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C-K (Carbon-Potassium) System

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Equilibrium Diagram

There is no phase diagram available for this system. It may be conjectured that it would be similar to that for the C-Li system, and so would show (a) the compound C_2K_2, (b) a liquid, and (c) a eutectic of composition very close to 100 at.% K and temperature very close to the M.P. of K.

The alkali metals exhibit three types of intermediate compounds with carbon. For potassium, these may be designated as:

(1) simple C-K compounds (C_2K_2 in this case)
(2) potassium-graphite intercalation compounds
(3) potassium-fullerene exohedral and endohedral compounds

In the chemical literature, molecular formulas for C-K have been written with either C or K placed first, without much consistency. In the present instance, C will always come before K.

C_2K_2 (potassium acetylide, potassium carbide) is prepared in two steps by the reaction of liquid potassium with acetylene [1866Ber, 1898Moi1, 1958Fop] and was characterized by XRD [1958Fop, 1999Hem] and neutron diffraction [1999Hem]. The reaction may also be carried out in liquid ammonia [1898Moi2]. The intermediate product in both cases is C_2HK, which upon heating in vacuo above 100 °C is converted to C_2K_2 [1898Moi1, 1898Moi2, 1999Hem]. The room-temperature form of C_2K_2 (α-C_2K_2) transforms at 147 °C to a different crystal structure (β-C_2K_2 [1999Hem]. Its melting behavior has not been reported, but like Na_2C_2 it probably decomposes before melting.

Potassium-Graphite Intercalation Compounds

These compounds are also called "lamellar," "layered," "interstitial" or "insertion" compounds. All alkali metals form graphite intercalation compounds, but those with Li or Na are fewer and are formed less easily than those of the heavier alkali metals. The structure of graphite and metal intercalation compounds is shown schematically in Fig. 1. Graphite has a layered structure, in which the carbon atoms are arranged in parallel planes, in each of which they form a network of hexagons [1971Nov]. There is no chemical bonding between the layers and so alkali metals can be inserted rather easily between the layers. The separation of the adjacent layers increases somewhat when alkali metal atoms are inserted.

The conventional method of indicating the stoichiometry of metal-graphite intercalation compounds is C_xM, where x may be an integer or fractional number. The formula C_xM does not represent a molecule of $(x + 1)$ atoms; rather, it indicates merely the atom ratio C/M in a macroscopic sample.

The intercalation compounds (Fig. 1) are named "first stage," "second stage," etc. according to the number of graphite layers separating the metal layers. The stoichiometry C_xM assigned to a particular stage compound should be regarded as nominal or average. The overall stoichiometry is established by chemical reaction and analysis, the particular stage by XRD.

The preparation and properties of alkali metal intercalation compounds have been reviewed extensively [1959Hen, 1959Rud, 1960Cro, 1971Nov, 1975Ber, 1975Boe, 1976Ebe, 1977Her, 1980Sel, 1981Dre, 1981Her, 1982Bar, 1990 Lag].

The common methods for the preparation of potassium-graphite intercalation compounds are

- reaction with potassium vapor or liquid at low pressures;
- in organic solvent solution;
- reaction with liquid potassium at high pressure.

The preparation of compounds at high pressure is described later in the "Pressure" section of this paper. Both natural and synthetic graphite have been used in these methods.

In the reaction of graphite with potassium vapor (the "two-bulb method" [1951Her1, 1951Her2, 1955Her]), graphite in one part of a vacuum apparatus is maintained at a given temperature and is then exposed to potassium vapor from a reservoir of molten metal maintained at a lower temperature. This is a preferred method for preparing these compounds of definite stoichiometry. The stoichiometry of the compound is controlled by the temperature difference between the graphite and metal reservoir. The stoichiometry is ultimately established by chemical reaction.

