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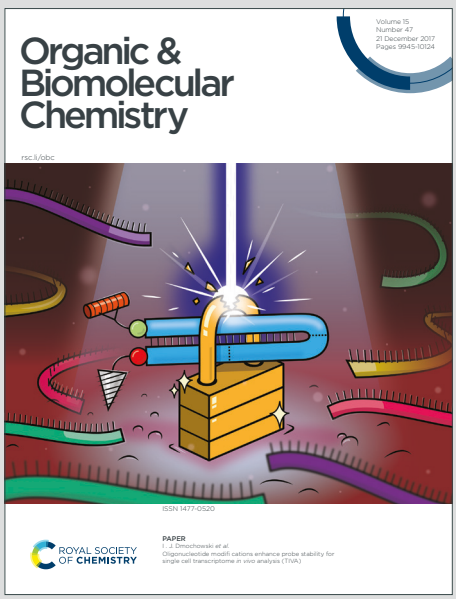

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COMMUNICATION

A cascade process for direct converting nitriles (RCN) to cyanamides (RNHCN) via SO_2F_2-activated Tiemann rearrangement

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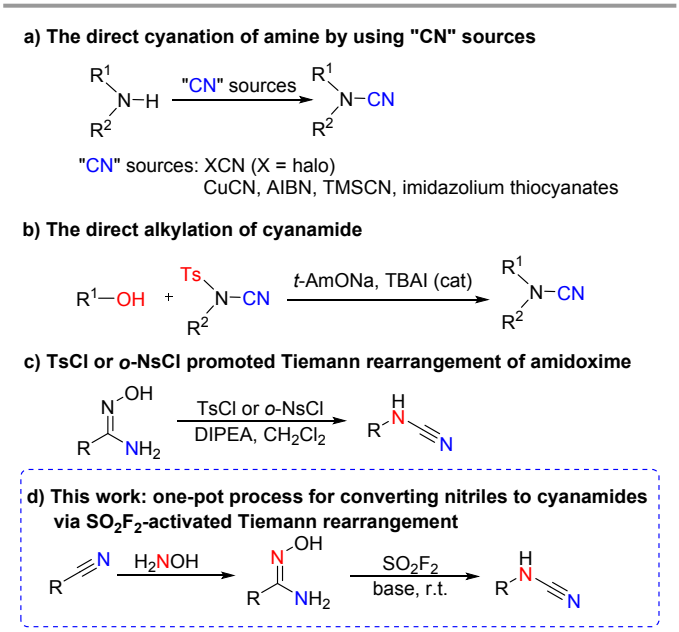
A simple, mild and practical process for direct converting nitriles to cyanamides was newly discovered feathering a wide substrate scope and great functional-tolerability (36 examples). In this efficient strategy the situ generated amidoximes, obtained from the reaction of nitriles with hydroxylamine, subsequently underwent Tiemann rearragement giving corresponding cyanamides in great isolated yields under SO_2F_2. Additionally, control experiments were reported to shed light the tentative mechanism involved formation and elimination of key intermediate, sulfonyl ester.

As a reactive N–C–N building blocks, the cyanamide moiety has been found in various bioactive molecules and functionalized materials.[1] But it is more commonly used as a precursor in the synthesis of pharmaceutically important *N*-containing heterocycles and *N*-alkyl or *N*-aryl imides.[2] Despite their versatile applications, only a limited number of synthetic routes for cyanamides have been reported in the literature.[3] The most frequently adopted method is the cyanation of amine using cyanogen halides,[4] which is overshadowed by its actue toxicity, unfavorable physical properties and sensitivity to moisture (Scheme 1, **a**).[5] Another straightforward approach is the direct alkylation of cyanamide, but *N*,*N*-dialkylated cyanamides are usually obtained due to the competing alkylation of the monoalkylated cyanamides (Scheme 1, **b**).[6] Other approaches include dehydrosulfurization of thiourea,[7] dehydration of urea, and the conversions from isocyanides, isocyanates, or isothiocyantes.[8] These methods are mutually complementary since they are all originated from the corresponding amines with multistep manipulations. And some of the transformations require harsh conditions or hazardous reagents. Recently, several new cyanide sources including CuCN,[9] AIBN,[10] TMSCN,[11] and imidazolium thiocyanates,[12] were achieved to the direct *N* - cyanation of amines access to cyanamides (Scheme 1, **a**). As an alternative approach, Tiemann rearrangement of amidoximes attracted our great interest for the synthesis of cyanamides.[13,14] Especially in 2014, Chien reported the benzenesulfonyl chlorides (TsCl or *o*-NsCl) promoted Tiemann rearrangement of amidoximes to generated corresponding cyanamides (Scheme 1, **c**).[14] However, it is highly dependent on the electronic effect of the substrates, requires rigorous reaction conditions and redundant work-up.



