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1,3,5-Triazine-2,4,6-triyltrisulfamic acid (TTSA): A new organic solid acid for the nitrosation of secondary amines and oxidation of urazoles in the presence of $NaNO_2$ under mild and heterogeneous conditions

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Abstract. Melamine reacted with chlorosufonic acid ($ClSO_3H$) to form a new sulfamic-type acid, 1,3,5-triazine-2,4,6-triyltrisulfamic acid (**TTSA**). Both nitrosation of secondary amines and oxidation of urazoles were accomplished by using **TTSA**/$NaNO_2$ system under mild and heterogeneous conditions with good to excellent yields.

Keywords. Oxidation; urazoles; nitrosation; secondary amines; 1,3,5-triazine-2,4,6-triyltrisulfamic acid (**TTSA**).

1. Introduction

Solid acids have many advantages such as simplicity in handling, decreased reactor and plant corrosion problems, and more environmentally safe disposal. Also, wastes and by-products can be minimized or avoided by developing cleaner synthetic routes.[1] Among the solid acids, sulfamic acid (NH_2SO_3H, **SA**) is a dry, non-volatile, non-hygroscopic, odourless, incorrodible crystalline solid with outstanding physical stability. Recently, **SA** emerged as a promising solid–acid catalyst for acid catalysed reactions, such as functional group protections and deprotections, and the synthesis of isoamyl acetate and polymeric ethers. Moreover, some important organic transformations, including Beckmann rearrangement, inter- and intramolecular imino Diels–Alder reactions, Pechmann and Biginelli condensations, have been also performed successfully in the presence of sulfamic acid.[2]

4-Substituted-1,2,4-triazole-3,5-diones (**TADs**), have been used both as substrates and reagents in various organic reactions. For example, they have been used in Diels–Alder, ene or [2 + 2] cycloadditions, dehydrogenation reactions, electrophilic aromatic substitution, condensation of dicarbonyl compounds, oxidation of alcohols to aldehydes and ketones.[3] Very recently aromatization of 1,4-dihydropyridines and pyrazolines as well as oxidation of thiols with **TAD**s were reported.[4] The unusual reactivity which makes **TAD**s (**2**, **4**) of interest also makes them hard to prepare and purify. It is interesting to note that, 4-phenyl-1,2,4-triazoline-3,5-dione (**2f**) is an extremely reactive dieonophile and enophile which is at least 1000 times more reactive than tetracyanoethylene in the Diels–Alder reaction with 2-chlorobutadiene and 2000 times more reactive than maleic anhydride. All of known synthetic methods of these compounds (1,2,4-triazolidine-3,5-diones) require oxidation of the corresponding urazoles (**1**, **3**).[5] Although a variety of reagents are capable of efficient oxidations of urazoles (**1**, **3**) to TADs, this transformation is not easy

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because these compounds are very sensitive to the oxidizing agents and reaction conditions.[4,5]

Nitrosation chemistry has been a fruitful area for mechanistic, organic, and biological chemists.[6] An effort has been also made to combine both the synthetic and mechanistic aspects of nitrosation or transnitrosation.[7,8] *N*-nitrosoamines have drawn considerable interest in recent years mainly due to their strong mutagenic and carcinogenic properties, vasorelaxant activity and their use as pesticides, antioxidants and lubricant additives. The most general reagent for the nitrosation of secondary amines is nitrous acid, generated from sodium nitrite and mineral acid in water or mixture of alcohol–water as solvent.[9] Other nitrosating agents, are Fremy's salt,[10] *bis*(triphenylphosphine)nitrogen(1+) nitrite,[11] *N*-haloamides and sodium nitrite,[12] oxyhyponitrite,[13] $[NO^{+}\cdot Crown\cdot H(NO_3)_2^{-}]$,[14] solid acids or acetic anhydride and sodium nitrite,[15] and N_2O_4 adducts.[16]

2. Experimental

2.1 *General*

Chemicals were purchased from Fluka, Merck and Aldrich. Products were characterized by comparison of their spectra (IR, ^{1}H NMR), TLC and physical data to the authentic samples which were given in our reported procedures.[21,22]

2.2 *Preparation of 1,3,5-triazine-2,4,6-triyltrisulfamic acid (**TTSA**)*

A 250 mL suction flask was used. It was equipped with a gas inlet tube for conducting HCl gas over an adsorbing solution i.e. water. It was charged with chlorosulfonic acid (5 mL, 75·2 mmol). While mixing, melamine (3·16 g, 25·07 mmol) was added in small portions over a period of 30 min at room temperature. HCl gas evolved from the reaction vessel immediately (scheme 1). When the addition of melamine was completed, the mixture was shaken for 30 min. A white solid **TTSA** was obtained.

