Eur. Phys. J. D **15**, 57–63 (2001)

THE EUROPEAN PHYSICAL JOURNAL D
EDP Sciences

Structures and energetics of carbon bridged C_{60} clusters

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Received 17 January 2001 and Received in final form 26 February 2001

Abstract. The structures and energetics of carbon bridged C_{60} clusters $(C_{60})_nC_m$ have been studied by simulated annealing technique within the tight-binding molecular-dynamics. The "sp^2 addition" ball-and-chain dimers exhibit odd-even alternations over the number of chain atoms, with the dimers containing even chain atoms more stable against dissociation than their immediate neighbors containing odd chain atoms. In addition to the usual "sp^2 addition" dimers, a pentagon-linked C_{121} isomer and a hexagon-linked C_{122} isomer are also found to be stable. Based on our tight-binding calculations, trimers and larger clusters can be simply regarded as being made up of independent or weakly interacting dimers, if the C–C_{60} joints on a single cage are not too close to each other. Large C_{60} clusters connected by chains each containing only one or two carbon atoms have similar stability to that of constituent dimers, indicating the possibility to form stable C_{60}-carbon polymers.

PACS. 61.48.+c Fullerenes and fullerene-related materials – 61.46.+w Nanoscale materials: clusters, nanoparticles, nanotubes, and nanocrystals – 36.40.Qv Stability and fragmentation of clusters

1 Introduction

Recently, the fullerene derivatives have attracted much interest due to their special physical and chemical properties, as well as their possible applications. Particular attentions have been paid to the structures and energetics of various fullerene clusters in some recent studies. The exposure of solid C_{60} to ultraviolet and visible laser light was found to produce strong bonds between the C_{60} molecules, yielding C_{60} oligomers or polymers [1,2]. The fullerene oxides ($C_{60}O$) and oxygen bridged C_{60} dimer (C_{60}–O–C_{60}) and trimer ($C_{180}O_2$) have been studied both experimentally [3,4] and theoretically [5–7]. Interest in carbon bridged fullerene clusters has also arisen. Martin and coworkers studied the gas-phase clusters produced by coevaporization of C_{60} and graphite, using a photoionization time-of-flight mass spectrometry [8]. They observed numerous clusters like $(C_{60})_nC_m$, with "n" up to "$n = 7$". Clusters with both odd and even carbon atoms were simultaneously found, and there was no preference for clusters containing an even number of carbon atoms. There existed a critical stability edge of $m \geq n-1$, due to the requirement that at least $n-1$ carbon atoms were needed to stabilize a cluster containing n C_{60} cages. Clusters with $m < n-1$ were significantly less stable and were depleted after a moderate laser heating. Upon laser heating, the clusters evaporated entire C_{60} molecules. Therefore, they concluded that the observed clusters were polymerized C_{60}-carbon clusters. The fragmentation behavior of C_{121}, C_{122}, C_{123} were also studied [8]. Chemically, the dumbbell-like molecules C_{121} and C_{122} have been synthesized, isolated and characterized [9,10]. Most recently, Dragoe *et al.* have isolated and characterized an unsymmetrical bisfullerene C_{121}, with the bridge atom connecting to one C_{60} molecule through an open [5.6] ring junction (*i.e.*, between a pentagon and a hexagon) and to another C_{60} molecule through a closed [6.6] ring junction. This unsymmetrical structure was found to be more stable than either of the symmetrical counterparts, though the two symmetrical molecules also existed in the experiment [11]. Shvartsburg *et al.* systematically studied the fullerene dimers obtained from the laser desorption products of C_{60} and C_{70}, by a high-resolution ion mobility measurements combined with trajectory calculations [12–14]. The laser desorption of fullerenes produced carbon clusters containing only even number of carbon atoms. The "exact dimers" and the species with slightly fewer atoms than the exact dimers were fully coalesced, large single-shell fullerenes and [2+2] cycloadducts. The species with more atoms than the exact dimers assumed a "ball-and-chain" structure besides the coalesced single cage fullerenes and [2+2] cycloadducts. The structures and energetics of ball-and-chain dimers with even chain atoms have been studied theoretically by the density functional tight binding(DFTB) method [12]. However, except for the [2+2] cycloaddition dimers and the ball-and-chain dimers with even chain atoms, the structures and the relative stabilities of the $(C_{60})_nC_m$ clusters are generally unknown. For example, schematic drawing of several possible

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configurations of tetramers was given in Figure 4 of reference [8]. However, these possible structures could not be determined from a mass spectrometry measurement [8]. In this paper, we carry out tight-binding molecular dynamics simulations to study the structures and energetics of various isomers of C_{60}-carbon clusters to fill the deficiency.

