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Tricyanomethane and its Ketenimine Tautomer Generated from Different Precursors and Analyzed in Solution, Argon Matrix, and Single Crystal

Klaus Banert,* Madhu Chityala, Manfred Hagedorn, Helmut Beckers,* Tony Stüker, Sebastian Riedel, Tobias Rüffer, and Heinrich Lang

Abstract: Solutions of azidomethylidenemalononitrile were photolyzed at low temperatures to produce the corresponding 2Hazirine and tricyanomethane, which were analyzed by lowtemperature NMR spectroscopy. The latter product was also observed after short thermolysis of the azide precursor in solution, whereas irradiation of the azide isolated in an argon matrix did not lead to tricyanomethane, but to unequivocal detection of the tautomeric ketenimine by IR spectroscopy for the first time. When the long-known "aquoethereal" greenish phase, generated from potassium tricyanomethanide, dilute sulfuric acid, and diethyl ether, was rapidly evaporated and sublimed, a mixture of hydronium tricyanomethanide and tricyanomethane was formed instead of previously claimed ketenimine tautomer. Under special conditions of sublimation, isolation of single crystals of tricyanomethane was possible and enabled analysis of the molecular structure by X-ray diffraction.

Initial attempts to prepare and isolate tricyanomethane (5), the socalled cyanoform, were performed as early as 1896 when Schmidtmann treated sodium tricyanomethanide (1a) with dilute sulfuric acid (Scheme 1).^[1] After addition of diethyl ether, he got a three-phase system, which included a greenish middle layer 2 that was claimed to contain cyanoform (5). But first experiments to remove the solvents did not lead to characterizable products. These phenomena were confirmed by Hantzsch and Osswald, who stated that tautomerism of 5 is likely to generate ketenimine 3.^[2] In 1963, Trofimenko repeated the synthesis of 2 by using the precursor 1b, and called the substance 2 "aquoethereal cyanoform".^[3,4] He was successful to liberate 2 from solvents by rapid evaporation and sublimation to get white crystals claimed to be composed of 3. Structure assignment of 3 was mainly based on elemental analysis and incomplete IR data, but ¹H NMR spectroscopy was missing, although precursor 2 was

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characterized by the latter method. When rapid evaporation of 2 was performed without sublimation, single crystals of hydronium tricyanomethanide (4) were obtained as revealed by X-ray diffraction crystallographic studies.^[5] Not only removal of the solvents from 2 followed by transport of the product into the gas phase, but also treatment of silver salt 1c with pure dry hydrogen sulfide was utilized to analyze the corresponding products by microwave spectroscopy^[6] and photoelectron spectroscopy.^[7] With the help of both methods, the gas-phase structure of 5 was confirmed. On the other hand, 5 as well as its tautomer 3 were discussed to explain the acidic properties of cyanoform.^[2,3,8] Thus, this compound is mentioned in textbooks of organic chemistry as one of the strongest carbon acids, with pK_a of about -5. The relative stabilities, spectroscopic features, and rearrangement reactions of 3, 5, and other C₄HN₃ species were studied using quantum chemical methods.^[9] Recently, the synthesis of 5 by treating 1d with an excess of anhydrous hydrogen fluoride was reported by Kornath et al.^[10] The product **5** could not be separated from 6 and turned out to be stable only below -40 °C. After removal of hydrogen fluoride, however, characterization was possible by low-temperature IR and Raman spectroscopy. Convincing NMR spectra (¹H, ¹³C, ¹⁴N), which were recorded at -45 °C by using solutions in $[D_6]$ acetone, were also reported. At first glance, the results of Trofimenko^[3] seem to be incompatible with those of the Kornath group.^[10] But the presence of the acidic salt 6 may have a strong influence on dissociation or exchange processes of 5 and possibly on the equilibration rate of the tautomers 3 and 5.



Scheme 1. Reported syntheses of ketenimine 3 and cyanoform (5).

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Herein, we report the first generation and unequivocal spectroscopic assignment of ketenimine **3**, and also the characterization of pure cyanoform (**5**) by a single crystal X-ray diffraction study. Furthermore, we clarify what kind of products are formed after evaporation and sublimation of **2**, and we explain why special conditions have to be fulfilled to measure NMR spectra of **5**.

