

Thermodynamic Properties of Organic Compounds. 3. Sublimation Enthalpy and Heat Capacities of 2,4,6-Trimethylbenzotrile *N*-Oxide

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Experimental heat capacities (6–319 K) and enthalpy of sublimation, $\Delta_{\text{sub}}H_m^\circ(313.5\text{K}) = 87.52 \pm 0.53 \text{ kJ mol}^{-1}$, are reported for 2,4,6-trimethylbenzotrile *N*-oxide. Heat capacity data show a reversible solid-phase transition in the 217–235 K temperature interval, for which the following thermodynamic functions have been computed: $T_{\text{tr}} = 224.3 \text{ K}$; $\Delta_{\text{tr}}H_m = 80 \pm 4 \text{ J mol}^{-1}$; $\Delta_{\text{tr}}S = 0.35 \pm 0.02 \text{ J mol}^{-1} \text{ K}^{-1}$. Thermodynamic properties of 2,4,6-trimethylbenzotrile *N*-oxide in the condensed state are tabulated at every 5 or 10 K interval throughout the 0–320 K temperature interval.

Introduction

Nitrile *N*-oxides are highly reactive organic compounds prepared from oxidation of aldoximes and dehydration of nitro compounds. Reported half-lives of aliphatic nitrile *N*-oxides range from a few seconds to minutes before rearrangement to isocyanates or polymerization occurs. Aromatic nitrile *N*-oxides are considerably more stable, particularly 1,4-dicyanobenzene di-*N*-oxide (1, 2), which can be stored for several months without noticeable degradation. Bulky substituents in the ortho positions of the benzene ring or on the α -carbon aliphatic derivatives enhance stability. Sterically hindered nitrile *N*-oxides such as 2,6-dimethylbenzotrile *N*-oxide and di-*tert*-butylacetone nitrile *N*-oxide are "permanently stable" at room temperature (3–5).

Chemical reactivity of the nitrile *N*-oxides undoubtedly arises because of the ionic nature of the N–O dative bond. Nuclear magnetic resonance ^{13}C chemical shift data for 2,4,6-trimethylbenzotrile *N*-oxide (6) suggest that the carbon atom carries substantial negative charge (i.e., the $\text{R}-\text{C}=\text{N}^+=\text{O}$ resonance form) and are in accord with published X-ray crystallographic bond distances for 4-methoxy-2,6-dimethylbenzotrile *N*-oxide (7). Surprisingly, very few precise thermochemical quantities are available in the literature for nitrile *N*-oxides. Acree, Pilcher, and co-workers (8, 9) previously reported the standard enthalpies of combustion and sublimation for 2,4,6-trimethylbenzotrile *N*-oxide, 2,4,6-trimethoxybenzotrile *N*-oxide, and 1,4-dicyanobenzene di-*N*-oxide as determined by static bomb calorimetric, "vacuum sublimation" drop microcalorimetric, and Knudsen effusion vapor pressure methods, respectively. Because of the scarcity of experimental data for this relatively novel class of compounds, particularly low-temperature thermodynamic data, we initiated a comprehensive thermodynamic study of 2,4,6-trimethylbenzotrile *N*-oxide, which included a recently published redetermination of the enthalpy of combustion (10), as well as heat capacity and sublimation measurements. In this paper we report results from the heat capacity and sublimation measurements.

Experimental Methods

2,4,6-Trimethylbenzotrile *N*-oxide was prepared by the procedures outlined by Grundmann and Dean (5) and Beltrame et al. (11). Hydroxylamine hydrochloride was added to a hot solution of 2,4,6-trimethylbenzaldehyde in methanol, and the resulting aldoxime was converted to the nitrile *N*-oxide with sodium hypobromite. The crude sample was further purified by three recrystallizations from a benzene–hexane mixture. Elemental analysis on the purified sample was in excellent agreement with calculated values. Anal. Calcd for $\text{C}_{10}\text{H}_{11}\text{NO}$: C, 74.51; H, 6.88; N, 8.69. Found: C, 74.58; H, 6.94; N, 8.62. High-performance liquid chromatographic analysis with ultraviolet detection at 254 nm further showed that the sample contained a single minor impurity. The sample purity was estimated to be 99.8+ % on the basis of the areas under the two observed chromatographic peaks, assuming identical detector responses for both 2,4,6-trimethylbenzotrile *N*-oxide and the unidentified impurity, which was likely unreacted 2,4,6-trimethylbenzaldehyde or a reaction byproduct. The sample was stored in a cool, dark place for several weeks and shipped to the Soviet Union via air mail during a period of heightened airport security.