2 Tight-binding molecular dynamics simulations

In this work, we use a tight-binding potential model for carbon [15]. This potential model is quite simple but it includes the essential quantum mechanics of the covalent bonding in carbon systems. In the tight-binding model, the total energy of the system is expressed as

$$E_{\mathrm{T}}(\{\mathbf{r}_i\}) = \sum_{n}^{\text{occupied}} \langle \Psi_n | H_{\mathrm{TB}}(\{\mathbf{r}_i\}) | \Psi_n \rangle + E_{\mathrm{rep}}(\{\mathbf{r}_i\}).$$

The first term is the electronic energy calculated from a parametrized tight-binding Hamiltonian $H_{\mathrm{TB}}(\{\mathbf{r}_i\})$, and the second term is a short-ranged repulsive potential. The potentials and the parameters were obtained by fitting to the first-principle local density approximation (LDA) results for the energies of diamond, graphite and linear chain as a function of bond length [15]. This tight-binding potential model has been used extensively to study the fullerene geometry from C_{20} to C_{102}, and was proved to be very successful [16–19]. For example, this tight-binding model gives the bond length of C_{60} to be 1.396 Å and 1.458 Å, respectively. The calculated cohesive energy per atom is 0.41 eV higher than that of graphite, and the HOMO-LUMO gap is 1.61 eV. These results are all in good agreement with the results of LDA [20].

We use a Verlet algorithm for the molecular dynamics, with a time step of 1.0 fs. The structures of the various isomers of $(C_{60})_nC_m$ clusters are obtained by means of simulated annealing technique. The various clusters are first equilibrated at 77 K, in consistency with the experiment where the clusters formed in a chamber cooled by liquid nitrogen [8]. Then the system is cooled down till $T = 0$ K, with the structure fully relaxed.

3 Results and discussions

The polymerized C_{60} clusters are made up of C_{60} cages connected by carbon atoms. Thus the clusters can be classified by the number of C_{60} cages included, the number of bridging carbon atoms, and the characteristics of C–C_{60} joints. There are several basic joints between the cage and the chain (see Fig. 3 of Ref. [12]). One is the "*sp* addition", with a single bond connecting an *sp* hybridized terminal atom of a chain to a fullerene atom. The other type of joint is "sp^2 addition" , with an sp^2 hybridized terminal atom of a chain added across a fullerene bond. In the special case of $(C_{60})_nC_{n-1}$, there is only one sp^3 hybridized carbon atom in each chain, thus we call the joints as "sp^3 additions", in the same spirit of "sp^2 addition". While there is an mixture of sp^2 addition and sp^3 addition joints in one cluster, the joints of the molecule are denoted as sp^2/sp^3. The C–C bond of a C_{60} cage between a six membered ring and a five membered ring is referred to as 65 bond; similarly we have bond 66. For both 66 and 65 sp^2 or sp^3 addition, the bonds on C_{60} cage are found to be severed, yielding open-65 and open-66 sp^2 or sp^3 joints, same as in DFTB calculations [12]. There is also a [2+2] cycloaddition while two 66 bonds on two cages come close and parallel to each other.

The structures, total energies, dissociation energies and the HOMO-LUMO gaps of the various C_{60}–C clusters and their isomers are calculated. For convenience of comparison, we define the relative total energy of the molecule as its total energy relative to that of the independent constituent C_{60} cages. The dissociation energy is defined as the energy cost for a molecule to dissociate along the lowest energy pathway. It was believed that the dissociation energy is close to the dissociation energy barrier in ball-and-chain dimers [12]. It was also shown in the photo-fragmentation mass spectra measurement that under low laser intensities, the C_{121} molecule dissociated into C_{60} and C_{61}, and the C_{122} dissociated into C_{60} and C_{62}. The dissociation barriers were determined to be between 1.5 and 2.5 eV [8]. The DFTB calculations also showed that the lowest energy dissociation pathway for a ball-and-chain dimer always involves the severance of one chain-C_{60} joint. The fragmentation into two tadpoles or two C_{60} cages and one chain requires much higher energy [12]. Present tight-binding calculations also confirm this point. The HOMO-LUMO gap is the energy difference between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO). The HOMO-LUMO gap may be related to the chemical reactivity of the molecule. It is found that the more abundant fullerenes in mass spectra have relatively large HOMO-LUMO gaps, while those absent in the mass spectra have very small gaps [17].