At first, we looked for an acid-free method to prepare the target compounds 3 and 5 at low temperature. We assumed that photolysis of the known^[11] vinyl azide 7 will hopefully lead not only to the probably highly unstable azirine 8,[12] but also to a product which results from loss of dinitrogen combined with migration of the hydrogen atom (Scheme 2). After irradiating a solution of 7 in CD₃CN at -40 °C, ¹H and ¹³C NMR spectra (-40 and +20 °C) indicated the formation of 8 (49% yield) and the aziridine 9 (45%). Since the latter product was possibly generated by addition of 3 or 5 at 8, we performed a similar photolysis of 7 in [D₈]THF at -60 °C. In the corresponding ¹H NMR spectra (-60 °C), the signals of 8 were not accompanied by those of 9; however, we found only an additional broad signal, which could not be assigned to 3 or 5. Finally, we irradiated a solution of 7 in [D₈]THF at -85 °C and measured the NMR spectra of the products at -95 °C. The isotopically labeled precursors ²H-7, ¹³C-7, and ¹⁵N-7 were included into our studies, and this supported assignments of signals and allowed to also analyze ¹⁵N NMR spectra. Besides signals of the main product 8, we detected at such low temperatures sharp signals of cyanoform (5) in the NMR spectra (¹H, ¹³C, ¹⁵N).^[13] Stepwise going up with the temperature of the NMR probe led to broader signals of 5; for example, the proton signal was very broad at -50 °C and undetectable at -30 °C (Figure 1). At the latter temperature, formation of 9 slowly started. When the probe was recooled to -95 °C, however, the sharp signals of 5 returned. Thus, NMR signals of 5 are "hidden" if the measurements are performed at the "wrong" temperatures.

At low temperatures, photolyses of **7** in [D₈]THF yielded also a small side product, that was identified as hydronium tricyanomethanide (**4**) because addition of highly soluble trihexyltetradecylphosphonium tricyanomethanide did not lead to a new set of NMR signals of the anionic part. Even with dried





Figure 1. ¹H NMR spectra of cyanoform (**5**) and 2*H*-azirine **8** in [D₈]THF (400 MHz) at different temperatures starting with -95 °C (top).

precursor **7** and anhydrous solvent, we were not able to completely avoid the signals of **4**, which might include those of **3** if very rapid exchange processes are supposed. With the help of our NMR methods, successful detection of **3** was not possible since a conclusive ¹⁵N NMR signal with large doublet coupling ¹*J*(¹⁵N,¹H) was not observed, even at -130 °C (Me₂O, [D₈]THF).

The "intrinsic" stability of cyanoform (5) cannot be determined when this species is generated by low-temperature photolysis of 7 because subsequent warming leads to the trapping reaction $5 + 8 \rightarrow 9$. Whereas vinyl azides generally form 2*H*-azirines as primary products after irradiation, it is also well known that the same substrates often affords mainly nitriles by thermolysis.^[14] Thus, we tested the stability of 5 by heating diluted solutions of 7 and ¹⁵N-7 in [D₈]THF. It turned out that heating at 60 °C for 1.5 min followed by rapid cooling to -95 °C and analysis by NMR spectroscopy (¹H, ¹³C, ¹⁵N) is a good compromise with acceptable conversion of the precursors (51–54%) and limited decay of 5 or ¹⁵N-5, which are produced with 49–52% yield based on reacted 7 or ¹⁵N-7, respectively.

The unexpected thermal stability of cyanoform (5) and our difficulties to detect the ketenimine 3 led to the question whether the former compound is also isolated after workup and sublimation of 2. When we repeated the experiment described by Trofimenko,^[3] low-temperature ¹H and ¹³C NMR spectra of the sublimed product indicated the formation of 4 and 5 instead of 3. Iterated sublimation gave a substance with an increased proportion of 5. As shown in Figure 2, NMR spectra of such mixtures of 4 and 5 in [D₈]THF are highly temperature dependent. Whereas sharp ¹H NMR signals were detected at -95 °C, very broad signals were observed at -50 °C, and decay started at 0 °C. By recooling to -95 °C, however, the sharp signals returned. Broadening of NMR signals at -50 °C was less pronounced when the mixture of 4 and 5 consisted mainly of 5. Obviously, Trofimenko^[3] isolated at least mixtures which include cyanoform (5). But he misinterpreted his results, possibly also because convincing characterization of 5 by NMR spectroscopy requires low-temperature measurements.

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Figure 2. ¹H NMR spectra of hydronium tricyanomethanide (4) and cyanoform (5) in $[D_8]$ THF (400 MHz) at different temperatures starting with -95 °C (top).