Scheme 1 Strategies for the synthesis of cyanamides.

Sulfuryl fluoride (SO_2F_2),[15] an inexpensive, abundant and relatively inert electrophile (stable up to 400 °C when dry) has recently attracted significant attention for Sulfur (VI) fluoride exchange (SuFEx) click chemistry and other versatile manipulations.[16] A perusal of the literature revealed that the proton of phenolic hydroxyl or oxime hydroxyl can activate the exchange of S-F bonds of SO_2F_2 for S-O bonds to make functional products, and fluorosulfate functional group (-OSO_2F) could be applied in a controllable and targeted manner

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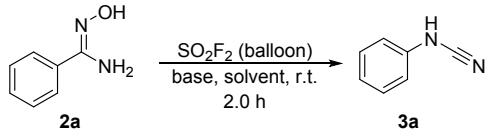
for varied transformations.[17] Most recently, prof Qin and our groups simultaneously reported a mild and robust method for efficiently converting aldehydes or aldoximes into corresponding nitriles mediated by SO_2F_2/base in a green manner.[18] Subsequently, an efficient activation of the Beckmann rearrangement of ketoximes for accessing amides or lactams utilizing the SO_2F_2 was developed in our lab.[19] Similarly to aldoxime or ketoxime, we speculated that the proton of amidoxime could also activate the exchange of S-F bond of SO_2F_2 for S-O bond to generate the corresponding intermediate sulfonyl ester under alkalinous conditions, which will subsequently fluorosulfonic ester elimination to promote Tiemann rearrangement. Herein, we report a cascade process for direct converting nitriles to cyanamides via SO_2F_2-activated Tiemann rearrangement (Scheme 1, **d**).

Table 1 Optimization of the reaction conditions.[a]



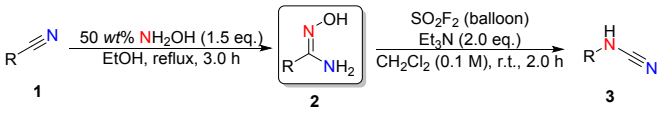
2a **3a**

Entry	Base (equiv)	Solvent	Isolated yield (%)
1	Et_3N (2.0)	CH_2Cl_2	94
2	DBU (2.0)	CH_2Cl_2	47
3	DIPEA (2.0)	CH_2Cl_2	73
4	Pyridine (2.0)	CH_2Cl_2	58
5	Et_3N (1.5)	CH_2Cl_2	66
6	Et_3N (3.0)	CH_2Cl_2	93
7	Et_3N (2.0)	CH_3CN	49
8	Et_3N (2.0)	CH_3OH	15
9	Et_3N (2.0)	EtOAc	85
10	Et_3N (2.0)	DMSO	62
11	Et_3N (2.0)	THF	30
12[b]	Et_3N (2.0)	CH_2Cl_2	96
13[c]	**Et_3N (2.0)**	**CH_2Cl_2**	**95**