The stage I compound is C_8K and the successive stages may be represented by $C_{12n}K$, where n is the stage number $(n \geq 2)$. Compounds of stages I-XIV have been prepared by the two-bulb method [1955Her, 1968Car, 1969Nix, 1980Bil, 1983Maa, 1983Mis, 1985Hei, 1986Nis]. The compounds are commonly characterized by XRD. C_8K was also prepared by the reaction of graphite in direct contact with molten potassium [1958Pod, 1968Aro], as were the higher-stage compounds $C_{24}K$, $C_{36}K$, $C_{48}K$ and $C_{60}K$ [1966Ott].

Higher-stage intercalation compounds were prepared by removing potassium from C_8K, either by evaporation at elevated temperatures [1954Rud1, 1954Rud2] or by reaction with CO [1971Dau], up to stage IX. They may also be prepared by reaction of C_8K with further graphite.

Potassium also reacts with other forms of carbon such as cokes, soot, carbon black, activated carbon, etc. [1926Fre,

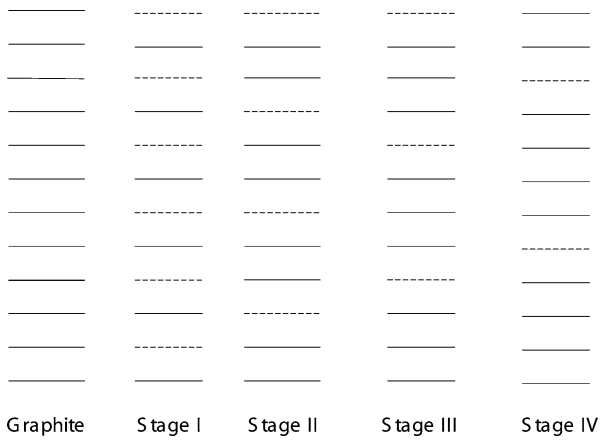

Fig. 1 Schematic representation of the structure of graphite and potassium-graphite intercalation compounds. *Solid lines*: graphite layers. *Dashed lines*: metal layers. The patterns repeat themselves in both directions

1929Fre, 1957Pla, 1970Ber, 1975Ber]. In these cases, well-defined compounds do not usually result because potassium may react with impurities or simply be adsorbed, as shown by Brunauer-Emmett-Teller (B.E.T.) isotherms [1975Ber]. The reaction with "glassy" amorphous carbon produces a highly unstable material which shatters violently [1968Hal, 1969Hal]. Well-defined compounds may, however, be prepared from synthetic graphite, e.g., "graphitized" carbon from the pyrolysis of *p*-phenylenevinylene polymer [1987Uen].

The preparation of a potassium intercalation compound of stoichiometry between C_8K and $C_{24}K$ was reported by [1932Sch, 1967Sal, 1968Aro, 1968Car, 1977Ehl]. The stoichiometry was variously reported as $C_{8\text{-}12}K$, $C_{10}K$ and $C_{16}K$. The assignments were based on EMF [1968Aro], thermogravimetry [1967Sal, 1968Car, 1972Car, 1977Ehl] and XRD [1932Sch, 1972Car]. This phase was called a stage II compound [1932Sch], but [1972Car] considered it to be a stage I compound in which the intercalated potassium layers are disordered. From the results of the EMF and thermogravimetric measurements, it may be concluded that this intermediate phase is stable only at elevated temperatures; the lower limit was given as 320 °C [1968Aro] or 370 °C [1972Car]. The more complete data of [1985Min] show that there are two disordered stage I compounds, which are stable only above 307 °C and 332 °C respectively.

Low-stage intercalation compounds may be prepared in solution at ambient temperature. The solvents were tetrahydrofuran (THF) [1965Ste1, 1965Ste2, 1966Ste] or methylTHF [1996Miz, 1997Miz]. The reaction carried out in pentane solution was effected with a cobalt-ethylene-trimethylphosphane complex [1980Kle].

Potassium-Fullerene Compounds

The fullerenes are closed carbon-cage molecules containing only pentagonal and hexagonal rings. They have the general formula C_n, where n is an even integer greater than 20 [1995Fow]. The best-known fullerenes are C_{60} ("soccer ball" shape) and C_{70}.