Experimental procedures and instrumentation are described in detail elsewhere (12, 13). Briefly, the heat capacity was measured in a vacuum adiabatic calorimeter TAU-1, fabricated by VNIIFTRI (National Scientific and Research Institute for Physical-technical and Radio-technical Measurements) in Moscow. The investigated sample (tablet with mass of 0.537 34 g) was put in a stainless-steel ampule filled with heat-exchange helium gas and sealed with an indium gasket. A small drop of silicon polymer (CKTH) was placed on the external surface of the ampule to improve heat exchange between the ampule and calorimetric device. The ampule was tightly inserted into the calorimetric sleeve and provided with a Manganin heater. The sleeve was suspended by three nylon cords within the adiabatic shield, which was also provided with a Manganin heater. The relative temperature between the adiabatic shield and the calorimetric sleeve was determined by a four-junction differential thermocouple: (0.999Cu + 0.001Fe)-to-Chromel. The electric potential of

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Table I. Experimental Molar Heat Capacities of Crystalline 2,4,6-Trimethylbenzotrile *N*-Oxide

$\langle T \rangle / K$	$\Delta T / K$	$C_{p,m} / R^a$	$\langle T \rangle / K$	$\Delta T / K$	$C_{p,m} / R^a$	$\langle T \rangle / K$	$\Delta T / K$	$C_{p,m} / R^a$	$\langle T \rangle / K$	$\Delta T / K$	$C_{p,m} / R^a$
Series 1											
5.974	0.1786	0.1031	9.319	0.5405	0.3211	13.78	0.5978	0.8550	17.28	0.7544	1.376
6.290	0.3712	0.1179	9.955	0.7041	0.3979	14.36	0.5509	0.9413	17.46	0.6791	1.406
6.674	0.3396	0.1337	10.64	0.6183	0.4749	14.63	1.0314	0.9830	18.01	0.6967	1.495
7.100	0.4594	0.1567	11.25	0.5584	0.5401	15.10	0.8938	1.041	18.69	0.6488	1.609
7.716	0.5557	0.1970	11.80	0.5109	0.6044	15.96	0.8040	1.173	20.24	0.7611	1.859
8.321	0.4991	0.2382	12.43	0.7221	0.6869	16.48	0.8243	1.254	20.98	0.7106	1.985
8.808	0.4487	0.2788	13.14	0.6562	0.7645	16.74	0.7323	1.301			
Series 2											
5.744	0.1926	0.09582	10.27	0.6392	0.4290	16.52	0.6801	1.251	33.06	1.3523	3.950
5.967	0.1845	0.09959	10.89	0.5678	0.5039	17.58	0.4759	1.421	34.45	1.4410	4.220
6.283	0.3783	0.1189	11.46	0.5153	0.5704	19.40	0.7012	1.720	35.84	1.3371	4.470
6.670	0.3449	0.1362	12.05	0.6123	0.6373	20.13	0.6381	1.850	37.13	1.2497	4.713
7.026	0.3174	0.1533	12.66	0.5590	0.7115	20.83	0.8129	1.968	38.87	2.2212	4.905
7.488	0.4975	0.1783	13.21	0.5167	0.7799	21.88	1.2673	2.132	40.98	2.0289	5.233
7.966	0.4442	0.2119	13.72	0.4841	0.8367	23.17	1.1115	2.356	43.15	2.3347	5.488
8.129	0.3647	0.2308	14.25	0.5052	0.9265	24.29	1.0634	2.534	45.37	2.1396	5.812
8.501	0.3376	0.2564	14.81	0.5268	1.001	25.48	1.4859	2.722	49.50	2.1912	6.433
8.846	0.3159	0.2797	15.53	0.7614	1.107	26.89	1.3436	2.959	51.61	2.0502	6.736
9.618	0.4373	0.3554	16.00	0.5687	1.177	28.46	1.7846	3.197			
Series 3											
52.85	2.8425	6.858	74.12	5.2151	9.761	93.11	3.4957	11.88	108.67	3.7151	13.48
56.20	3.8732	7.366	81.72	4.3613	10.68	96.99	3.7450	12.31	112.30	3.6203	13.80