3.1 C_{121} and C_{122}

The C_{121} and C_{122} dimers are quite abundant in the products of co-evaporization of graphite and C_{60} fullerenes [8]. Their various isomers are therefore carefully studied in this work. The bridging atom in the "sp^3 addition" C_{121} isomer is tetrahedrally coordinated and robustly bonded, thus the open 65/65, 65/66, 66/66 "sp^3 addition" isomers should be stable. The calculated energies support this point. As shown in Table 1, all of the three isomers have relatively low energies, large dissociation energies and large HOMO-LUMO gaps. Among the three isomers, the open 65/65 is the most stable one, while open 65/66 isomer has similar stability to that of open 66/66 isomer, but is less stable than open 65/65 isomer. In C_{122} and the other ball-and-chain dimers with longer chains, similar trend is found, in agreement with the DFTB calculations [12]. The recent finding of an unsymmetrical C_{121}

Table 1. Relative total energies, dissociation energies, and HOMO-LUMO gaps of the dimers C_{121} and C_{122}. The "connection" indicates the two joints of the dimer. The relative energies are the energies of the molecules excluding the part of the independent constituent C_{60} cages; dissociation energy is the energy cost along the lowest dissociation pathway, and the data in parentheses is the result of DFTB [12].

Molecule	Connection	Total energy (eV)	Dissociation energy (eV)	HOMO-LUMO (eV)
C_{121}	65/65 sp^3	−7.13	2.53	1.32
	65/66 sp^3	−6.32	1.81	0.88
	66/66 sp^3	−5.69	1.80	0.84
	pentagon	−5.52	2.96	0.98
	65/65 sp^2	−5.95	1.36	0.25
	66/66 sp^2	−5.52	1.64	0.46
	sp	−2.57	0.0	0.28
C_{122}	65/65 sp^2	−14.25	2.77 (2.61)	1.18
	65/66 sp^2	−13.51	2.02	0.86
	66/66 sp^2	−12.78	2.08 (2.16)	0.79
	double [2+2]	−11.06	3.27 (2.80)	1.42
	hexagon	−10.16	2.38	0.68

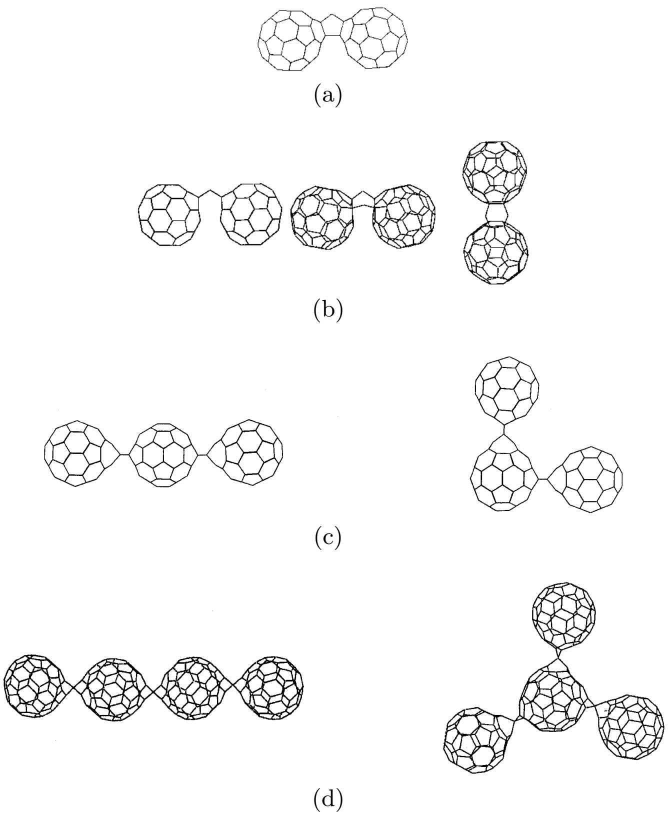

Fig. 1. C_{121} and C_{122} isomers: (a) pentagon linked C_{121}; (b) hexagon linked C_{122}; (c) linear and "L" shape open-66 sp^3 addition C_{182}; (d) linear and star-like open-65 sp^3 addition C_{243}.

