Thermochimica Acta, 219 (1993) 97–104
Elsevier Science Publishers B.V., Amsterdam

Thermodynamic properties of organic compounds. Part 4. First update of enthalpy of fusion and melting point temperature compilation

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(Received 19 August 1992; accepted 6 September 1992)

Abstract

Published enthalpies of fusion and melting point temperatures have been gathered from the chemical literature and are presented in tabular form according to increasing carbon and hydrogen atom numbers. References are also provided to indicate the literature sources consulted.

INTRODUCTION

The number of experimental and theoretical thermochemical investigations on organic substances and mixtures has increased significantly in recent years as society has become more technologically oriented. The development of micro-methods in combusion calorimetry, combined with "vacuum drop" sublimation and/or Knudsen indirect pressure–temperature measurement, has enabled standard gas-phase enthalpies of formation of small sample sizes (50 mg or less) to be determined to within an average uncertainty of about 0.2%. Modern flow calorimeters, continuous dilution dilatometers and vibrating-tube densimeters permit the experimental determination of excess enthalpies, heat capacities and volumes of non-electrolyte liquid mixtures to be determined with convenience and accuracy. Automated chromatographic head-space sampling devices have reduced the experimental time needed for the determination of excess Gibbs free energies nad activity coefficients through conventional vapor pressure measurements. But even with today's modern instrumentation, it would be physically impossible to measure the thermodynamic properties for the 6 million plus distinct known chemical substances [1], let alone the seemingly infinite number of possible binary, ternary and higher-order multicomponent mixtures.

To address this problem, researchers have turned to predictive methods as a means to generate desired quantities from structural information. Group contribution methods for enthalpies of formation [2–7] , enthalpies

of vaporization [2, 8], solid and liquid heat capacities [2, 9, 10], and for activity coefficients, excess free energies and excess enthalpies of mixing [11–19] have been in existence for several years now. Enthalpies of fusion were largely ignored, however, perhaps because of the various solid–solid, solid–liquid-crystalline and other mesomorphic transitions which may occur before the compound actually melts. It was only recently that Chickos and coworkers [20, 21] derived a group additivity approach for estimating entropies of fusion based upon the mathematical relationships.
For acyclic and aromatic hydrocarbons

$$\Delta_{fus}\bar{S} = \sum_i n_i C_i G_i + \sum_j n_j C_j G_j + \sum_k n_k C_k G_k \quad (1)$$

For cyclic hydrocarbons

$$\Delta_{fus}\bar{S} = [8.41 + 1.025(n-3)] + \sum_i n_i C_i G_i + \sum_j n_j C_j G_j + \sum_k n_k C_k G_k \quad (2)$$

For polycyclic molecules

$$\Delta_{fus}\bar{S} = [8.41N + 1.025(R-3N)] + \sum_i n_i C_i G_i + \sum_j n_j C_j G_j + \sum_k n_k C_k G_k \quad (3)$$

$$K = \sum_k n_k \quad (4)$$

where n refers to the number of equivalent methylene groups necessary to simulate the size of the ring, R is the total number of ring atoms and N indicates the total number of rings in polycyclic molecules. Hydrocarbon components are identified by the subscript i, the j subscript identifies the carbon(s) bearing the functional group(s) and the k subscript denotes the different functional groups in the molecule.

Each group contribution to the entropy of fusion represents the product of the number of identical groups in the molecule (n) times the group value (G) times an empirical coefficient (C) which modifies each group contribution according to the structural environment around the carbon atom bearing substituent functional groups. Inherent in the group addivity estimational scheme is the underlying assumption that the contribution of any particular atom or group of atoms to entropy change associated in going from a rigid anisotropic solid state to the highly isotropic liquid state is fundamentally constant. Entropies for mesomorphic liquid crystalline transitions are thus included in the overall melting process, if possible. The corresponding enthalpies of fusion are obtained by multiplying $\Delta_{fus}\bar{S}$ by the observed melting point temperature.