60.44	4.6442	7.948	85.81	3.8208	11.10	100.99	3.9800	12.68			
66.53	7.5672	8.768	89.54	3.6447	11.50	104.90	3.8390	13.08			
Series 4											
69.27	3.0533	9.108	100.82	4.1825	12.67	127.10	3.4300	15.11	161.66	4.4656	17.76
73.97	4.5687	9.755	104.93	4.0285	13.08	130.50	3.3597	15.41	166.14	4.3922	18.14
78.38	4.2488	10.28	108.89	3.8945	13.47	135.90	4.8949	15.76	170.56	4.3232	18.51
86.39	3.7901	11.17	112.73	3.7763	13.85	142.90	4.8359	16.22	174.92	4.2615	18.86
90.10	3.6183	11.58	116.45	3.6737	14.21	147.70	4.7295	16.61	179.22	4.2025	19.21
93.65	3.4731	11.95	120.09	3.5830	14.52	152.46	4.6343	16.98			
97.06	3.3472	12.32	123.63	3.5022	14.84	157.10	4.5454	17.39			
Series 5											
182.73	6.2998	19.49	196.14	7.8026	20.60	211.66	7.4946	21.97	219.22	7.3655	22.62
189.05	6.1818	20.04	203.97	7.6388	21.30	216.95	5.3945	22.43			
Series 6											
219.86	0.2019	22.81	223.07	1.1401	23.48	225.48	1.1402	23.25	231.52	1.1393	22.77
221.26	1.1486	23.07	224.09	0.1689	23.71	227.01	0.1697	23.14	232.73	1.1403	22.61
221.73	0.1706	23.18	224.31	0.1688	23.74	227.92	0.1680	23.01	233.73	2.6160	22.61
222.57	0.1705	23.30	224.54	0.1695	23.40	228.35	2.6178	23.01	235.20	1.1370	22.62
222.78	0.1698	23.43	224.76	0.1692	23.52	230.31	1.1402	22.81			
222.99	0.1695	23.56	224.98	0.1698	23.22	231.04	2.6235	22.61			
Series 7											
238.57	5.2786	22.86	254.46	5.1240	24.02	275.66	6.1069	26.89	300.71	5.8991	28.67
241.76	2.5808	23.05	259.68	5.0752	24.46	281.94	6.0466	27.38	306.91	5.7993	29.90
244.87	2.9607	23.29	264.86	5.0123	25.14	288.20	6.0059	27.68	313.05	5.6867	31.40
249.20	5.1752	23.61	269.97	4.9344	26.10	294.46	5.9578	28.09	319.22	5.5870	32.81

^a Note: The mean molar $C_{p,m}$ in the temperature interval ΔT are given at the mean temperature $\langle T \rangle$. $R = 8.31451 \text{ J mol}^{-1} \text{ K}^{-1}$.

the thermocouple after preliminary amplification went to the terminals of an analogue regulation block, which assured automatic adiabatic shield control with proportional integral/differential action. The thermometric measurements were made by a capsule-type (rhodium + 0.5% atomic Fe) resistance thermometer ($R_0 = 45.32 \Omega$) installed on the inner surface of the adiabatic shield. The thermometer was fabricated and calibrated by VNIIFTRI. The reliability of the calorimetric apparatus was established by measuring the heat capacities of standard benzoic acid K-1 and high-purity copper [sample OSCh 11-4, standard of USSR]. The probable error of the heat capacity values is approximately 2% at liquid helium temperature and 1% in the interval 15–20 K, decreases to 0.4% for temperatures up to 40 K, and does not exceed 0.4% from 40 to 320 K. Heat capacities of all auxiliary substances were included in the 2,4,6-trimethylbenzotrile *N*-oxide heat capacity calculations. The heat capacities of the sample at temperatures 10, 100, and 300 K were

approximately 45%, 30%, and 35% of the total heat capacity of the calorimeter, respectively.

