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STATISTICAL THEORY OF FULLERITE SOLUBILITY

In this paper, calculated the solubility of fullerite in order to clarify the features of the temperature dependence in the process. This takes into account the implementation in different temperature ranges of the crystalline structures of fullerite formed by fullerene molecules of various modifications. Experimental studies of solutions of fullerene molecules in various organic media made it possible to establish the features of the temperature dependence of the solubility of fullerite C_{60}. Instead of the expected increase in solubility with increasing temperature, the extremes of its temperature dependence with a maximum at room temperature were revealed, the endothermicity of the dissolution process was established. The anomalies of the temperature dependence of the solubility of fullerenes C_{60} in organic matrices are due to a change in the structure of bonds in the framework of fullerene molecules, which, in turn, affect the structure of solid-phase fullerite. More accurate results using the formulas obtained can be calculated if the true values of the energy parameters are known from independent experiments. However, it can be assumed that the nature of the revealed patterns will remain.

Key words: materials, molecule, fullerite, Face Centered Cubic (FCC), Body-Centered Cubic (BCC), Simple Cubic (SP).

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Фуллерит ерігіштігінің статистикалық теориясы

Бұл жұмыста осы процеске температураның тәуелділік ерекшелігін анықтау мақсатында фуллереннің ерігіштігі есептелген. Онымен қоса, әр түрлі температура интервалында кристалл құрылымды, әр түрлі модификациядағы фуллерен молекулаларының жасалуы, фуллериттің пайда болуы есепке алынады. Әр түрлі органикалық орталардағы фуллерен молекулаларының ерітіндісінің эксперименталды зерттелуі фуллерит C_{60} ерігіштігінің температуралық тәуелділігін анықтауға мүмкіндік берді. Температура жоғарылаған сайын ерігіштіктің жоғарылауы болжанған болатын, оның орнына бөлме температурасындағы максимум экстремалды температуралық тәуелділік анықталды. Органикалық матрицаларда C_{60} фуллерен ерігіштігінің температураға тәуелді болу аномалиясының себебі – фуллерен молекуласы қаңқасындағы байланыстардың құрылымдарының өзгеруі, олар өз кезегінде қатты фазалы фуллерит құрылымына әсер етеді. Егер энергия параметрлерінің тәуелсіз мәндері тәуелсіз эксперименттерден белгілі болса, алынған формулаларды пайдалана отырып, дәлірек нәтижелерді есептеуге болады.

Түйін сөздер: материалдар, молекула, фуллерит, беткі орталықтандырылған текше, көлемді орталықтандырылған текше, қарапайым текше.

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Статистическая теория растворимости фуллерита

В данной работе рассчитана растворимость фуллерита с целью выяснения особенностей температурной зависимости этого процесса. При этом учитывается реализация в разных температурных интервалах кристаллических структур фуллерита, сформированных фуллереновыми молекулами разной модификации. Экспериментальные исследования растворов фуллереновых молекул в различных органических средах позволили установить особенности температурной зависимости растворимости фуллерита C_{60}. Вместо предполагаемого увеличения растворимости с повышением температуры выявлена экстремальность ее температурной зависимости с максимумом при комнатной температуре, установлена эндотермичность процесса растворения. Аномалии температурной зависимости растворимости фуллеренов C_{60} в органических матрицах обусловлены изменением структуры связей в каркасе фуллереновых молекул, которые, в свою очередь, влияют и на структуру твердофазного фуллерита. Более точные результаты с использованием полученных формул могут быть рассчитаны, если будут известны из независимых экспериментов истинные значения энергетических параметров. Однако можно предположить, что характер выявленных закономерностей сохранится.

Ключевые слова: материалы, молекула, фуллерит, гранецентрированная кубическая (ГЦК), объёмно-центрированная кубическая (ОЦК), простая кубическая (ПК).