Although Chickos and coworkers [20, 21] did present a very convincing set of comparisons between predicted and experimental enthalpy of fusion data for select organic molecules to document the applicability of their

TABLE 1

Molar enthalpies of fusion (J mol^{-1}) and melting point temperature (°C)

Formula	Chemical name	T_{mp}	$\Delta_{fus}\bar{H}$	Ref.
$C_4H_4N_2$	Pyrazine	55.0	12,950	22
$C_5F_{13}N$	Perfluoromethyldiethylamine	−123.5	7,160	23
C_6Cl_6	Hexachlorobenzene	228.9	25,180	24
C_6HCl_5	Pentachlorobenzene	83.8	20,100	24
$C_6H_2Cl_4$	1,2,3,5-Tetrachlorobenzene	50.6	18,320	24
$C_6H_2Cl_4$	1,2,4,5-Tetrachlorobenzene	139.4	29,940	24
$C_6H_2Cl_4$	1,2,3,4-Tetrachlorobenzene	46.5	16,960	24
$C_6H_3N_3O_6$	1,3,5-Trinitrobenzene	121.0	15,900	25
$C_6H_4ClNO_2$	4-Nitro-1-chlorobenzene	81.5	11,850	26
$C_6H_4N_2O_4$	1,2-Dinitrobenzene	114.5	22,180	25
$C_6H_4N_2O_4$	1,3-Dinitrobenzene	86.9	17,570	25
$C_6H_4N_2O_4$	1,4-Dinitrobenzene	171.0	26,360	25
$C_6H_6O_2$	1,2-Dihydroxybenzene	104.7	15,000	27
$C_6H_6O_2$	1,3-Dihydroxybenzene	110.4	15,250	27
$C_6H_6O_2$	1,4-Dihydroxybenzene	172.8	21,090	27
$C_7H_5N_3O_6$	2,4,6-Trinitrotoluene	79.0	23,430	25
$C_7H_6N_2O_4$	2,6-Dinitrotoluene	54.3	23,850	25
$C_7H_6N_2O_4$	2,3-Dinitrotoluene	56.6	17,570	25
$C_7H_6N_2O_4$	2,4-Dinitrotoluene	67.6	22,180	25
$C_7H_6N_2O_4$	3,4-Dinitrotoluene	56.3	18,830	25
$C_7H_7NO_2$	*p*-Nitrotoluene	48.7	18,410	25
$C_7H_{10}O_2$	2-Norbornanone	95.5	3,390	21
$C_8H_8O_2$	Methyl benzoate	−12.5	13,900	28
$C_9H_{10}O$	Cinnamyl alcohol	34.9	15,730	21
$C_9H_{10}O_2$	4-Ethylbenzoic acid	111.5	14,060	21
$C_9H_{20}O_2S$	3-(*n*-Hexylthio)-1,2-propanediol	15.8	48,500	29
$C_{10}H_7Cl$	2-Chloronaphthalene	58.0	14,000	30
$C_{10}H_8$	Naphthalene	80.7	19,000	31
$C_{10}H_{10}N_4O_2S$	4-Amino-*N*-2-pyrimidinylbenzenesulfonamide	265.6	31,230	32
$C_{10}H_{10}O_4$	1,2-Dicarbomethoxybenzene	0.0	15,700	28
$C_{10}H_{10}O_4$	1,3-Dicarbomethoxybenzene	67.5	25,300	28
$C_{10}H_{10}O_4$	1,4-Dicarbomethoxybenzene	142.0	32,900	28
$C_{10}H_{12}N_2O_3S$	4-Amino-*N*-(5-methyl-3-isoxazolyl)-benzenesulfonamide	166.3	28,670	32
$C_{10}H_{12}O_2$	1-(2-Hydroxy-2,6-dimethylphenyl)ethanone	60.0	1,360	33
$C_{10}H_{14}O$	Thymol	51.0	22,180	21
$C_{10}H_{22}O_2S$	3-(*n*-Heptylthio)-1,2-propanediol	15.8	27,300	29
$C_{10}H_{22}O_3$	3-(*n*-Heptyloxy)-1,2-propanediol	14.8	28,800	29
$C_{10}H_{23}NO_2$	3-(*n*-Heptylamino)-1,2-propanediol	51.7	28,800	29
$C_{11}H_{12}N_3O_3S$	4-Amino-*N*-(3,4-dimethyl-5-isoxazolyl)-benzenesulfonamide	195.0	29,250	32
$C_{11}H_{12}N_4O_2S$	4-Amino-*N*-(4-methyl-2-pyrimidinyl)-benzenesulfonamide	242.0	31,550	32
$C_{11}H_{12}N_4O_3S$	4-Amino-*N*-(5-methoxy-2-pyrimidinyl)-benzenesulfonamide	213.5	34,540	32
$C_{11}H_{14}O_2$	4-*tert*-butylbenzoic acid	163.5	17,910	21