The sublimation enthalpy of 2,4,6-trimethylbenzotrile *N*-oxide was determined by a heat-conduction calorimeter of the Calvet type (13). We constructed two vacuum calorimetric cells (A and B) for measuring enthalpies of vaporization in accordance with the ampule method to be used with a commercially available differential microcalorimeter MID-2000. The cells were calibrated by enthalpies of vaporization of four reference substances: benzoic acid K-1, naphthalene twice sublimed from a sample "pure for analysis" (standard of the USSR), *n*-decane of gas chromatographic standard grade, and doubly distilled water. The studied substances (mass from 0.01 to 0.15 g) were preliminarily outgassed in vacuum and sealed in glass ampules. Despite the significant differences of saturated vapor pressures of these reference substances (2–3160 Pa) at the experimental temperatures (293–342 K), we obtained in all cases practically identical

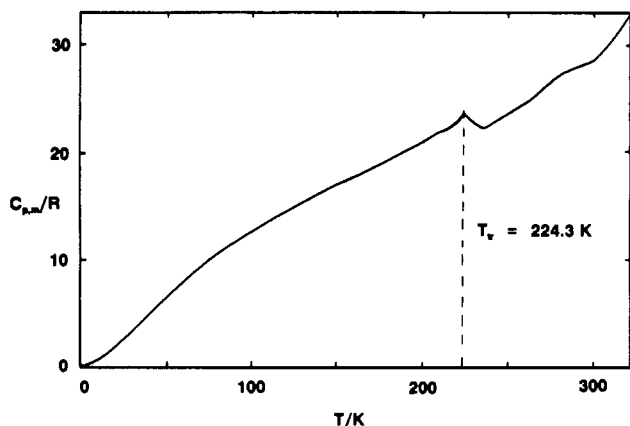


Figure 1. Temperature dependence of $C_{p,m}/R$ of 2,4,6-trimethylbenzotrile *N*-oxide. The vertical line indicates the temperature of the phase transition. The solid curve represents the "smoothed" heat capacity data.

Table II. Molar Thermodynamic Functions of 2,4,6-Trimethylbenzotrile *N*-Oxide at Select Temperatures

T/K	$C_{p,m}/R$	$\Delta^{\circ}T S^{\circ}_m/R$	$\Delta^{\circ}T H^{\circ}_m/RT$	$\Phi^{\circ}_m(T,0)/R$
0	0	0	0	0
5	0.05836	0.01945	0.01459	0.004863
10	0.3993	0.1432	0.1058	0.03735
15	1.032	0.4183	0.3036	0.1147
20	1.823	0.8202	0.5819	0.2383
25	2.652	1.317	0.9140	0.4034
30	3.451	1.871	1.270	0.6009
35	4.311	2.466	1.641	0.8246
40	5.070	3.093	2.025	1.069
45	5.779	3.732	2.403	1.329
50	6.497	4.377	2.776	1.601
60	7.909	5.687	3.515	2.173
70	9.213	7.006	4.237	2.769
80	10.46	8.321	4.940	3.381
90	11.56	9.617	5.615	4.002
100	12.62	10.89	6.263	4.627
110	13.59	12.14	6.885	5.254
120	14.49	13.36	7.482	5.879
130	15.32	14.55	8.053	6.500
140	16.09	15.72	8.601	7.117
150	16.81	16.85	9.124	7.729
160	17.61	17.96	9.629	8.334
170	18.45	19.06	10.12	8.932
180	19.26	20.13	10.61	9.525
190	20.05	21.19	11.08	10.11
200	20.93	22.25	11.55	10.69
210	21.81	23.29	12.02	11.27
220	22.84	24.32	12.49	11.84
230	22.92	25.35	12.95	12.40
240	22.94	26.32	13.36	12.96
250	23.66	27.27	13.75	13.52
260	24.49	28.21	14.15	14.06
270	26.10	29.16	14.56	14.60
280	27.25	30.14	15.00	15.14
290	27.79	31.10	15.43	15.68
298.15	28.40	31.88	15.77	16.11
300	28.59	32.06	15.85	16.21
310	30.63	33.03	16.29	16.73
320	32.99	34.04	16.78	17.26

values of cell constants, which were correspondingly $K_A = 185.65 \pm 0.43 \text{ mV W}^{-1}$ and $K_B = 200.00 \pm 0.63 \text{ mV W}^{-1}$. The uncertainty of the measurements of enthalpies of vaporization was not greater than 0.5–1.0% over a large range of saturated vapor pressures provided that the measured powers of heat flows did not exceed 0.5 W. The calorimetric procedure is described in greater detail elsewhere (13).