C_{60} may be prepared in an ac arc between graphite electrodes in subatmospheric Ar or He. The C_{60} is extracted from the smoke of carbon particles by an organic solvent [1990Hau, 1990Kra, 1990Tay, 1991Bet]. It may also be prepared by a vaporizing laser beam falling on a graphite surface under a high-pressure stream of He [1985Kro] or in a static low pressure Ar atmosphere [1990Mei].

All the alkali metals form *exohedral* and *endohedral* compounds with fullerene. In exohedral potassium compounds, the K atoms and fullerene molecules occupy sites on a crystalline lattice; in endohedral compounds, the K atoms are inside the fullerene cages [1998Lah]. The endohedral compound of potassium is conventionally indicated by $K@C_{60}$.

$C_{60}K$ is prepared by direct reaction. This may be done by depositing K on a fullerene substrate in vacuo [1992Win]. Crystals are prepared by direct reaction with K vapor or liquid at 500 K or higher [1994Teg, 1995Zhu, 1998Win, 2003Sko]. It was characterized by XRD [1993Zhu]. $C_{60}K$ is stable only above 150 °C; at room temperature the final equilibrium products are C_{60} and $C_{60}K_3$ [1995Zhu, 1996Rob]. From measurements of the dipole moment, it was concluded that $C_{60}K$ is almost entirely ionic in nature [2000Ant].

$C_{60}K_2$ was prepared as thin films in vacuo, but was thought to be a transient species [1992Win]. The principal method of preparing $C_{60}K_3$ is by direct reaction in vacuo or inert gas. K vapor may be used [1991Heb, 1991Spa, 1992Xia,] or K liquid [1991Hol, 1991Ste] at elevated temperatures. Thin films are made by vapor deposition on a fullerene substrate [1991Had].

Thin films or crystals may be prepared from C_{60}-K compounds of different stoichiometry, either by dilution with C_{60}or by distillation of excess K [1991Mcc, 1994Dou, 1995Knu]. Identification and characterization are routinely made by XRD or NMR [1991Ste, 1992Mur].

$C_{60}K_3$ may be prepared in solution. An electrochemical method used acetonitrile as solvent [1993Wil]; the direct reaction was carried out in toluene or tetralin, with or without previous dry ball milling [1998Bul, 2000Dyt]. Liquid ammonia was used by [1993Buf].

$C_{60}K_4$ was prepared as thin films but was thought to be a transient species [1992Win]. It is in fact stable at room temperature, and was prepared by dilution of $C_{60}K_6$ [1992Mur, 1995Knu] or by distillation of excess K from compounds more deficient in metal [1995Knu]. XRD, electron diffraction and NMR were used in these studies for characterization.

$C_{60}K_6$ is sometimes described as the "saturated" potassium-fullerene compound. It is best prepared by direct reaction in vacuo at 250-350 °C [1991Zho, 1992Mur]. It was characterized by XRD in these reports.

The endohedral compound $K@C_{60}$ was prepared as thin films by bombardment of fullerene layers by potassium ions [1996Cam, 1996Tel]. Ab initio calculations [1995Tom] suggest that this compound is strongly ionic in nature.

Although no global C-K phase diagram is available, C_{60}-K phase diagrams have been proposed.

The system $C_{60}K_x$ (x = 0-4) was studied by X-ray photoelectron spectroscopy and a phase diagram was proposed [1993Poi, 1993Wea, 1994Poi]. The earlier T-x diagram shows phase boundaries among the phases of x = 0, 1, 3, and 4, with a eutectoid at 150 °C for the equilibrium $C_{60}K = C_{60} + C_{60}K_3$. The pressure is not explicitly stated, but evidently is the equilibrium vapor pressure. The same authors present a fuller phase diagram [1995Poi] incorporating compounds with x = 0, 1, 3, 4, and 6. The pressure is 1 atmosphere and the temperature range is extended to the vapor phase (>1500°C).