Introduction

At present, carbon materials, including nanostructured carbon modifications or their derivatives are belong to the rapidly developing field of materials science. A large number of scientists and engineers are engaged in their research. Carbon modifications include particles, one of the dimensions of which does not exceed 100 nm. To a greater extend, the term nanostructured modifications of carbon can be attributed to fullerenes and nanotubes and other similar structures. Fullerenes are spherical hollow carbon clusters with the number of atoms n = 30-120. C_{60}, C_{70}, C_{76}, and others [1–5] are known to be obtained in sufficiently large quantities. The most stable form is C_{60}, which spherical hollow structure consists of 20 hexagons and 12 pentagons. According to X-ray analysis, the average diameter of the sphere is 0.714 nm [1-5]. The internal bond in fullerene between the atoms is larger than the external one, therefore fullerenes form a solid body during consideration, with the preservation of their internal structure (fullerite) with a density of 1.65 g / cm^3. The ionization potential of fullerene is 7.6 eV, the affinity for an electron is 2.6-2.8 eV. The dissociation energy with a separation of C_2 and the formation of C_{58}-4.6 eV [6-12]. Fullerite is a molecular crystal formed from C_{60}, C_{70} and other molecule clusters, called fullerenes. The interest in the study of fullerenes increased significantly after the developing technology of their synthesis in macroscopic, gram quantities. According to some publications, arc synthesis is the most accessible and common method for producing fullerene-containing soot. Almost all technological parameters, the chemical frequency of evaporated graphite, as well as the size and shape of graphite electrodes, have a strong effect on the yield of fullerenes. The interest in studying the behavior of fullerenes in solutions is twofold and has both fundamental and applied aspects [13-19]. Fundamental interest in this issue is primarily due to the fact that fullerenes belong to the only one of the three currently known allotropic carbon modifications (graphite, diamond, fullerenes), which have appreciable solubility in a wide class of organic solvents. Another reason for the increased interest in the study of fullerenes in solutions is the fact that the there are existing methods for the preparation and cleaning of fullerenes in macroscopic quantities based on their use in solvents. As a result of intense thermal effects on the surface of crystalline graphite, soot is formed, which contains up to 20% fullerenes. In this case, it is most convenient to carry out thermal action by

means of an arc discharge between graphite electrodes in an atmosphere of a buffer gas, for which helium is usually used [20–26]. This report presents the results of calculations of the solubility of fullerite in aromatic and other media.

Results of the experiments and discussion

To solve the problem, the free energy of the system was calculated. The following provisions were taken into account:

1) implementation of three possible phases of fullerite with SC, BCC, FCC structures formed by fullerene molecules of different modifications (Φ_α, Φ_β, Φ_γ) and different activities [9],

2) the method of average energies [6-9], in which the interaction energy of molecules is determined by the intermolecular distance and their chemical nature;

3) model of spherically symmetric hard balls [10, 11], in which the interaction is taken into account not between atoms, but between molecules, as independent structural units;

4) the lack of order in the placement of all molecules at the sites of the crystal lattice;

5) neglect of the correlation in the substitution of the positions of the crystal lattices with both fullerene molecules and solvent molecules;

6) taking into account only the pair interaction and only the neighbouring molecules;

7) arbitrary composition of fullerite, as well as its single or two phase in different temperature ranges;

8) implementation in solution in different temperature ranges of SC, BCC, or FCC lattices, in some positions of which P solvent molecules are located. The latter position is realized at the boundary of a solid fullerite crystal and a liquid solvent.

When calculating the phase were designated as follows:

α phase – fullerite with SC lattice and
Φ_α molecules,
β phase – fullerite with BCC lattice and
Φ_β molecules,　　　　(1)
γ phase – fullerite with FCC lattice and
Φ_γ molecules.

The parameters of the considered lattices depend on the temperature and are somewhat different in various literature sources. We have taken the following values:

a_α = 14,08 Å for the PC structure,
a_β = 14,13 Å for the BCC structure,　　(2)
a_γ = 14,17 Å for the FCC structure,

The nearest intermolecular distances for each structure was determined by the following formulas:

$$r_\alpha = a_\alpha = 14{,}08 \text{ Å}, \quad r_\beta = a_\beta \sqrt{3}/2 = 12{,}23 \text{ Å},$$
$$r_\gamma = a_\gamma \sqrt{2}/2 = 10{,}02 \text{ Å}. \quad (3)$$

Thus, intermolecular distances are reduced in the following sequence: SC $\to$ BCC $\to$ FCC. The interaction energy due to this factor should increase in absolute value. However, it still depends on the activity of the fullerene molecules themselves, which largely determines the nature of the interaction.