dimer, with the bridging atom connecting to one C_{60} cage through an open 65 bond, while connecting to another cage through a closed 66 bond [11], is unfortunately unattainable in this tight-binding scheme. We always get open sp^2 additions or open sp^3 additions, which is the same in DFTB [12]. Compared with the *ab initio* results [11], we guess this is due to the minimal sp^3 basis used in the tight-binding schemes. Another stable structure of C_{121} is the closed 66/66 pentagon-linked isomer (Fig. 1a), which is very similar to the most stable structure of a hetronuclear fullerene dimer C_{60}–O–C_{60} [6,7]. As shown in Table 1, its total energy is not high, and the dissociation energy is quite large. Thus, the pentagon-linked C_{121} isomer should be very stable. All the four isomers described above might exist in the products of co-evaporization of C_{60} and graphite. Table 1 shows the structure of C_{121} *sp* addition is very unstable; the C_{121} sp^2 addition structures are also not very stable. These three structures might not exist or would be very scarce. The dissociation energy of C_{121} was measured to be about 2.0 eV [8]. Since the measured C_{121} molecules should be a mixture of several stable isomers, the corresponding dissociation energy is an average of the various clusters. From Table 1, we learn that an average dissociation energy of about 2.0 eV for the stable C_{121} isomers is reasonable, consistent with the experiment.

For C_{122}, the open 65/65, 65/66, 66/66 sp^2 addition isomers are also stable, as the two bridging atoms in each isomer are sp^2 hybridized and strongly bonded. The obtained dissociation energies are in good agreement with the DFTB results (see Tab. 1). The energy difference between the 66/66 and 65/65 isomer is about 1.5 eV, larger than the DFTB result [12]. However, present tight-binding model does give the correct relative energy order of the various isomers. A closed-66 "double [2+2]" isomer for C_{122} is also found stable, consistent with the DFTB result [12]. Besides all the C_{122} isomers described above, a closed 66/66 hexagon-linked structure (Fig. 1b) is obtained by simulated annealing, which can be viewed as a deformation of the "benzene-link" structure (Fig. 8d of Ref. [12]). The "benzene-link" structure was found to be unstable from a DFTB optimization, although the ion mobility measurement preferred a structure close to that [8]. In our simulated annealing studies, we start from the "benzene link" structure, and find that the *sp* joints of the "benzene link" structure deform to let the two C_{60} cages go closer to each other. This is energetically favorable, since two hexagons on the two C_{60} cages are brought close and face to face. The distances between the nearest atoms on the two cages range from 2.2 Å to 2.6 Å, where the carbon atoms exhibit attractive interactions at such distances. Similar phenomenon in ball-and-chain dimers also exists [12]. The obtained total energy of this deformed "benzene link" (referred to as "hexagon link" hereafter) is only 0.9 eV higher than that of the double [2+2] isomer, the dissociation energy of which is large (2.38 eV), and its HOMO-LUMO gap is not very small (0.68 eV). So this structure is expected to be stable. It appears to have been observed using ion mobility measurements [12,21].

3.2 Ball-and-chain dimers

Produced from laser desorption of fullerenes, the ball-and-chain dimers with even chain atoms have been reported by Shvartsburg *et al.* [12]. However, dimers with odd chain atoms should exist, since both odd numbered and even numbered clusters were simultaneously observed [8]. Open 66/66 and 65/65 sp^2 ball-and-chain dimers (up to C_{130}) have been calculated in this work. We find odd-even alternations over the number of chain atoms for both structures and energetics of the ball-and-chain dimers.