2.3 *General procedure for the oxidation of urazoles and N-nitrosation of secondary amines*

A suspension of sodium nitrite, **TTSA** (for the molar ratio of **TTSA** and sodium nitrite to the substrate; please see the tables 1, 2), substrate (1 mmol), and wet SiO_2 [(50% w/w), 0·4 g] in dichloromethane (5 mL) was stirred vigorously magnetically at room temperature. After the completion of the reaction, the reaction mixture was filtered and the residue was washed with CH_2Cl_2 (2 × 5 mL). Then anhydrous Na_2SO_4 (4 g) was added to the filtrate and filtered off after 20 minutes. The solvent was evaporated and the products were obtained in good to excellent yields (tables 1 and 2).

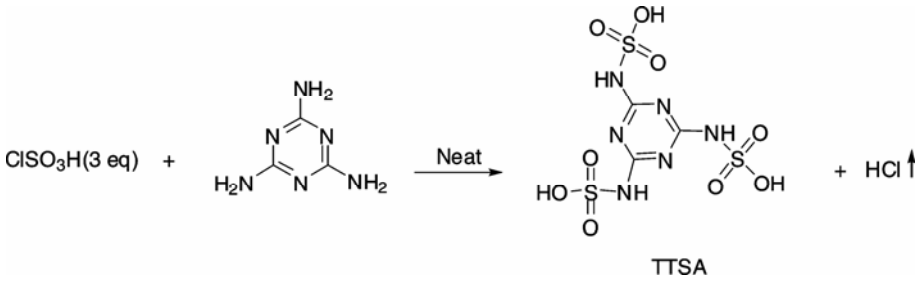
2.4 *Spectral data for the TTSA*

2.4a *Microanalysis data:* $C_3N_6H_6S_3O_9$, Found: (C = 9·81%; N = 22·95%; H = 1·64%), Calcd.: (C = 9·83%; N = 22·95%; H = 1·64%). IR, ν cm^{-1}: 1069, 1175, 1509, 1654, 2621, 3133.

2.5 *Spectral data for the N-nitroso compounds*[23]

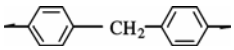
2.5a *N-Nitroso-N-phenyl benzyl amine* (**6a**): IR, ν cm^{-1}: 3050–3100, 2900, 1500, 1600, 1450, 1300, 1100. ^{1}H-NMR, δ (ppm): 6·90–7·25 (*m*, 10H), 4·98 (*s*, 2H).

2.5b *N-Nitrosodibenzyl amine* (**6b**): IR, ν cm^{-1}: 3050–3100, 2900, 1500, 1600, 1450, 1320, 1100. ^{1}H-NMR, δ (ppm): 6·90–7·14 (*m*, 10H), 4·99 (*s*, 2H), 4·46 (*s*, 2H). ^{13}C-NMR, δ (ppm): 44·6, 54·5, 139·30, 133·6, 128·50, 128·30, 128·06, 127·92, 127·36. Mp = 55–56°C.



Scheme 1.

Table 1. Oxidation of urazoles (**1**) and *bis*-urazoles (**3**) with **TTSA** and $NaNO_2$ in the presence of wet SiO_2 in dichloromethane at room temperature.

Entry	Substrate			Reagent/substrate (mmol)		Time (h)	Yields[a] (%)
		R_1	R_2	**TTSA**	$NaNO_2$		
1	1a	H	Me	0·67	2	1	100[b]
2	1b	H	Et	0·67	2	1	100[b]
3	1c	Na	*n*-Pr	1	2	3	56
4	1d	H	*n*-Bu	0·67	2	1	91
5	1e	H	Cyclohexyl	0·67	2	1	92
6	1f	H	Ph	0·67	2	0.5	99
7	1g	H	4-Cl-Ph	0·67	2	1.5	97
8	1h	H	3,4-Cl_2-Ph	0·67	2	1.5	99
9	1i	H	4-NO_2-Ph	0·67	2	2	88
10	1j	H	4-OMe-Ph	0·67	2	2	87
11	1k	H	4-*t*-Bu-Ph	0·67	2	1.5	94
12	1l	H	4-Naphtyl	0·67	2	2	96
13	3a	Na	$-(CH_2)_6-$	2·33	7	7	83
14	3b	H		1·33	4	1.5	86

[a]Isolated yields; [b]Conversion

2.5c *N-Nitroso dicyclohexyl amine* (**6c**): IR, ν cm^{-1}: 2850–2950, 1450, 1350, 1100. ^{1}H-NMR, δ (ppm): 4·76 (*m*, 1H), 3·65 (*m*, 1H), 1·3–1·85 (*m*, 20H). Mp = 94–96°C.