A similar investigation used XRD and Knudsen cell mass spectrometry [2001Sko]. The composition range covered was $C_{60}K_x$ (x = 0-6), and activities of both components were deduced from vapor pressures. The temperature range illustrated is 298-925 K at 1 atmosphere pressure. A P-x diagram at 925 K was also given.

There is qualitative agreement between the two proposed T-x phase diagrams, that of [1995Poi] being the more detailed but more conjectural.

Crystal Structures and Lattice Parameters

These are presented in Tables 1 and 2 respectively.

The structure of α-C_2K_2 was determined from powder XRD [1958Fop, 1999Hem] and neutron diffraction [1999Hem]. The C_2^{2-} ions were described [1958Fop] as distorted ellipsoids in an overall tetragonal structure similar to that of room-temperature C_2Na_2; alternatively, as a distorted variant of the anti-fluorite structure [1999Hem]. The high-temperature form (β-C_2K_2) is fcc (undistorted antifluorite structure with disordered C_2^{2-} dumbbells [1999Hem]).

The temperature dependence of the lattice parameters of α-C_2K_2 was measured in the interval 368-402 K and of β-C_2K_2 at 407-465 K by XRD [2004Zib] (data not tabulated, points on diagram only).

The structure of potassium-graphite intercalation compounds is derived from the structure of graphite itself (Fig. 1). The data have been reviewed extensively {1959Hen, 1959Rud, 1971Nov, 1975Ber, 1975Boe, 1976Ebe, 1979Fis, 1979Her, 1980Sel, 1981Dre, 1982Bar]. The order of the carbon atoms in the graphite layers is very little affected by the process of intercalation [1969Nix, 1976Gue].

The first stage compound C_8K is the only one in which the K atoms are completely ordered at room temperature. The K atoms are ordered in a centered hexagonal arrangement [1954Rud1]. The resulting three-dimensional structure may be described as based on a hexagonal unit cell (hP36) with a = 0.495 nm, c = 2.1388 nm [1932Sch, 1954Rud1, Pearson3] showing direct similarity to graphite itself. An alternative more accurate description is an orthorhombic unit cell [1960Wol, 1970Swa, 1977Par, 1978Lag1, 1978Lag2, 1981Her]. The space group of this orthorhombic arrangement was variously identified as C222 [1960Wol] or F_{ddd} [1978Lag1, 1978Lag2], Fdd2 [1977Par] or F222 [1970Swa]. The designation Fdd2 is preferred [1982Tre]. The unit cell was described by [1978Lag1, 1978Lag2] as "orthohexagonal" ($b = a\sqrt{3}$), since a set of three orthorhombic unit cells, twisted 120° with respect to one another, gives an apparent hexagonal symmetry [1978Lag1, 1978Lag2, 1981Her].

There is a great deal of information concerning the structure of Stage II and higher compounds. Most, if not all, of this information is focused on interlayer spacings and order in the potassium layers. For $C_{24}K$ (stage II), reference may be made to [1968Nix, 1971Dau, 1974Bil, 1979Her, 1982Boe, 1985Rou, 1986Nis]. Similarly for stages III-XI, [1969Nix,1980Bil,1985Hei] are pertinent.

The molecular structure of fullerene C_{60} was suggested by [1985Kro, 1987Kro, 1990Kra] to resemble closely that of a soccerball. Ab initio calculations [1988Sch] indicated that a cluster of C atoms having 60 vertices and 32 faces (20 hexagonal and 12 pentagonal) would be energetically favored. This structure, a truncated icosahedron, would