Results and Discussion

Results of the low-temperature heat capacity measurements

obtained in seven series of measurements are reported in Table I, where $C_{p,m}$ is the experimental molar heat capacity at the mean temperature of the experiment (T) and ΔT is the temperature increase of the calorimeter corrected for heat exchange with the environment. Care was taken to ensure that thermal equilibrium was achieved. Measurements between 5 and 12 K required unusually long equilibration times, and were 5–6 times greater than those typically noted for organic samples. The reason for the longer equilibration times is not clear; however, we suspect that it may result from slow adsorption/desorption of the helium heat-exchange gas upon the solid sample at these low temperatures. Examination of the heat capacity data (see Figure 1) reveals that there is a small anomaly in the $C_{p,m}/R$ versus T curve around 217–235 K caused by a reversible solid-phase transformation. A second unexplained anomaly (not reversible, likely trace occluded solvent removal) is observed around 260–280 K, and at higher temperatures there is a sharp increase in the $C_{p,m}$ of the substance. Inspection of the solid sample after the heat capacity measurements was finished showed no visual color change and no signs of thermal degradation.

Table II lists the thermodynamic functions of crystalline 2,4,6-trimethylbenzotrile *N*-oxide calculated at every 5 or 10 K temperature change over the 0–320 K temperature interval. The Debye heat capacity equation was used in extrapolating measured heat capacity data to $T \rightarrow 0$:

$$C_{p,m} = 3RD(\Theta_D/T) \quad (1)$$

where $\Theta_D = 79.4 \text{ K}$ is the characteristic temperature based upon $C_{p,m}$ data in the temperature interval of 5.74–8.31 K. Calculated thermodynamic functions at 298.15 K are

$$C_{p,m} = 236.10 \pm 0.94 \text{ J mol}^{-1} \text{ K}^{-1} \quad (2)$$

$$H^{\circ}_{298.15} - H^{\circ}_0 = 39101 \pm 158 \text{ J mol}^{-1} \quad (3)$$

$$S^{\circ}_{298.15} - S^{\circ}_0 = 265.08 \pm 1.15 \text{ J mol}^{-1} \text{ K}^{-1} \quad (4)$$

$$(H^{\circ}_{298.15} - H^{\circ}_0)/T = 131.15 \pm 0.53 \text{ J mol}^{-1} \text{ K}^{-1} \quad (5)$$

$$-(G^{\circ}_{298.15} - H^{\circ}_0)/T = 133.93 \pm 1.27 \text{ J mol}^{-1} \text{ K}^{-1} \quad (6)$$

On the basis of an analysis of the nonregular contribution to the heat capacity over the 217–235 K temperature interval, the following thermodynamic functions have been computed:

$$T_{tr} = 224.3 \text{ K} \quad (7)$$

$$\Delta_{tr}H_m = 80 \pm 4 \text{ J mol}^{-1} \quad (8)$$

$$\Delta_{tr}S = 0.35 \pm 0.02 \text{ J mol}^{-1} \text{ K}^{-1} \quad (9)$$

for the reversible solid-phase transformation. Transition properties were based upon three sets of heat capacity measurements [one set is provided in Table I as series 6] over the temperature interval of 217–235 K:

$$\Delta_{tr}H_m = \int_{T=217}^{T=235} C_p(\text{excessive}) dT \quad (10)$$

$$\Delta_{tr}S = \int_{T=217}^{T=235} [C_p(\text{excessive})/T] dT \quad (11)$$

where $C_p(\text{excessive})$ represents the difference between the experimental and regular heat capacity; i.e., $C_p(\text{excessive}) = C_p(\text{exp}) - C_p(\text{reg})$. The value of $C_p(\text{reg})$ was found by extrapolating the gradual parts of the heat capacity as a function of temperature, $C_p = f(T)$. The transition temperature was computed as the peak temperature in the C_p versus T curve (see Figure 1). Uncertainties represent the mean squared deviation of the mean arithmetic value by the