2.5d *Dinitroso piperazine* (**6d**): IR, ν cm^{-1}: 2850–2950, 1430, 1350. ^{1}H-NMR, δ (ppm): 4·51 (*s*, 4H), 4·42 (*t*, 4H, J = 4·8 Hz), 4·06 (*t*, 4H, J = 4·8 Hz), 3·83 (*s*, 4H). ^{13}C-NMR, δ (ppm): 37·16, 40·52, 46·58, 49·23. Mp = 156–160°C.

2.5e *N-Nitroso-2-methyl piperidine* (**6e**): IR, ν cm^{-1}: 2850–2950, 1450, 1350, 1050. ^{1}H-NMR, δ (ppm): 5·25 (*m*, 1H), 4·6 (*m*, 1H), 4·51 (*m*, 2H), 3·82 (*m*, 2H,), 3·71 (*m*, 3H), 1·51–1·92 (*m*, 18H), 1·5 (*d*, 3H, J = 1·3 Hz), 1·10 (*d*, 3H, J = 1·3 Hz), 0·92 (*d*, 3H, J = 1·3 Hz). Dec. p = 142°C.

*2.5*f *N-Nitroso piperidine* (**6f**): IR, ν cm^{-1}: 2850–2950, 1450, 1350, 1100. ^{1}H-NMR, δ (ppm): 4·06 (*b*, 1H), 3·64 (*t*, 2H, J = 4·1 Hz), 1·51–1·67 (*m*, 6H). Dec. p = 170°C.

2.5g *N-Nitroso-N-phenyl piperazine* (**6h**): IR, ν cm^{-1}: 3050–3100, 2900, 1500. ^{1}H-NMR, δ (ppm): 6·9–7·1 (*m*, 5H), 4·31(*t*, 2H, J = 4·8 Hz), 3·78 (*t*, 2H, J = 4·8 Hz), 3·31 (*t*, 2H, J = 4·8 Hz), 3·07 (*t*, 2H, J = 4·8 Hz). Mp = 45–47°C.

2.5h *N-Nitroso diethyl amine* (**6i**): IR, ν cm^{-1}: 2850–2950, 1450, 1375, 1350, 1040. ^{1}H-NMR, δ (ppm): 3·87(*q*, 2H, J = 6·3 Hz), 3·33 (*q*, 2H, J = 6·3 Hz), 1·16 (*t*, 3H, J = 6·3 Hz), 0·85 (*t*, 3H, J = 6·3 Hz). Bp = 173–176°C.

2.5i *N-Nitroso diisopropyl amine* (**6j**): IR, ν cm^{-1}: 2850–2950, 1450, 1375, 1350, 1040. ^{1}H-NMR, δ (ppm): 4·94 (*h*, 1H, J = 5·9 Hz), 4·19 (*h*, 1H, J = 5·9 Hz), 1·41 (*d*, 6H, J = 5·9 Hz), 1·07 (*d*, 6H, J = 5·9 Hz). ^{13}C-NMR, δ (ppm): 46·06, 20·4, 51·94, 25·14. Mp = 43–45°C.

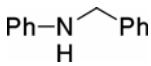
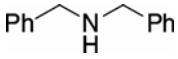
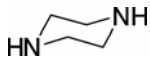
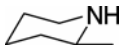
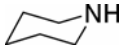
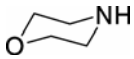
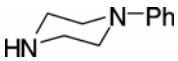
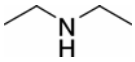
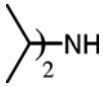
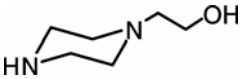
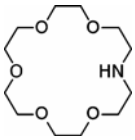
2.5j *Dinitroso cryptofix 22* (**6m**): IR, ν cm^{-1}: 2850–2950, 1450, 1350, 1100–1300, 1040. ^{1}H-NMR, δ (ppm): 4·32 (*t*, 8H, J = 8·1 Hz), 3·82 (*t*, 8H, J = 8·1 Hz), 3·81 (*t*, 4H, J = 8·2 Hz), 3·82 (*t*, 4H, J = 8·2 Hz), 3·58 (*s*, 4H), 3·56 (*m*, 4H), 3·11 (*m*, 4H), 2·91 (*s*, 4H), 3·42 (*m*, 8H). Mp = 41–43°C.