Table 1 C-K crystal structure data

Phase	Composition at.% K	Pearson symbol	Space group	Strukturbericht designation	Prototype	Reference
C	0	*hP*4	$P6_3/mmc$	A9	C(graphite)	[King1]
C	0	*cF*8	$Fd\bar{3}m$	A4	C(diamond)	[King1]
C	0	*cF*240	$Fm\bar{3}m$		C_{60}	[1991Hei]
$C_{60}K$(a)	1.6	*cF**				[1993Zhu]
$C_{60}K_3$	4.8	*cF*252	$Fm\bar{3}m$			[1991Ste]
$C_{60}K_4$	6.3	*tI**				[1991Fle1]
$C_{60}K_6$	9.1	*cI**	*Im*3			[1991Zho]
C_8K	11.1	*oF*72	*Fdd*2			[1970Swa, 1982Tre]
C_2K_2	50	*tI*32	$I4_1/acd$			[1958Fop]
C_2K_2(b)	50	*cF*16	$Fm\bar{3}m$			[1999Hem]
K	100	*cI*2	$Im\bar{3}m$	A2	W	[King1]

(a) Above 150 °C
(b) Above 147 °C

Table 2 C-K lattice parameter data

Phase	Composition at.% K	Lattice parameter, nm			Reference
		a	*b*	*c*	
C (dia)	0	0.54308	…	…	[Pearson3]
C(graphite)	0	0.2464	…	0.6711	[Pearson3]
C(C_{60})	0	1.411	…	…	[1991Ste]
		1.4161	…	…	[1992Tan]
		1.417	…	…	[1991Hei]
		1.4155	…	…	[1992Aga]
$C_{60}K$(a)	0.02	1.407	…	…	[1993Zhu]
		1.4138	…	…	[1997Bor]
$C_{60}K_3$	4.8	1.4240	…	…	[1991Ste, 1992Tan]
		1.4253	…	…	[1991Fle2]
		1.4237	…	…	[1997Bor]
$C_{60}K_4$	6.3	1.1886	…	1.0774	[1991Fle1]
$C_{60}K_6$	9.1	1.139	…	…	[1991Zho]
		1.140	…	…	[1998Bor]
C_8K	11.1	0.496	0.860	2.134	[1970Swa]
		0.4961	0.8592	2.376	[1977Par]
		0.4965	0.8599	2.1584	[1978Lag1, 1978Lag2]
		0.492	0.852	2.376	[1981Her]
		0.496	0.859	2.14	[1982Tre]
C_2K_2	50	0.758	…	1.469	[1958Fop]
		0.7594	…	1.4521	[1999Hem]
C_2K_2(b)	50	0.7553	…	…	[1999Hem]
K	100	0.5321	…	…	[King1]

(a) At 200 °C
(b) At 167 °C

have I_h overall symmetry, i.e., all the C atoms are equivalent. This assignment was confirmed by NMR measurements [1990Joh, 1991Yan] and Raman/IR spectra [1991Bet]. The arrangement of the C_{60} molecules in the crystal lattice was deduced from powder XRD [1990Kra, 1991Hei]. Originally it was thought that the spheres were stacked in a hexagonal close-packed lattice, with a = 1.002 nm and c = 1.636 nm [1990Kra]. Later more careful examination showed that the crystal structure is fcc [1991Hei] at ambient temperature.

At −24 °C the structure becomes simple cubic (cP240) and a = 1.404 nm at 11 K [1991Hei]. According to [1992Jin], this transition probably represents a crystal/plastic crystal transformation. The plausibility of such a first-order transition is supported by ab initio calculations [1991Guo].

The structures of K fullerides were obtained by XRD on powder samples. They are all closely related to the crystal structure of C_{60} itself. Thus $C_{60}K$ (stable only at high temperature) is fcc [1993Zhu]; $C_{60}K_3$ is fcc [1991Fle2, 1991Ste]; $C_{60}K_4$ is bct [1991Fle1, 1992Mur, 1997Kun]; $C_{60}K_6$ is bcc [1991Zho, 1992Mur].