In remarkable contrast to our NMR spectra of 5 in [D₈]THF, which indicate very broad signals at -50 °C (see, for example, Figures 1 and 2), sharp NMR signals were previously reported ^[10] and shown for the same compound measured in [D₆]acetone at -45 °C. When we prepared 5 from 2 or by photolysis of 7 and tried to measure low-temperature NMR spectra in [D₆]acetone, we did not get the corresponding signals because an addition of the substrate at the solvent molecule completely transformed 5 into the alcohol 10a (Scheme 3). This surprising result was confirmed by treating 5 with non-deuterated acetone, which led to the product 10b. After warming the solutions of 10a or 10b to room temperature, decay of the adducts was observed, possibly via liberation and decomposition of 5. The reaction of 5 with acetaldehyde produced alcohol 10c, which was slightly more stable and could be characterized by NMR spectroscopy at ambient temperature.



Scheme 3. Reaction of cyanoform (5) with acetone or acetaldehyde.

A closer look revealed that previous NMR spectra were not measured in [D₆]acetone, but liquid sulfur dioxide was used as solvent and [D₆]acetone as external (!) standard (and lock).^[15] When we synthesized **5** from **2** and tried to measure ¹H and ¹³C NMR spectra of the corresponding suspension in liquid sulfur dioxide at -45 °C, we did not get any signal of cyanoform (**5**), even after long-term measurement. After addition of trifluoroacetic acid, however, we detected ¹H and ¹³C NMR spectra of **5** with sharp signals, which are very similar to those published^[10] previously. Obviously, the presence of an additional Brønsted acid has a strong effect on the NMR spectra of **5** in liquid sulfur dioxide, and measurement of **5** in this solvent at -45 °C is perhaps not possible without such an acid, for example, acidic salt **6**.

When we completely removed the solvents from 2 at -20 °C and 10^{-2} Torr and rapidly used the residue for sublimation into an uncooled horizontal glass tube by heating from 20 to 50 °C at 10^{-2}

Torr with an open connection to the pump, we obtained brilliant colorless crystals.^[13] It turned out that the product consisted of single crystals of cyanoform (**5**), which were appropriate for X-ray diffraction studies (Figure 3). As a pure solid, **5** obviously is by far more stable than the same compound in solution. Most likely, this can be attributed to the presence of layers in the solid state owing to intermolecular hydrogen bridges. The C-H donor function is involved in the formation of three different hydrogen bonds with the correponding N atoms as acceptor function (see Figures S2–S4 and Table S2 in the Supporting Information). These non-classical hydrogen bridges are also remarkable in view of the properties of **5** as one of the strongest carbon acids.



Figure 3. Molecular structure of cyanoform (5) (50% probability level).

The Raman spectrum of crystalline 5 (see Supp. Info., Figure S5) reveals strong and characteristic CH and CN stretching bands at 2882.8 and 2283.3 cm⁻¹ (Table 1). These band positions are surprisingly close to those reported previously^[10] for a mixture of **5** and **6** (2885 and 2287 cm⁻¹, respectively), indicating that if intermolecular interactions to the terminal H or N atoms of 5 are considered, there influence on these two band positions is at least very similar. The influence of such intermolecular interactions can be evaluated by comparison with a matrix-isolation spectrum of 5, where molecules of 5 will be isolated at cryogenic temperatures (< 20 K) in a host of solid argon This was achieved by evaporating colorless crystalline 5 at 7 °C in a flow of argon at a rate of 1 mL min⁻¹ and its deposition on the matrix support, a rhodium plated mirror cooled to 4 K. The argonmatrix isolation spectrum of 5 is shown in Figure 4, and the band positions are listed and compared to quantum chemical results in Table 1. As expected, the CH stretching vibration of the isolated molecule appeared at 2927.6 cm⁻¹, and thus at significantly higher



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Table 1. Experimental IR and Raman as well as calculated band positions (in cm^{-1}) of cyanoform (5) and their tentative assignment.

IR ^[a]	Raman ^[b]	B3LYP	Anharm.	Assignment
		(IR-Int.) ^[c]	CCSD(T) ^[d]	-
2927.6	2882.8	2909 (12)	2929	v(HC), A₁
2806.1				2268 + 556
	2288,3	2302 (0)	2260	v _s (CCN), A ₁
2268.1	2269.6	2294 (2)	2261	v _a (CCN), E
1267.9	1254.3	1244 (7)	1266	δ(HCC), E
	1266.7			
1023.4	1025.9	987 (37)	1012	v _a (CC ₃), E
1013.9 ^[e]				
889.5				556 + 334
822.5	822.4	811 (5)	822	v _s (CC ₃), A ₁
	570.9	558 (0)	556	$\delta_{s}(CC_{3}), A_{1}$
556.4	561.2	552 (1)	553	$\delta_a(CC_3), E$
	348.8 ^[f]	345 (0)	336	δ _s (CCN), A ₂
	348.8 ^[f]	344 (0)	334	δ _a (CCN), E
	164.4	162 (25)	161	$\delta_{s}(C(CN)_{3}), A_{1}$
	149.0	128 (15)	128	$\delta_a(C(CN)_3), E$