2.5k *N-Nitroso cryptofix 21* (**6n**): IR, ν cm^{-1}: 2850–2950, 1450, 1350, 1100–1300, 1050. ^{1}H-NMR, δ (ppm): 4·27 (*t*, 4H, J = 3·2 Hz), 3·5–3·76 (*m*, 20H). Mp = 92–94°C.

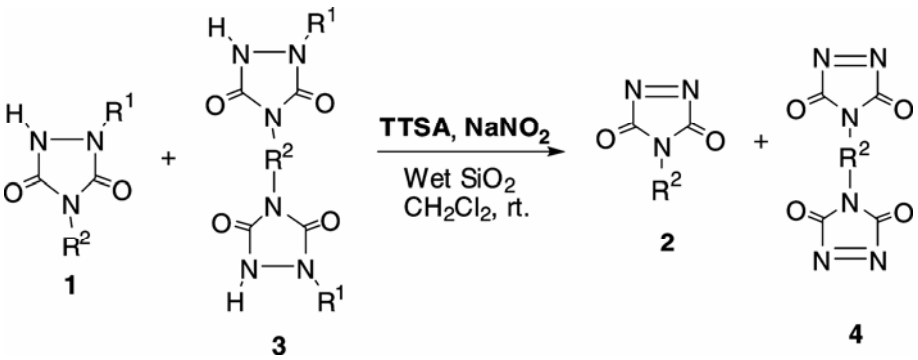
3. Results and discussion

On the basis of our experiences in the application of solid acids,[15] we found that melamine reacts with

Table 2. Nitrosation of secondary amines (**5**) with **TTSA** and $NaNO_2$ in the presence of wet SiO_2 in dichloromethane at room temperature.

Entry		Substrate (**5**)	Reagent/substrate (mmol) **TTSA**	Reagent/substrate (mmol) **$NaNO_2$**	Time (h)	Yields[a] (%)
1	a		0·67	2	0·08	89
2	b		0·67	2	0·17	98
3	c		0·67	2	0·08	99
4	d[d]		1·33	4	0·08	81
5	e		0·67	2	0·08	94
6	f		0·67	2	0·25	97
7	g		0·67	2	0·08	98
8	h		0·67	2	0·17	56[b]
9	i		0·67	2	0·08	83
10	j		0·67	2	0·08	88
11	k		0·67	2	0·17	–[c]
12	l		0·67	2	0·17	–[c]
13	m[d]		1·33	4	0·25	93
14	n		1·67	5	2	91

[a]Isolated yields; [b]Purified by a short pad of silica gel; [c]Undesired products; [d]Dinitrosated product



Scheme 2.

chlorosulfonic acid to give 1,3,5-triazine-2,4,6-triyltrisulfamic acid (**TTSA**). It is interesting to note that the reaction is easy and clean without any required work-up procedure, because the HCl gas is evolved from the reaction vessel immediately (scheme 1).

However, we hoped that the TTSA would be a superior proton source compared to other reported solid supported acids or acidic resins for running acid mediated reactions under mild and heterogeneous conditions.

In continuation of our studies on the oxidation of urazoles[17] and nitrosation of secondary amines,[18] we were interested in using the **TTSA** for the *in situ* generation of HNO_2 in combination with $NaNO_2$ and wet SiO_2 and eventually oxidation of urazoles as well as nitrosation of secondary amines. Here, as the first application of **TTSA**, we wish to report a simple, economical and effective method for oxidation of urazoles, *bis*-urazoles and also nitrosation of secondary amines under mild and heterogeneous conditions using TTSA/$NaNO_2$ system (schemes 2 and 3).

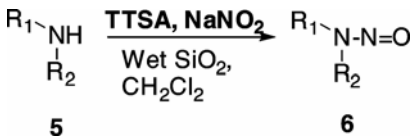
A wide range of urazoles (1) and *bis*-urazoles (3) were subjected to the oxidation reaction with a mixture of TTSA and $NaNO_2$ in the presence of wet SiO_2 in dichloromethane at room temperature (table 1).

Also, nitrosation of secondary amines (**5**) as well as oxidation of urazoless occurred under the same conditions. Thus, **TTSA**/$NaNO_2$/wet SiO_2 is a suitable system for the nitrosation of various kinds of secondary amines under mild and heterogeneous conditions (scheme 3 and table 2).