C_{60} fullerene—like many unsaturated organic compounds—polymerizes under certain conditions [1993Rao, 1994Iwa]. $C_{60}K$ behaves similarly. When this compound is cooled from high temperature, a number of metastable phases may appear before room temperature is reached [1995Pus, 1995Ste]. Among them are a dimer phase and a polymer phase. Though these are thermodynamically metastable, they persist long enough at lower temperatures to be characterized. Both are orthorhombic (space group Pmnn). The lattice parameters of the polymer were determined by XRD [1995Osz, 1995Pek, 1998Bor]. Similar values were found by [1994Ste]. For the dimer at this temperature, a = 0.961 nm, b = 0.979 nm, c = 1.419 nm [1995Osz, 1995Pek].

Endohedral compounds have been prepared only as thin films, and no crystalline structure data are available. Since the K atoms are inside the C_{60} cages, it is expected that the crystalline structure of K@C_{60} would be very similar to C_{60} itself.

Thermodynamics

Thermodynamic data for the reaction between graphite and potassium were reviewed by [1959Hen, 1960Set, 1962Set, 1968Tak, 1971Nov, 1972Bal, 1979Her]. The heat of reaction between graphite and the liquid metal was measured by thermal analysis at 64 °C [1926Fre] and by direct calorimetry at 80 °C [1964Sae] and in the range 66-95 °C [1952Qua]. The reaction in question in these measurements is

$$K(l) + 8C(gr) = C_8K\ (cr) \qquad \text{(Eq 1)}$$

In the later work [1952Qua, 1964Sae] different samples of artificial and natural graphite were used, and the reported values for the enthalpy of reaction (1) lie in the range −32.7 to −39.8 kJ mol^{-1}.

Thermodynamic properties of K-graphite intercalation compounds were determined from EMF measurements in the range 200-350 °C [1968Aro]. In later work [1983Nis, 1984Nis, 1985Min] the equilibrium vapor pressure of these compounds was measured as a function of temperature, with simultaneous XRD measurements on the solid. The results were combined in [1985Min] in the form of a P-T phase diagram, which is reproduced in Fig. 2. The derived thermodynamic quantities for the equilibrium reactions among the various stages are presented in Table 3, and were calculated from the equation

$$\ln(P/P^\circ) = \Delta H/RT - \Delta S/R \qquad \text{(Eq 2)}$$

where P is the vapor pressure, P° is the reference pressure (1 atm) and R is the gas constant. The enthalpies and entropies from this equation are presented in Table 3. For the [1985Min] entries in Table 3, the values were derived from the slopes and intercepts read off Fig. 2.

The enthalpy and entropy of formation of potassium intercalated compounds were calculated from the data in Table 3, in the manner described in [1968Aro]. The values were obtained by summing the stages, together with the assumptions that the enthalpy and entropy changes of the reaction

$$60C(gr) + K(g) = C_{60}K(cr) \qquad \text{(Eq 3)}$$

are −140 kJ mol^{-1} and −91.9 J K^{-1} mol^{-1} respectively [1968Aro]. The data were converted to refer to K(l) at 277 °C with the use of the data $\Delta_{vap}H = 87.2$kJ mol^{-1} and $\Delta_{vap}S = 86.1$ J K^{-1} mol^{-1}, derived from recommended thermodynamic data for liquid and gaseous potassium [1985Ohs].

From the results (Table 4), the direct calorimetric determination of the standard enthalpy of formation of the stage I compound [1952Qua, 1964Sae] would correspond to the partially or completely disordered form, rather than the completely ordered C_8K, as might be expected.

Standard enthalpies of formation at 25 °C are listed in [1982Wag], and are −33.5 kJ mol^{-1} for C_8K, decreasing to −44.8 kJ mol^{-1} for $C_{60}K$.

The Gibbs energies of the various metastable modifications of $C_{60}K$ (relative to the high temperature form) were found from differential scanning calorimetry [1995Pek].

The Gibbs energies of formation of the fullerene compounds $C_{60}K_x$ (x = 1, 3, 4, 6) at 925 K were determined from Knudsen effusion mass spectrometry [1998Bor, 2001Sko]. The data are given in Table 4.