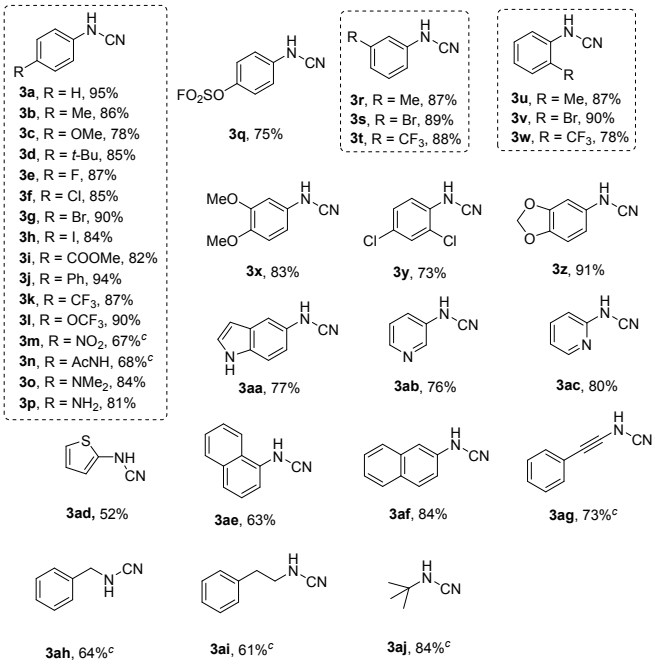
[a] Reaction conditions: benzamidoxime **2a** (0.5 mmol), base (1.0 mmol, 2.0 eq.), solvent (2.5 mL, 0.2 M), and SO_2F_2 balloon, room temperature, 2.0 h. [b] 5.0 mL of solvent (0.1 M). [c] Benzonitrile **1a** (1.0 mmol), 50 *wt*% NH_2OH (100 mg, 1.5 mmol, 1.5 eq.), and EtOH (10 mL, 0.1 M), reflux, 3.0 h; then the mixture was concentrated, CH_2Cl_2 (10 mL, 0.1 M) and Et_3N (280 uL, 2.0 mmol, 2.0 eq.) was added, and SO_2F_2 was introduced by slow bubbling through a SO_2F_2 balloon, room temperature, 2.0 h.

Initially, we conducted our investigation by examining the representativesubstrate benzamidoxime **2a** to test the feasibility of the proposed Tiemann rearrangement. Accordingly, after screening a large variety of conditions as shown in Table 1. We are pleased to observe that the desired Tiemann product, *N*-phenylcyanamide **3a**, was isolated in great yield of 94% under SO_2F_2 atmosphere at room temperature when 2.0 equiv. of triethylamine (Et_3N) was employed in CH_2Cl_2 (Table 1, entry 1). Inspiringly, various bases, including organic bases (Table 1, entries 2-4) and inorganic bases were further screened (see ESI† for a more detailed account of optimization conditions). Although inorganic bases have significant advantages over their organic counterparts,[20] we were disappointed to find that using inorganic bases, such as *t*-BuONa, Na_2CO_3 and K_2CO_3 provided only a trace amount of the desired product, even though increasing the reaction time (Table S1, entries 4-6). It is worth noting that reducing the loading of Et_3N to 1.5 equiv. cause obvious decreasing yield of **3a** (Table 1, entry 5, 66 % yield), while increasing Et_3N to 3.0 equiv., the yield is basically unchanged (Table 1, entry 6, 93 % yield). Subsequently, the usage of other common solvents, such as CH_3CN, CH_3OH, EtOAc, DMSO and THF, did not improve the yield of this transformation (Table 1, entries 7-11). Furthermore, reducing the concentration of substrate bring a superior isolated yield of **3a** in 96% (Table 1, entry 12). Alternatively, considering of developing sustainable and cost-effective methods from readily available and abundant starting materials in a PASE (pot, atom and step-economical) manner with the least requirements of isolation or purification of intermediates,[21] a series of continuous conversions,[22] including oximation of nitriles and cyanation of amidoximes, was tested by using benzonitrile **1a** as starting material. It's exciting to find that the final product **3a** was still generated in 95 % isolated yield (Table 1, entry 13).