TABLE 1 (continued)

Formula	Chemical name	T_{mp}	$\Delta_{fus}\bar{H}$	Ref.
$C_{11}H_{24}O_2S$	3-(*n*-Octylthio)-1,2-propanediol	31.9	39,800	29
$C_{11}H_{24}O_3$	3-(*n*-Octyloxy)-1,2-propanediol	22.9	33,400	29
$C_{11}H_{25}NO_2$	3-(*n*-Octylamino)-1,2-propanediol	62.7	45,100	29
$C_{12}H_8N_2O_5$	4,4′-Dinitrodiphenyl ether	145.0	10,290	26
$C_{12}H_9N$	Carbazole	239.0	27,200	31
$C_{12}H_{12}N_2O$	4,4′-Diaminodiphenyl ether	192.3	7,740	26
$C_{12}H_{12}O_6$	1,2,3-Tricarbomethoxybenzene	102.0	32,700	28
$C_{12}H_{14}N_4O_2S$	4-Amino-*N*-(2,6-dimethyl-4-pyrimidinyl)-benzenesulfonamide	250.4	42,650	32
$C_{12}H_{14}N_4O_2S$	4-Amino-*N*-(4,6-dimethyl-4-pyrimidinyl)-benzenesulfonamide	198.5	31,120	32
$C_{12}H_{26}O$	Dodecanol	24.8	40,170	21
$C_{12}H_{26}O_2S$	3-(*n*-Nonylthio)-1,2-propanediol	29.6	23,000	29
$C_{12}H_{26}O_3$	3-(*n*-Nonyloxy)-1,2-propanediol	24.3	29,500	29
$C_{12}H_{27}NO_2$	3-(*n*-Nonylamino)-1,2-propanediol	69.2	53,200	29
$C_{13}H_{10}O_2$	(2-Hydroxyphenyl)phenylmethanone	35.0	665	33
$C_{13}H_{14}N_2$	bis-(4-Aminophenyl)methane	90.6	9,220	26
$C_{13}H_{15}N_5$	4-Amino-4-(*N*-methylanilino)-6-isopropenyl-1,3,5-triazine	118.0	31,400	34
$C_{13}H_{28}O_2S$	3-(*n*-Decylthio)-1,2-propanediol	38.7	17,300	29
$C_{13}H_{28}O_3$	3-(*n*-Decyloxy)-1,2-propanediol	37.8	38,900	29
$C_{13}H_{29}NO_2$	3-(*n*-Decylamino)-1,2-propanediol	73.4	54,800	29
$C_{14}H_{10}$	Anthracene	217.5	29,000	31
$C_{14}H_{12}$	9,10-Dihyrdophenanthrene	33.4	12,790	35
$C_{14}H_{12}O_4$	1,2-Dicarbomethoxynaphthalene	85.1	27,600	28
$C_{14}H_{12}O_4$	1,3-Dicarbomethoxynaphthalene	104.5	30,500	28
$C_{14}H_{12}O_4$	1,4-Dicarbomethoxynaphthalene	66.0	20,400	28
$C_{14}H_{12}O_4$	1,5-Dicarbomethoxynaphthalene	119.5	26,400	28
$C_{14}H_{12}O_4$	1,6-Dicarbomethoxynaphthalene	98.6	22,100	28
$C_{14}H_{12}O_4$	1,7-Dicarbomethoxynaphthalene	89.0	20,000	28
$C_{14}H_{12}O_4$	1,8-Dicarbomethoxynaphthalene	102.5	27,800	28
$C_{14}H_{12}O_4$	2,3-Dicarbomethoxynaphthalene	50.0	20,200	28
$C_{14}H_{12}O_4$	2,6-Dicarbomethoxynaphthalene	191.2	38,400	28
$C_{14}H_{12}O_4$	2,7-Dicarbomethoxynaphthalene	137.2	26,600	28
$C_{14}H_{14}O_8$	1,2,3,4-Tetracarbomethoxybenzene	131.5	40,400	28
$C_{14}H_{14}O_8$	1,2,3,5-Tetracarbomethoxybenzene	116.0	32,600	28
$C_{14}H_{14}O_8$	1,2,4,5-Tetracarbomethoxybenzene	144.8	35,700	28
$C_{14}H_{17}N_5$	2-Amino-4-(*N*-ethylanilino)-6-isopropenyl-1,3,5-triazine	107.0	27,600	34
$C_{14}H_{30}O_2S$	3-(*n*-Undecylthio)-1,2-propanediol	44.2	18,300	29
$C_{14}H_{30}O_3$	3-(*n*-Undecyloxy)-1,2-propanediol	38.5	43,100	29
$C_{14}H_{31}NO_2$	3-(*n*-Undecylamino)-1,2-propanediol	75.6	58,200	29
$C_{15}H_{14}O$	1,3-Diphenylacetone	34.4	20,210	21
$C_{15}H_{14}O_2$	(2-Hydroxy-4,6-dimethylphenyl)-phenylmethanone	132.0	674	33
$C_{15}H_{19}N_5$	2-Amino-4-(*N*-propylanilino)-6-isopropenyl-1,3,5-triazine	107.0	31,500	36
$C_{15}H_{30}O_2$	Methyl myristate	19.0	50,210	21