To calculate the solubility of fullerite in an organic medium, the free energies of the α, β, γ phases were determined using the initial formula

$$F_i = E_i - kT \ln G_i - kTN_i \ln \lambda_i,$$
$$i = \alpha, \ \beta, \ \gamma, \quad (4)$$

where E_i is the internal configuration energy, determined by the sum of the energies of pair interactions of the nearest fullerite and solvent molecules, G_i is the thermodynamic probability of the distribution of fullerene molecules and solvent molecules, determined by the rules of combinatorics, N_i is the number of fullerene molecules in the solvent, λ_i is their activity, k is constant Boltzmann, T – absolute temperature.

The formula (4) takes into account the dependence of the free energy of the system on the number of fullerene molecules $(Ni \neq const)$ in the solvent, which changes during the dissolution of fullerite.

Configuration energy was determined by the expression:

$$E_i = -N_{\varphi\varphi}^{(i)} \upsilon_{\varphi\varphi}^{(i)} - N_{pp}^{(i)} \upsilon_{pp}^{(i)} - N_{\varphi p}^{(i)} \upsilon_{\varphi p}^{(i)}, \quad (5)$$

where $N_{\varphi\varphi}^{(i)}$, $N_{pp}^{(i)}$, $N_{\varphi p}^{(i)}$ are the numbers of the nearest pairs of *FF, PP, FR* in the phase, $\upsilon_{\varphi\varphi}^{(i)}$, $\upsilon_{pp}^{(i)}$, $\upsilon_{\varphi p}^{(i)}$ are the energies of their

interaction, taken with the opposite sign, $i = \alpha, \beta, \gamma$ are the type of phase.

The numbers of the nearest pairs of molecules in each phase i are equal

$$N_{\varphi\varphi}^{(i)} = \frac{1}{2} z_i N_i^2 / N, \quad N_{pp} = \frac{1}{2} z_i N_p^2 / N,$$

$$N_{\varphi p}^{(i)} = z_i N_i N_p / N, \tag{6}$$

where

$$N = N_i + N_p \neq const \tag{7}$$

is the number of lattice sites occupied by Φ_i and P molecules, N_i и N_p – the number of Φ_i and P molecules, z_i is the coordination number, which equals for the structures:

$$z_i = 6, 8, 12, \text{ respectively, for SC, BCC,} \atop \text{and FCC lattices.} \tag{8}$$

Configuration energy E_i taking into account formulas (5) – (7) is equal to

$$E_i = -\frac{1}{2} z_i \frac{1}{N_i + N_p} \times$$

$$\times (N_i^2 \upsilon_{\varphi\varphi}^{(i)} + N_p^2 \upsilon_{pp}^{(i)} + 2 N_i N_p \upsilon_{\varphi p}^{(i)}). \tag{9}$$

Thermodynamic probability is determined by the rules of the combinatory formula

$$G_i = \frac{N!}{N_i! N_p!}, \tag{10}$$

its natural logarithm with regard to the Stirling formula $\ln X! = X(\ln X–1)$, valid for large numbers X, is equal to

$$\ln G_i = N \ln N - N_i \ln N_i - N_p \ln N_p. \tag{11}$$

We obtain the free energy by substituting expression (4) into (9) and (11)

$$F_i = -\frac{z_i}{2(N_i + N_p)} (N_i^2 \upsilon_{\varphi\varphi}^{(i)} + N_p^2 \upsilon_{pp}^{(i)} + 2 N_i N_p \upsilon_{\varphi p}^{(i)}) -$$

$$-kT \Big[(N_i + N_p) \ln(N_i + N_p) - N_i \ln N_i - N_p \ln N_p - kT N_i \ln \lambda_i \Big]. \tag{12}$$

The equilibrium concentration of fullerenes in the solid solution is determined by minimizing the free energy. The latter is conveniently carried out using the Lagrange indeterminate multiplier method. For this the function is composed

$$\Psi_i = F_i + a\vartheta, \tag{13}$$

where a is the Lagrange multiplier, and

$$\vartheta \equiv N_i + N_p - N \tag{14}$$

bond condition (7).