Table 2 Substrate scope of the SO_2F_2-promoted Tiemann rearrangement.[a,b]



1 **2** **3**



[a] Reaction conditions: nitrile substrate **1** (1.0 mmol), 50 *wt*% NH_2OH (100 mg, 1.5 mmol, 1.5 eq.), and EtOH (10 mL, 0.1 M), reflux, 3.0 h; then the mixture was concentrated, CH_2Cl_2 (10 mL, 0.1 M) and Et_3N (280 uL, 2.0 mmol, 2.0 eq.) was added, and SO_2F_2 was introduced by slow bubbling through a SO_2F_2 balloon, room temperature, 2.0 h. [b] Isolated yields based on **1**. [c] 5.0 h.

Having the optimization reaction conditions in our hand, we further pursued the scope and generality of this process with respect to the other substrates (Table 2). Gratifyingly, a broad range of aromatic and aliphatic substituted amidoximes, which were obtained (mostly in up to nearly quantitative yield) from

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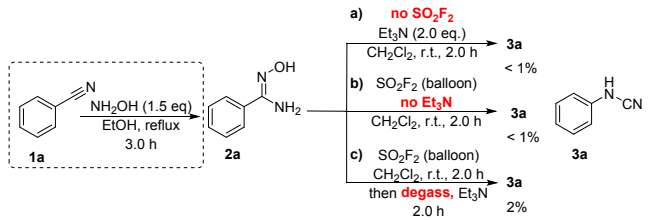
corresponding nitriles without purification, was found to be rearranged smoothly by this protocol to afford the corresponding cyanamides in good to excellent yields and a functional group-tolerant fashion. Irrespective of strongly activating (**3b**-**d** and **3i**, **3x**, **3z**), weakly deactivating (**3e**-**h** and **3y**), moderately deactivating (**3j**) and strongly deactivating (**3k**-**n**), the nature of substituents attached to the phenyl ring is equally effective. Especially the transformative moiety, iodic substituent was well tolerated (**3h**). Surprisingly, the derivatives bearing oxidation-sensitive groups (NMe_2 and NH_2) also reacted efficiently, providing moderate yields of the desired cyanamides without the generation of other side products (**3o**, **3p**). Remarkably, the 4-hydroxybenzonitrile **1q** derived amidoxime containing both SuFEx-sensitive aromatic hydroxy group and Tiemann-active oxime hydroxy group was smoothly achieved to form 4-cyanamidophenyl sulfurofluoridate **3q** in 75% yield. Notably, the satisfactory results showed the position of substituents on thearyl rings exhibited insignificant influence on the efficiency (**3b**, **3r**, **3u**, and **3g**, **3s**, **3v**, and **3k**, **3t**, **3w**). Moreover, this transformation of aromatic nitriles featuring a heterocyclic aromatic ring (**3aa**-**ad**) and an aromatic fused ring (**3ae**, **3af**) was furnished their corresponding cyanamides in gratifying yields. Besides, the phenylethynyl nitriles (**3ag**) were also achieved to give final products in 73% isolated yield. As regards aliphatic moiety, representative nitriles (**3ah**-**aj**) were also successfully transformed into their corresponding cyanamides in good yields.



Scheme 2 Control experiments for mechanism investigation.

As illustrated in Scheme 2, a series of control experiments were conducted to gain further insight into the mechanism of this SO_2F_2-promoted Tiemann rearrangement. Mixing the benzamidoxime **2a**, which was generated from benzonitrile **1a** without any by-product, just with Et_3N without the presence of SO_2F_2, the desired *N*-phenylcyanamide **3a** was not generated (Scheme 2, **a**). Coincidentally, only a trace amount of **3a** was observed while mixing **2a** just with SO_2F_2 without 2.0 equiv. of Et_3N (Scheme 2, **b**). Moreover, pre-mixing the **2a** with SO_2F_2, then deaerating SO_2F_2 and adding 2.0 equiv. of Et_3N, the *N*-phenylcyanamide was formed in negligible yield (Scheme 2, **c**), which demonstrating the generation and elimination of sulfonyl ester were crucial for this Tiemann rearrangement.