TABLE 1 (continued)

Formula	Chemical name	T_{mp}	$\Delta_{fus}\bar{H}$	Ref.
$C_{15}H_{32}O_2S$	3-(*n*-Dodecylthio)-1,2-propanediol	52.3	20,300	29
$C_{15}H_{32}O_3$	3-(*n*-Dodecyloxy)-1,2-propanediol	49.8	51,400	29
$C_{15}H_{33}NO_2$	3-(*n*-Dodecylamino)-1,2-propanediol	78.7	62,100	29
$C_{16}H_{14}O_6$	4,4′-Dicarboxy-α,ω-diphenoxyethane	354.0	56,610	37
$C_{16}H_{14}O_6$	1,2,3-Tricarbomethoxynaphthalene	89.8	23,700	28
$C_{16}H_{14}O_6$	1,2,4-Tricarbomethoxynaphthalene	120.2	32,100	28
$C_{16}H_{14}O_6$	1,2,5-Tricarbomethoxynaphthalene	89.8	25,500	28
$C_{16}H_{14}O_6$	1,2,6-Tricarbomethoxynaphthalene	144.0	35,900	28
$C_{16}H_{14}O_6$	1,2,7-Tricarbomethoxynaphthalene	155.0	36,100	28
$C_{16}H_{14}O_6$	1,2,8-Tricarbomethoxynaphthalene	92.8	24,800	28
$C_{16}H_{14}O_6$	1,3,5-Tricarbomethoxynaphthalene	129.8	25,900	28
$C_{16}H_{14}O_6$	1,3,6-Tricarbomethoxynaphthalene	196.5	37,400	28
$C_{16}H_{14}O_6$	1,3,7-Tricarbomethoxynaphthalene	172.5	37,200	28
$C_{16}H_{14}O_6$	1,3,8-Tricarbomethoxynaphthalene	114.0	27,700	28
$C_{16}H_{14}O_6$	1,4,5-Tricarbomethoxynaphthalene	129.8	26,500	28
$C_{16}H_{14}O_6$	1,4,6-Tricarbomethoxynaphthalene	137.0	30,200	28
$C_{16}H_{14}O_6$	2,3,5-Tricarbomethoxynaphthalene	128.8	41,000	28
$C_{16}H_{14}O_6$	2,3,6-Tricarbomethoxynaphthalene	125.5	34,400	28
$C_{16}H_{15}N$	4′-Propylbiphenyl-4-carbonitrile	65.6	22,700	38
$C_{16}H_{16}O_{10}$	Pentacarbomethoxybenzene	151.5	38,000	28
$C_{16}H_{21}N_5$	2-Amino-4-(*N*-butylanilino)-6-isopropenyl-1,3,5-triazine	78.0	31,800	34
$C_{16}H_{27}N$	*trans,trans*-4′-Propylbicyclohexyl-4-carbonitrile	80.6	1,800	38