Equilibrium equations were found from the conditions

$$\partial \Psi_i / \partial N_i + a \frac{\partial \vartheta}{\partial N_i} = 0,$$

$$\partial \Psi_i / \partial N_p + a \frac{\partial \vartheta}{\partial N_p} = 0, \quad i = \alpha, \beta, \gamma. \tag{15}$$

Substituting in (15) the free energy (12) and the bond condition (14), one can find the relations

$$-z_i(c_i \upsilon_{\varphi\varphi}^{(i)} + c_p \upsilon_{\varphi p}^{(i)}) + \frac{1}{2} z_i(c_i^2 \upsilon_{\varphi\varphi}^{(i)} + c_p^2 \upsilon_{pp}^{(i)} + 2 c_i c_p \upsilon_{\varphi p}^{(i)}) + kT \ln c_i - kT \ln \lambda_i + a = 0,$$

$$\tag{16}$$

$$-z_i(c_p \upsilon_{pp}^{(i)} + c_p \upsilon_{\varphi p}^{(i)}) + \frac{1}{2} z_i(c_i^2 \upsilon_{\varphi\varphi}^{(i)} + c_p^2 \upsilon_{pp}^{(i)} + 2 c_i c_p \upsilon_{\varphi p}^{(i)}) + kT \ln c_p + a = 0,$$

where

$$c_i = N_i/N, \quad c_p = N_p/N \qquad (17)$$

concentrations of fullerene molecules and solvent molecules.

Subtracting the second equation (16) from the first equation, eliminaing the Lagrange multiplier, the ratio can be obtained

$$z_i(\upsilon_{\varphi\varphi}^{(i)} - \upsilon_{pp}^{(i)} - c_i\omega_i) = kT \ln \frac{c_i}{\lambda_i(1-c_i)} \qquad (18)$$

where

$$\omega_i = 2\upsilon_{\varphi p}^{(i)} - \upsilon_{\varphi\varphi}^{(i)} - \upsilon_{\partial\partial}^{(i)} \qquad (19)$$

- mixing energy of each phase and it is taken into account that

$$c_i + c_p = 1 \qquad (20)$$

Formula (18) can be rewritten as

$$c_i = \left[1 + \frac{1}{\lambda_i} \exp \frac{-z_i(\upsilon_{\varphi\varphi}^{(i)} - \upsilon_{pp}^{(i)} - c_i\omega_i)}{kT} \right]^{-1}. \qquad (21)$$

This formula or formula (18) determines the temperature dependence of the solubility of fullerite in the matrix $c_i = c_i(T)$. The form of this dependence is determined by the energy parameter

$$V_i = z_i(\upsilon_{\varphi\varphi}^{(i)} - \upsilon_{pp}^{(i)} - c_i\omega_i), \qquad (22)$$

i.e. the nature of intermolecular interactions.

The study of function (16) on the extremum showed that for each phase the dependence $c_i = c_i(T)$ is monotonic: with increasing temperature, the solubility of fullerite increases or decreases depending on the sign of the energy V_i (22). However, in a wide temperature range, when phase transitions occur in the SC $\rightleftarrows$ BCC $\rightleftarrows$ FCC and the sign of the energy parameter V_i is possible, the dependence $c_i = c_i (T)$ can be extreme. Thus, the

developed statistical theory of the solubility of fullerite in an organic matrix, taking into account the formation of various modifications in different temperature ranges of fullerene molecules, i.e. different chemical activity and realization of fullerite with SC, BCC and FC lattices in these temperature ranges allowed us to explain and justify the temperature dependence of the solubility of fullerite in organic solvents, observed experimentally in matrix media such as hexane, toluene, xylene, etc. This fact gives hope on the existence of three stable isomers of C_{60} molecules, differing in activity and temperature stability, determined by the processes of changes in interatomic bonds with intramolecular on external molecular is realistic. We also note that more accurate results with the execution of the obtained formulas can be calculated if the true values of the energy parameters are known from independent experiments. However, it can be assumed that the nature of the revealed patterns remains.

Conclusion

Thus, the developed statistical theory of the solubility of fullerite in an organic matrix, taking into account the formation of various modifications in different temperature ranges of fullerene molecules, i.e. different chemical activity and realization of fullerite with PC, bcc and fc lattices in these temperature ranges allowed us to explain and justify the temperature dependence of the solubility of fullerite in organic solvents, observed experimentally in matrix media such as hexane, toluene, xylene, etc. This fact confirms the existence of three stable isomers of C60 molecules, distinguished by their activity and temperature stability, determined by the processes of changing intra-molecular and inter-atomic bonds to external molecular ones, is realistic. We also note that more accurate results with the execution of the formulas obtained can be calculated if the true values of the energy parameters are known from independent experiments. However, it can be assumed that the nature of the revealed patterns will remain.

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