2b). A three-necked flask equipped with a gas outlet tube, a thermometer, and a magnetic stirrer was charged with 5.8 g (0.028 mol) of HEDP and 50 mL of water. After complete dissolution, 2.4 g (0.014 mol) of glycoluryl **1a** was added, the mixture was cooled to –5°C, and then 5.0 g (0.07 mol) of sodium nitrite was added in portions. The reaction mixture was stirred for 90 min, then heated to room temperature and stirred for 50 min. The precipitate that formed was filtered off, washed with water, and recrystallized from DMF. The resulting light yellow powder was a mixture of mononitroso glycoluryl **2a** and dinitrosoglycoluryl **2b** in a 1 : 1 ratio.

Compound **2a**. Yield 0.5 g (43%), decomp. point 198–200°C (DMF). IR spectrum, ν, cm^{-1}: 3245 br (NH), 2994 s (CH), 1703 s (C=O), 1676 s (C=O), 1337–1452 br (N=O), 1084–1144 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 5.34 d (1H, C<u>H</u>CH, *J* 8.0 Hz), 5.64 d (1H, CHC<u>H</u>, *J* 6.2 Hz), 7.76 s (1H, NH), 7.96 s (1H, NH), 9.40 s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 62.1 (<u>C</u>HCH), 63.5 (CH<u>C</u>H), 152.3 (C=O), 160.6 (C=O). Found, %: C 28.01; H 2.90; N 40.73; O 28.31. $C_4H_5N_5O_3$. Calculated, %: C 28.08; H 2.95; N 40.93; O 28.05. *M* 171.12.

Compound **2b**. Yield 0.7 g (50%), decomp. point 198–200°C (DMF). IR spectrum, ν, cm^{-1}: 3351 br (NH), 2927 s (CH), 1745 s (C=O), 1337–1494 br (N=O), 1084–1144 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 5.63 s (2H, C<u>H</u>C<u>H</u>), 9.95 s (4H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 60.2 (CHCH), 152.0 (C=O). Found, %: C 24.01; H 2.00; N 42.03; O 31.96. $C_4H_4N_6O_4$. Calculated, %: C 24.01; H 2.01; N 42.00; O 31.98. *M* 200.11.

2,6-Dimethyl-4-nitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2e) and 2,6-dimethyl-4,8-dinitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2h). A three-necked flask equipped with a gas outlet tube, a thermometer, and a magnetic stirrer was charged with 5.8 g (0.028 mol) of HEDP and 50 mL of water. After complete dissolution, 2.4 g (0.014 mol) of 2,6-dimethylglycoluryl (**1d**) was added, the mixture was cooled to –5°C, and then 5.0 g (0.07 mol) of sodium nitrite was added in portions. The reaction mixture was stirred for 90 min, then heated to room temperature and stirred for 50 min. The crystals of dinitrosodimethylglycoluryl **2h** that precipitated were filtered off, washed with cold water, and dried. The remaining reaction mixture was left in a refrigerator for 1 day, and the precipitate that formed was filtered off and washed with cold water to obtain mononitrosodimethylglycoluryl **2e**.

Compound **2h**. Yield 0.33 g (31%), light yellow crystals, decomp. point 191–193°C (H_2O). IR spectrum, ν, cm^{-1}: 2992 s (CH), 2925 s (CH_3), 1713 s (C=O), 1450 br (N=O), 1037–1111 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 2.89 s (6H, CH_3), 5.65 s (2H, C<u>H</u>C<u>H</u>). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 30.9 (CH_3), 65.4 (CHCH), 152.1 (C=O). Found, %: C 31.31; H 3.59; N 36.93; O 28.17. $C_6H_8N_6O_4$. Calculated, %: C 31.58; H 3.53; N 36.83; O 28.05. *M* 228.17.