$C_{16}H_{33}NO$	*N*-Butyl-dodecanamide	48.9	39,000	39
$C_{16}H_{34}O_2S$	3-(*n*-Tridecylthio)-1,2-propanediol	57.4	22,700	29
$C_{16}H_{34}O_3$	3-(*n*-Tridecyloxy)-1,2-propanediol	51.0	51,400	29
$C_{16}H_{35}NO_2$	3-(*n*-Tridecylamino)-1,2-propanediol	81.7	68,700	29
$C_{17}H_{16}O_6$	4,4′-Dicarboxy-α,ω-diphenoxypropane	323.5	51,920	37
$C_{17}H_{34}O_2$	Methyl palmitate	31.5	68,200	21
$C_{17}H_{36}O_2S$	3-(*n*-Tetradecylthio)-1,2-propanediol	63.2	26,800	29
$C_{17}H_{36}O_3$	3-(*n*-Tetradecyloxy)-1,2-propanediol	58.1	62,100	29
$C_{17}H_{37}NO_2$	3-(*n*-Tetradecylamino)-1,2-propanediol	83.0	64,900	29
$C_{18}H_{15}N$	Triphenylamine	126.7	24,890	21
$C_{18}H_{16}O_8$	1,2,3,4-Tetracarbomethoxynaphthalene	151.5	35,900	28
$C_{18}H_{16}O_8$	1,2,4,5-Tetracarbomethoxynaphthalene	166.2	36,400	28
$C_{18}H_{16}O_6$	1,2,5,6-Tetracarbomethoxynaphthalene	198.2	42,100	28
$C_{18}H_{16}O_8$	1,2,6,7-Tetracarbomethoxynaphthalene	134.2	34,200	28
$C_{18}H_{16}O_8$	1,4,5,8-Tetracarbomethoxynaphthalene	204.5	36,100	28
$C_{18}H_{16}O_8$	2,3,6,7-Tetracarbomethoxynaphthalene	186.2	42,200	28
$C_{18}H_{18}O_6$	4,4′-Dicarboxy-α,ω-diphenoxybutane	339.5	65,480	37
$C_{18}H_{18}O_{12}$	Hexacarbomethoxybenzene	190.5	22,500	28
$C_{18}H_{25}N_5$	2-Amino-4-(*N*-hexylanilino)-6-isopropenyl-1,3,5-triazine	68.0	24,100	36
$C_{18}H_{30}O$	2,4,6-Tri-*tert*-butylphenol	129.6	19,460	21
$C_{18}H_{37}NO$	*N*-Butyl-tetradecanamide	62.9	45,000	39
$C_{18}H_{38}NO$	Octadecanamide	105.9	59,830	21
$C_{18}H_{38}O$	1-Octadecanol	57.5	70,080	21