Based on the results of the control experiments and our previous works about SO_2F_2-activated transformations of oximes,[18,19] a plausible mechanism for this Tiemann rearrangement process was proposed (Figure 1). Initially, the nitrile **1** reacted with NH_2OH in polar solvent (EtOH) to generate amidoxime **2** through a nucleophilic addition and dehydration process. Then amidoxime **2** was deprotonated with SO_2F_2 under the promotion of Et_3N to form the key intermediate, sulfonyl ester **A**. Subsequently, the N–O bond cleavage occurred with concomitantly R group migration over C–N bond to furnish the *N*-substituted cyanamides **3**.

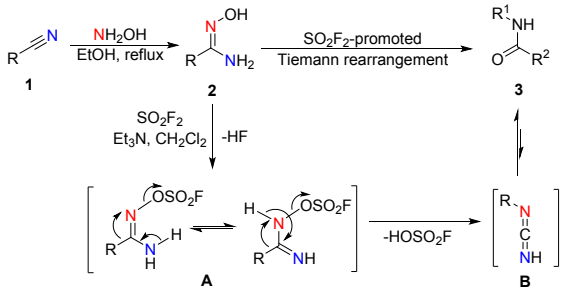
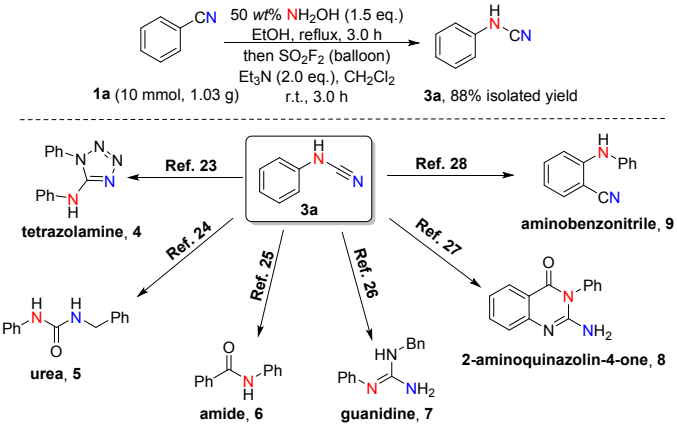


Figure 1 A plausible mechanism for the SO_2F_2-promoted Tiemann rearrangement.

To demonstrate the synthetic utility of this one-pot strategy for direct converting nitriles to cyanamides, a gram-scale (10 mmol, 1.03 g) reaction was performed under the standard conditions (Scheme 3). The desired *N*-phenylcyanamide **3a** was obtained in 88% isolated yield. Due to the resulting product *N*-phenylcyanamide **3a** have been widely applied in direct and efficient synthesis of many bioactive molecules as estimable building blocks, this protocol is particularly useful. For instance, **tetrazolamine 4**,[23] **urea 5**,[24] **amide 6**,[25] **guanidine 7**,[26] **2-aminoquinazolin-4-one 8**,[27] and **aminobenzonitrile 9.**[28] These representative transformations clearly demonstrate the versatilities of cyanamides in organic chemistry.



Scheme 3 A gram-scale preparation and further transformations of *N*-phenylcyanamide **3a**.

In summary, we have developed a novel, mild, practical and robust method for direct converting nitriles into cyanamides promoted by SO_2F_2 with the presence of Et_3N in a PASE manner. More than 36 structurally diverse cyanamides were synthesized with moderate to great isolated yields, demonstrating that the high efficiency, broad scope and functional-group compatibility of this new protocol. A gram-scale reaction was performed to demonstrate the applicability of cyanamides, which can be efficiently converted to various

structures. In addition, a SO_2F_2-activated Tiemann rearrangement mechanism was also proposed.

Conflicts of interest

There are no conflicts to declare.

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