Calculated and experimental values of the enthalpy of formation of exohedral potassium fullerides are given in Table 5.

The heat of polymerization of $C_{60}K$, from DSC measurements, was 26 kJ mol^{-1} [1995Pek].

The enthalpy of formation of endohedral K@C_{60}, calculated by the Born-Haber cycle, was reported [1993Wan] as −195 kJ mol^{-1}.

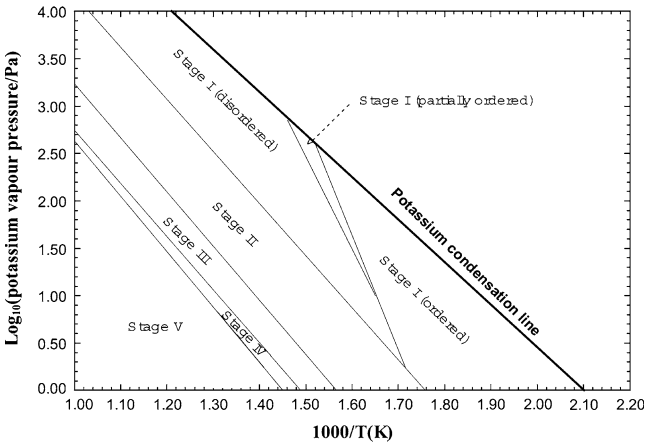

Fig. 2 Pressure of potassium vapor above potassium-graphite intercalation compounds [1985Min]

Pressure

The preparation and properties of potassium-graphite intercalation compound under pressure has been investigated in great detail. A recent review is [1995Gue]. Reaction conditions vary considerably: pressure ≥0.3 GPa, temperature ≥300 K. Stoichiometries C_3K, C_4K, C_6K and C_8K have been reported and characterized.

Table 3 Thermodynamic properties of potassium-graphite intercalation reactions, per g-atom of K (g) [Eq 2]

	[1985Min]		[1968Aro]	
Stage equilibrium reaction	**$-\Delta H$, kJ**	**$-\Delta S$, J K^{-1}**	**$-\Delta H$, kJ**	**$-\Delta S$, J K^{-1}**
I (partially ordered) = I (ordered)	219	285	…	…
I (disordered) = I (partially ordered)	181	222	162	189
II = I (disordered)	105	88.2	103	105
III = II	110	75.6	119	91.1
IV = III	109	65.5	128	91.5
V = IV	114	68.6	131	91.9

Table 4 The Gibbs energies of formation of potassium fullerides at 925 K according to the equation $xK(l) + C_{60}(cr) = C_{60}K_x(cr)$ (from [1998Bor, 2001Sko])

Compound	$-\Delta_f G^\circ$ (cr, 925 K) kJ mol^{-1}
$C_{60}K$	179 ± 12
$C_{60}K_3$	424 ± 21
$C_{60}K_4$	535 ± 27
$C_{60}K_6$	703 ± 37

Table 5 Calculated and experimental enthalpies of formation of exohedral potassium fullerides according to the equation $C_{60}(cr) + xK(cr) = C_{60}K_x(cr)$

		$-\Delta_f H^\circ$, kJ mol^{-1}		
Method	Reference	$C_{60}K$	$C_{60}K_3$	$C_{60}K_6$
Calculation Ab initio	[1992Mar]	135	492	984
Calculation Ab initio	[1991Sai]	…	1910	…
Calculation Born-Haber cycle	[1993Tom]	92	605	1083
Calculation Born-Haber cycle	[1996Cla]	…	466	…
Experiment Differential scanning calorimetry	[1993Che]	…	…	956

The lattice parameter of $C_{60}K_3$ was measured by XRD as a function of pressure up to 29 kbar and the relation may be represented by the equation [1992Zho]

$$d(\ln\ a)/dP = -1.20E - 3\ k\,bar^{-1}$$

The corresponding value for C_{60} itself is approximately –2.3E–3 k bar^{-1} [1992Zho].

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