Compound **2e**. Yield 1.1 g (60%), light yellow powder, decomp. point 191–193°C (H_2O). IR spectrum, ν, cm^{-1}: 3354 br (NH), 2992 s (CH), 2925 s (CH_3), 1713 s (C=O), 1675 s (C=O), 1344–1428 br (N=O), 1037–1111 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 2.67 s (3H, CH_3), 2.90 s (3H, CH_3), 5.20 d.d (1H, C<u>H</u>CH, *J* 7.9, 2.2 Hz), 5.75 d (1H, CHC<u>H</u>, *J* 7.8 Hz), 8.10 d (1H, NH, *J* 2.2 Hz). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 28.2 (CH_3), 30.7 (CH_3), 63.8 (<u>C</u>HCH), 66.6 (CH<u>C</u>H), 151.5 (C=O), 159.3 (C=O). Mass spectrum, *m/z* (I_{rel}, %): 200.1 (3.6) $[M + H]^+$. Found, %: C 36.01; H 4.59; N 35.23; O 24.17. $C_6H_9N_5O_3$. Calculated, %: C 36.18; H 4.55; N 35.16; O 24.10. *M* 199.17.

2,8-Dimethyl-4-nitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2f). A three-necked flask

equipped with a gas outlet tube, a thermometer, and a magnetic stirrer was charged with 5.8 g (0.028 mol) HEDP and 50 mL of water. After complete dissolution, 2.4 g (0.014 mol) of 2,8-dimethylglycoluryl (**1f**) was added, the mixture was cooled to –5°C, and then 5.0 g (0.07 mol) of sodium nitrite was added in portions. The reaction mixture was stirred for 90 min and then heated to room temperature, and, after 50-min stirring, left in a refrigerator for 1 day. The precipitate that formed was filtered off and washed with cold water. Yield 0.8 g (69%), light yellow powder, decomp. point 195°C (H_2O). IR spectrum, ν, cm^{-1}: 3243 br (NH), 2992 s (CH), 2925 s (CH_3), 1713 s (C=O), 1675 s (C=O), 1329–1450 br (N=O), 1037–1111 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 3.12 s (6H, CH_3), 5.25 d (1H, C<u>H</u>CH, *J* 4.4 Hz), 6.05 d (1H, CHC<u>H</u>, *J* 7.5 Hz), 8.16 s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 30.9 (CH_3), 57.3 (<u>C</u>HCH), 71.1 (CH<u>C</u>H), 152.3 (C=O), 159.4 (C=O). Mass spectrum, *m/z* (I_{rel}, %): 200.1 (3.6) $[M + H]^+$. Found, %: C 36.44; H 4.37; N 35.04; O 24.15. $C_6H_9N_5O_3$. Calculated, %: C 36.18; H 4.55; N 35.16; O 24.10. *M* 199.17.

2,8-Dimethyl-4-nitroso-2,4,6,8-tetraazabicyclo-[3.3.0]octane-3,7-dione (2c) and 1,5-dimethyl-2,6-dinitroso-2,4,6,8-tetraazabicyclo[3.3.0]octane-3,7-dione (2g). A three-necked flask equipped with a gas outlet tube, a thermometer, and a magnetic stirrer was charged with 5.8 g (0.028 mol) HEDP and 50 mL of water. After complete dissolution, 2.4 g (0.014 mol) of 1,5-dimethylglycoluryl (**1d**) was added, the mixture was cooled to –5°C, and then 5.0 g (0.07 mol) of sodium nitrite was added in portions. The reaction mixture was stirred for 90 min and then heated to room temperature and stirred for 50 min. The precipitate that formed was filtered off, washed with water, and recrystallized from DMF to obtain a mixture of *N*-mononitrosodimethylglycoluryl **2c** and dinitrosodimethylglycoluryl **2g** in a 7 : 3 ratio.