TABLE 1 (continued)

Formula	Chemical name	T_{mp}	$\Delta_{fus}\bar{H}$	Ref.
$C_{19}H_{20}O_6$	4,4′-Dicarboxy-α,ω-diphenoxypentane	288.5	61,090	37
$C_{20}H_{12}$	Perylene	277.8	31,870	40
$C_{20}H_{29}N_5$	2-Amino-4-(*N*-octylanilino)-6-isopropenyl-1,3,5-triazine	61.0	24,600	36
$C_{21}H_{14}N_2O_4$	bis-(4-Maleic acidimide-phenyl)methane	157.8	18,230	26
$C_{22}H_{18}O_6$	bis-(4-Methoxyphenyl)isophthalate	156.3	42,200	41
$C_{22}H_{33}N_5$	2-Amino-4-(*N*-decylanilino)-6-isopropenyl-1,3,5-triazine	67.0	32,200	36
$C_{22}H_{45}NO$	*N*-Hexyl-hexadecanamide	69.9	57,000	39
$C_{22}H_{46}$	*n*-Docosane	42.0	39,760	42
$C_{24}H_{37}N_5$	2-Amino-4-(*N*-dodecylanilino)-6-isopropenyl-1,3,5-triazine	66.0	41,000	36
$C_{24}H_{50}$	*n*-Tetracosane	50.5	57,310	42
$C_{26}H_{34}O_6$	4,4′-Dicarboxy-α,ω-diphenoxydodecane	262.0	46,740	37
$C_{26}H_{41}N_5$	2-Amino-4-(*N*-tetradecylanilino)-6-isopropenyl-1,3,5-triazine	66.0	45,800	36
$C_{26}H_{54}$	*n*-Hexacosane	56.0	63,920	42
$C_{28}H_{45}N_5$	2-Amino-4-(*N*-hexadecylanilino)-6-isopropenyl-1,3,5-triazine	72.0	51,500	36
$C_{28}H_{58}$	*n*-Octacosane	60.8	66,520	42
$C_{30}H_{49}N_5$	2-Amino-4-(*N*-octadecylanilino)-6-isopropenyl-1,3,5-triazine	72.0	60,710	36
$C_{34}H_{42}O_6$	bis-(4-Heptyloxyphenyl)isophthalate	116.9	71,900	41
$C_{38}H_{48}N_2O_2S_4$	2,6-Di(4-octyloxybenzylthio)benzo-[1,2-*d*:5,4-*d*′]-bisthiazole	150.0	43,900	43
$C_{46}H_{64}N_2O_2S_4$	2,6-Di(4-dodecyloxybenzylthio)benzo-[1,2-*d*:5,4-*d*′]-bisthiazole	145.0	57,400	43
$C_{54}H_{80}N_2O_2S_4$	2,6-Di(4-hexyldecyloxybenzylthio)benzo-[1,2-*d*:5,4-*d*′]-bisthiazole	139.0	65,300	43

approach, one must recognize that only a very limited number of functional groups and "parent" molecules were represented. Continued development of group contribution methods does require the ready availability of a large $\Delta_{fus}\bar{H}$ data base. To provide the scientific community with a convenient reference source for $\Delta_{fus}\bar{H}$ data, the present author has previously compiled experimental values for approx. 600 organic compounds retrieved from the chemical literature. In this follow-up communication, enthalpy data for an additional 149 compounds are listed in Table 1. Compounds are arranged according to molecular formula, beginning with increasing carbon and hydrogen number. Care was taken to eliminate typographical errors, but readers should be aware that in a compilation of this magnitude a few errors will undoubtedly occur. Literature references are given in the last column to indicate actual sources consulted. It is hoped that this tabulation will not only provide a data base for developing better estimational schemes for

fusion enthalpies, but will facilitate thermodynamic modelling of solid–liquid equilibria, where $\Delta_{fus}\bar{H}$ values are required as input parameters for derived predictive equations.

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