The presented oxidation/nitrosation reaction can be readily carried out only by addition of **TTSA**, $NaNO_2$ and wet SiO_2 to a suspension of urazoles (**1**), *bis*-urazoles (**3**) or a solution of secondary amines (**5**) in CH_2Cl_2 and efficiently stirring the resulting heterogeneous mixture at room temperature. The products (**2**, **4** and **6**) are obtained by simple filtration and evaporation of the solvent. According to the previous reports[17,18] and the current results, we propose the following mechanism for the oxidation/nitrosation reaction (scheme 4).

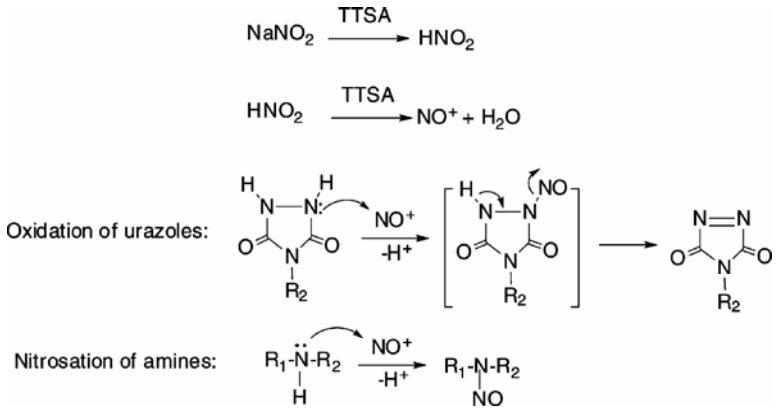
In continuation of our studies on the chemistry of 1,4-dihydropyridines[19] and thiols[20] we were also interested in using the above mentioned oxidizing reagents for the oxidation of 1,4-dihydropyridines (**7**) and coupling of thiols (**8**), but the reactions were sluggish and impractical. Several efforts for optimizing the reaction conditions failed and the yields of the desired products (i.e. pyridines (**9**) or disulfides (**10**) were very low which resulted to unidentified by-products. Also, based on our reported procedure for the oxidation of urazoles with $KClO_3$,[21] we tried to use the TTSA/$KClO_3$ system for the oxidation of urazoles, but this reaction was not suitable for this purpose (scheme 5).

Nitrosation of benzyl-phenylamine shows a good chemoselectivity, providing *N*-nitrosobenzylphenylamine as the only product. The described system behaves different from previously reported methods,[18a] because nitrosonium ion (NO^+) attacks the nitrogen sites of the secondary amines even where an aromatic moiety is connected directly to the nitrogen atom.

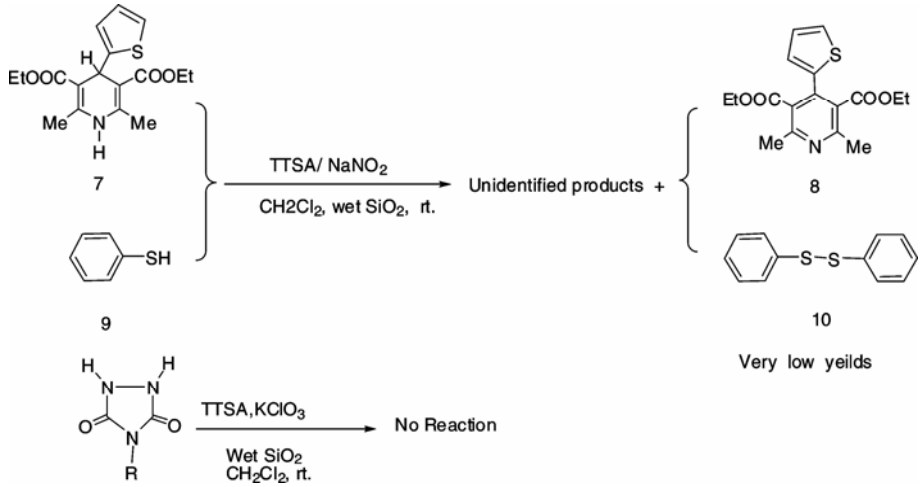


5 **6**

Scheme 3.



Scheme 4.



Scheme 5.

4. Conclusion

TTSA is a superior proton source considering convenience, cost effectiveness and simplicity of its production procedure. Also, **TTSA** is insoluble in most of the organic solvents and is an excellent proton mediated source for the organic reactions under heterogeneous conditions.

Acknowledgment

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