[a] Isolated in solid argon at 4 K (strongest matrix sites are listed); [b] solid sample, see Figure S5; [c] This work: DFT B3LYP/cc-pVTZ level, IR intensities in parentheses (in km mol⁻¹); [d] Ref. 9a, computed by adding VPT2 anharmonic corrections calculated at MP2/cc-pVTZ level to harmonic frequencies calculated at the CCSD(T)/cc-pVTZ level; [e] strong matrix site; [f] overlapped.

wavenumbers than in the solid state. The matrix-isolation spectrum is in excellent agreement with previously calculated anharmonic vibrational frequencies obtained by adding VPT2 anharmonic frequency corrections (predicted at the MP2/cc-pVTZ level) to the CCSD(T)/cc-pVTZ harmonic frequencies.^[9a] In agreement with preliminary TD-DFT B3LYP/cc-pVTZ calculations (see Supp. Info., Table S3), our attempts to initiate a photo-rearrangement of **5** failed (Figure S6).

After we have successfully proved that 5 can not only be obtained from 2, but also by heating or irradiating azide 7 in [D₈]THF solution, the question arises whether the conversion of 7 to 5 is a pure monomolecular process that can be studied under matrix-isolation conditions. Photolysis of 7 probably involves the azirine intermediate 8, where the rearrangement from 8 to 5 has recently been shown requires an appreciable energy to surmount a barrier of about 60 kcal mol⁻¹ at the CCSD(T)/cc-pVTZ level.^[9a] We have studied the photolysis of 7 isolated in a solid argon matrix, prepared by gently heating (30-50 °C) 7 and its deposition with an excess of argon gas (ca. 1:1000) onto the matrix support held at 15±1 K. To facilitate the band assignment, the experiments were repeated using ¹⁵N labelled 7, in which either the terminal or the α -nitrogen atoms of the azide group were substituted by ¹⁵N. Spectra of 7 and of ¹⁵N-7 are shown in Figure S7. Band positions and their ^{14/15}N isotope shifts are compared to calculated values in Table S4. As expected, photolysis of 7 or ¹⁵N-7 using a solidstate laser of λ = 266 nm efficiently produces the corresponding isotopologues 8 and ¹⁵N-8, respectively (Figure S8 and Table S5). However, a second photolysis product was detected, showing a rather strong N-H and a strong ketenimine absorption at 3360 and 2096 cm⁻¹, respectively (Figure S8). Based on these vibrations and their characteristic 14/15N isotope shifts, we assigned this species to the hitherto unknown ketenimine 3 (Table S6). We note that absorptions of cyanoform (5) were not detected in these experiments, and attempts to produce 5 by irradiating the deposit failed.^[13] We can also exclude the formation of 3 from 8 by secondary photolysis reactions, because i) further photolysis did not change the relative amount of the primary photolysis products, and ii) the **8** to **3** conversion is at least a two-step process, involving a rather high-barrier H-migration and subsequent C-N bond cleavage.^[9a] Thus **3** is a product of the photolysis of **7**. This assumption is verified by our DFT B3LYP/cc-pVTZ calculations (Figure S8), which revealed a concerted reaction of **7** by elimination of N₂ with concurrent hydrogen shift from the carbon to the nitrogen atom (see Figure S9).

In conclusion, detection of ketenimine **3** was possible by irradiation of azide **7** in noble gas matrix only. We assume that photolysis in solution leads also to the primary product **3** since similar transformations^[16] are known for other vinyl azides. In solution, however, very rapid tautomerism most probably induces the reaction **3** \rightarrow **5**. Pure cyanoform (**5**) is obviously available via special sublimation of **4**. Not only by using neat **5** (see Scheme 3) or **2**, but also by gentle thermolysis of **7**, we were able to initiate nucleophilic attack at different types of carbon electrophiles. We will report on these reactions, which may be valuable for carbon–carbon bond formation and heterocyclic ring transformation, in the near future.

Conflict of Interest

The authors declare no conflict of interest.