Compound **2c**. Yield 1.23 g (63%), light yellow powder, decomp. point 250°C (DMF). IR spectrum, ν, cm^{-1}: 3226 br (NH), 2933 s (CH_3), 1701 s (C=O), 1661 s (C=O), 1314–1425 br (N=O), 1079–1142 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 1.42 s (3H, CH_3), 1.53 s (3H, CH_3), 7.80 s (1H, NH), 8.14 s (1H, NH), 9.47 s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 17.1 (CH_3), 17.9 (CH_3), 74.0 (<u>C</u>HCH), 78.1 (CH<u>C</u>H), 151.5 (C=O), 158.8 (C=O). Found, %: C 36.41; H 4.30; N 34.90; O 24.39. $C_6H_9N_5O_3$. Calculated, %: C 36.18; H 4.55; N 35.16; O 24.10. *M* 199.17.

Compound **2g**. Yield 0.35 g (36%), light yellow powder, decomp. point 250°C (DMF). IR spectrum, ν, cm^{-1}: 3225 br (NH), 2933 s (CH_3), 1701 s (C=O), 1314–1451 br (N=O), 1079–1143 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 1.61 s (6H, CH_3), 10.20 s (2H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 21.8 (CH_3), 75.4 (CHCH), 150.5 (C=O). Found, %: C 31.61; H 4.10; N 36.40; O 27.89. $C_6H_8N_6O_4$. Calculated, %: C 31.58; H 3.53; N 36.83; O 28.05. *M* 228.17.

2-Nitroso-1,5-diphenyl-2,4,6,8-tetraazabicyclo-[3.3.0]octane-3,7-dione (2d). A three-necked flask equipped with a gas outlet tube, a thermometer, and a magnetic stirrer was charged with 5.8 g (0.028 mol) of HEDP and 50 mL of water. After complete dissolution, 4.1 g (0.014 mol) of 1,5-diphenylglycoluryl (**1b**) was added, the mixture was heated until a stable suspension formed and cooled to –5°C, and 5.0 g (0.07 mol) of sodium nitrite was added in portions. The reaction mixture was stirred for 90 min and then heated to room temperature and stirred for 50 min. The precipitate that formed was filtered off and washed with water and acetone. Yield 0.45 g (10%), light green powder, decomp. point 287–290°C (H_2O). IR spectrum, ν, cm^{-1}: 3232 br (NH), 3064 s (CH), 2923 s (CH), 1818–2000 ov (Ph), 1710 s (C=O), 1670 s (C=O), 1226–1446 br (N=O), 1209 br (N–N). ^1H NMR spectrum (DMSO-d_6), δ, ppm: 7.08–7.11 m (10H, Ph), 8.57 s (1H, NH), 8.74 s (1H, NH), 10.15 s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 80.1 (C–Ph), 83.5 (C–Ph), 127.0, 127.6, 127.9, 128.2, 128.5, 135.9, 136.3 (Ph), 157.3 (C=O), 159.8 (C=O). Found, %: C 59.61; H 4.10; N 21.40; O 14.89. $C_{16}H_{13}N_5O_3$. Calculated, %: C 59.44; H 4.05; N 21.66; O 14.85. *M* 323.31.

CONCLUSIONS

For the first time, a series of *N*-nitroso derivatives of glycoluryls **2a–2h**, including monosubstituted products **2a–2f** and disubstituted products **2b**, **2g**, and **2h**, were obtained using a commercially readily available acid (HEDP) as a catalyst. It was found that at a substrate to HEDP ratio of 1 : 2, *N*-mononitroso-substituted glycoluryls **2a–2f** were obtained. *N*-nitrosoglycoluryls **2a–2h** were isolated in yields of 10–70%, and the relatively low yield of N-nitrosodiphenylglycoluryl **2d** is due to the poor solubility of starting substrate **1c** in water. The procedure of the synthesis is quite simple, the

reaction proceeds in an aqueous heterophase medium, which is typical for *N*-nitrosation processes [4, 6], and excludes the use of aggressive acids.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

SUPPLEMENTARY INFORMATION

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