Table III. Calorimetric Results for the Sublimation Enthalpy of 2,4,6-Trimethylbenzotrile *N*-Oxide^a

no.	type of cell	<i>m</i> /g	τ_2 /s	$\int_{\tau_0}^{\tau_2} \Delta E d\tau / (\text{mV s})$	$\Delta_{\text{sub}}H/\text{J}$	$\Delta_{\text{sub}}H^\circ_m / (\text{kJ mol}^{-1})$
1	A	0.054 05	20 000	5416.0	29.173	87.01
2	A	0.052 49	10 317	5333.3	28.728	88.22
3	A	0.043 45	16 327	4403.2	23.718	87.99
4	B	0.020 62	9 200	2226.4	11.132	87.01
5	B	0.044 65	5 021	4844.0	24.220	87.44
6	B	0.056 55	19 035	6111.9	30.559	87.42
						av: 87.52

^a The average square deviation of the average arithmetic value is 0.20 kJ mol⁻¹. Taking into account the Student's *t* test criterion (*t*₉ = 2.571) for the 95% confidence interval, we obtain $\Delta_{\text{sub}}H^\circ_m(313.5\text{K}) = 87.52 \pm 0.53$ kJ mol⁻¹. Note: The sublimation enthalpy $\Delta_{\text{sub}}H$ of the specimen and the molar enthalpy of sublimation $\Delta_{\text{sub}}H^\circ_m$ were calculated from the expressions $\Delta_{\text{sub}}H = K^{-1} \int_{\tau_0}^{\tau_2} \Delta E d\tau$ and $\Delta_{\text{sub}}H^\circ_m = (\Delta_{\text{sub}}H)M/m$, where *m* is the specimen mass, *M* is the molar mass, *K* is the calorimeter cell constant, ΔE is the thermocouple potential difference corresponding to the temperature imbalance between the cell and calorimeter thermostat at the moment τ , and τ_2 is the experiment duration.

Student's *t* test factor (*t*₉ = 4.3) for three sets of replicate heat capacity measurements.

Sublimation measurements were performed on the sample as received via air mail and after subsequent purification by vacuum sublimation to remove trace impurities which may have resulted from prolonged shelf storage and/or X-ray inspections at airport security checkpoints. For the original sample four sets of measurements were performed; however, in every case the sample was not completely vaporized after 5 h in vacuum at 318.4 K. A significant quantity of brown material was found on the bottom of the calorimetric cell. Because of the incomplete vaporization, the calculated enthalpies varied from 384 to 892 J g⁻¹ and the values were not included in the statistical analysis. Six sets of measurements on the purified sample gave very reproducible sublimation enthalpies as shown in Table III. The average of $\Delta_{\text{sub}}H^\circ_m(313.5\text{K}) = 87.52 \pm 0.53$ kJ mol⁻¹ is in excellent agreement with an earlier value of $\Delta_{\text{sub}}H^\circ_m(319.2\text{K}) = 87.41$

± 1.81 kJ mol⁻¹ (10), which was based upon an integral Knudsen effusion vapor pressure method.

On the basis of these two values corrected to 298.15 K, the recommended value of sublimation enthalpy is $\Delta_{\text{cr}}H^\circ_m(298.15\text{K}) = 87.8 \pm 1.9$ kJ mol⁻¹. Assuming the residual entropy equals zero, $S^\circ_m(c,298.15\text{K}) = 265.08$ J mol⁻¹ K⁻¹. The saturated vapor pressure $P^\circ(298.15\text{K})$ can be calculated by insignificant extrapolation of experimental data to 298.15 K using the polynomial dependence reported earlier (10). Then $P^\circ(298.15\text{K}) = 0.109$ Pa. In accordance with this set of data, the standard entropy of the gaseous 2,4,6-trimethylbenzotrile *N*-oxide at 298.15 K is equal to

$$S^\circ_m(g,298.15\text{K}) = S^\circ_m(c,298.15\text{K}) + \Delta_{\text{cr}}H^\circ_m(298.15\text{K})/298.15\text{K} + R \ln [P^\circ(298.15)/101325 \text{ Pa}] = 445.3 \text{ J mol}^{-1} \text{ K}^{-1} \quad (12)$$

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