Keywords: azides • cyanides • cyanoform • ketenimines reactive intermediates

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- [1] H. Schmidtmann, Ber. Dtsch. Chem. Ges. 1896, 29, 1168–1175.
- [2] A. Hantzsch, G. Osswald, Ber. Dtsch. Chem. Ges. 1899, 32, 641–650.
- [3] S. Trofimenko, J. Org. Chem. **1963**, 28, 217–218.
- [4] For a more detailed description of the cyanoform story, see the excellent introduction in ref. [5].
- [5] D. Šišak, L. B. McCusker, A. Buckl, G. Wuitschik, Y.-L. Wu, W. B. Schweizer, J. D. Dunitz, *Chem. Eur. J.* **2010**, *16*, 7224–7230.
- [6] B. Bak, H. Svanholt, J. Mol. Struct. **1977**, 37, 153–156.
- [7] H. Bock, R. Dammel, Z. Naturforsch. **1987**, 42b, 315–322.
- [8] a) R. H. Boyd, J. Phys. Chem. 1963, 67, 737–744; b) E. Raamat, K. Kaupmees, G. Ovsjannikov, A. Trummal, A. Kütt, J. Saame, I. Koppel, I. Kaljurand, L. Lipping, T. Rodima, V. Pihl, I. A. Koppel, I. Leito, J. Phys. Org. Chem. 2013, 26, 162–170; c) A. Kütt, T. Rodima, J. Saame, E. Raamat, V. Mäemets, I. Kaljurand, I. A. Koppel, R. Y. Garlyauskayte, Y. L. Yagupolskii, L. M. Yagupolskii, E. Bernhardt, H. Willner, I. Leito, J. Org. Chem. 2011, 76, 391–395.
- a) M. Szczepaniak, J. Moc, J. Phys. Chem. A 2017, 121, 1319–1327; b)
 H. Brand, J. F. Liebman, A. Schulz, Eur. J. Org. Chem. 2008, 4665–4675;
 c) B. Bak, C. Bjorkman, J. Mol. Struct. 1975, 25, 131–140; d) S. S.

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Stoyanov, J. A. Tsenov, I. G. Binev, Asian Chem. Lett. 2009, 13, 155–162; e) S. A. Elroby, Chem. Cent. J. 2016, 10, 20.

- [10] T. Soltner, J. Häusler, A. J. Kornath, Angew. Chem. 2015, 127, 13979– 13980; Angew. Chem. Int. Ed. 2015, 54, 13775–13776.
- [11] a) H. Schubert, M. Regitz, *Synthesis* **1982**, 149–151; b) K. Friedrich, *Angew. Chem.* **1967**, *79*, 980; *Angew. Chem. Int. Ed. Engl.* **1967**, *6*, 959– 960; c) P. P. Koriuta, A. I. Kalenskaya, V. I. Shevchenko, *Zh. Obshch. Khim.* **1971**, *41*, 2390–2395.
- [12] For investigation of **8** by quantum chemical methods, see ref. [9a].
- [13] For details, see the Supporting Information.
- [14] For the chemistry of vinyl azides, see: a) K. Banert in Organic Azides, Syntheses and Applications (Eds.: S. Bräse, K. Banert), Wiley, Chichester, 2010, pp. 115–166; b) S. J. Collier in Science of Synthesis, Vol. 33 (Ed.: G. A. Molander), Thieme, Stuttgart, 2006, pp. 541–563; c) K. Banert in Houben-Weyl, 4th ed., Vol. E15 (Eds.: H. Kropf, E. Schaumann), Thieme, Stuttgart, 1993, pp. 818–875; d) A. Hassner in Azides and Nitrenes (Ed.: E. F. V. Scriven), Academic Press, Orlando, 1984, pp. 35–94; e) G. L'abbé, Angew. Chem. 1975, 87, 831–837; Angew. Chem. Int. Ed. Engl. 1975, 14, 775–782; f) G. Smolinsky, C. A. Pryde in The Chemistry of the Azido Group (Ed.: S. Patai), Wiley, New York, 1971, pp. 555–585; g) G. L'abbé, A. Hassner, Angew. Chem. 1971, 83, 103–109; Angew. Chem. Int. Ed. Engl. 1971, 10, 98–104; h) B. Hu, S. G. DiMagno, Org. Biomol. Chem. 2015, 13, 3844–3855.
- [15] T. Soltner, dissertation, LMU München (Germany), 2011.
- [16] a) G. Smolinsky, J. Org. Chem. 1962, 27, 3557–3559; b) J. Finnerty, U. Mitschke, C. Wentrup, J. Org. Chem. 2002, 67, 1084–1092.

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Held incommunicado in argon matrix, a long-sought ketenimine can be detected after irradiation of an appropriate vinyl azide, whereas photolysis or thermolysis of the same precursor in solution leads to cyanoform, which is also available as single crystals from potassium tricyanomethanide.



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