The total energies of the ball-and-chain dimers decrease linearly with increasing chain atoms, with no odd-even alternation. For both the open 66/66 and 65/65 sp^2 dimers, the total energy decreases about 7.2 eV upon adding one carbon atom to the chain, which is only 0.8 eV higher than the average cohesive energy of C_{60}. But the structures are a bit different. For sp^2 adducts, the chain and the joint bond in the cage form a plane. For those with odd number of chain atoms, the two joint planes are approximately perpendicular to each other, while for those with even number of chain atoms, the two joint planes are approximately parallel to each other. This phenomena can be explained by the π bond couplings. The dissociation energies also exhibit an odd-even alternation over the chain length (see Fig. 2). The dimers with an even number of chain atoms have a larger dissociation energy than their immediate neighbors with an odd number of chain atoms. This might be due to the odd-even alternation of an independent carbon chain. The lowest energy pathway of dissociation for ball-and-chain dimers is to sever one C_{60}-carbon joint, yielding a cage and a "tadpole". It is well known that short carbon chains show strong odd-even alternations, with the odd-numbered chains being more stable than the even-numbered ones [22]. The energy of the "tadpole" also shows a strong odd-even alternation, where the "tail" of the "tadpole" behaves like an independent chain. From our calculations, it is clear that a tadpole with odd carbon atoms in the tail is lower in energy (by average) than a tadpole with even atoms in the tail. As a result, the energy cost for the odd numbered-chain dimer to dissociate into a C_{60} cage and a tadpole is smaller. Hence, dimers with odd chain atoms are less stable than dimers with even chain atoms. From Figure 2, we also learn that the dissociation energies of ball-and-chain dimers decrease with increasing number of C_2 units in the chain. Thus those dimers with long chains are less stable than those with short chains. As shown in Figure 3, the HOMO-LUMO gaps of ball-and-chain dimers also shows an odd-even alternation. For the open 65/65 sp^2 dimers, the HOMO-LUMO gaps of those with even chain atoms are smaller than those with odd chain atoms, just opposite to the order of dissociation energy. For the open 66-66 sp^2 dimers, the odd-even alternation of the gap is not so evident. The HOMO-LUMO gaps also decrease with increasing number of C_2 units in the chain. We conclude that ball-and-chain dimers with even number of chain atoms are more stable against dissociation than their immediate neighbors with odd number of chain atoms.

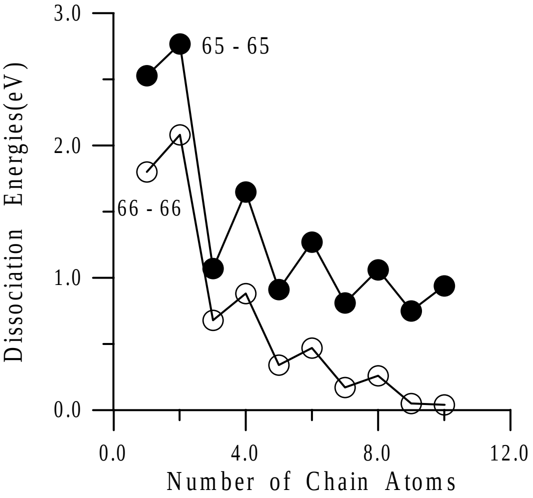

Fig. 2. Dissociation energies of the ball-and-chain dimers *versus* the number of chain atoms.

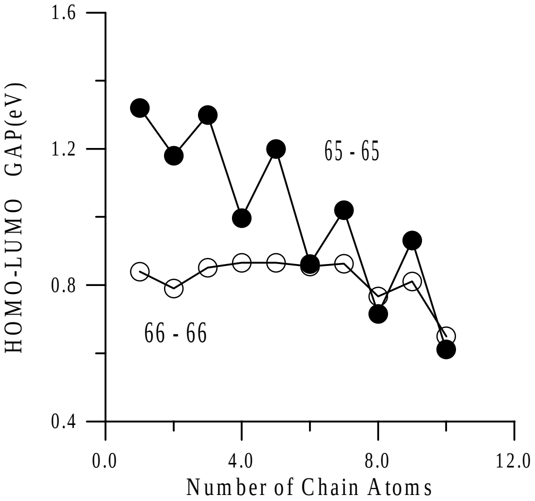

Fig. 3. HOMO-LUMO gaps of the ball-and-chain dimers *versus* the number of chain atoms.

Table 2. Relative total energies, dissociation energies and HOMO-LUMO gaps of the trimers C_{182}, C_{183}, and C_{188}. The cluster shape is indicated in the parentheses, and all C_{188} isomers considered are linear shape. The two integers in parentheses in the connection column of C_{188} indicate the number of atoms in the two bridges. For C_{188}, the dissociation energies are the energy cost severing the longer chain-C_{60} joint; the data in the parentheses of dissociation energy column is the energy cost severing the shorter chain-C_{60} joint.

Molecule	Connection	Total energy (eV)	Dissociation energy (eV)	HOMO-LUMO (eV)
C_{182}	66 sp^3 (linear)	−11.34	1.80	0.67
	66 sp^3 (L)	−11.47	1.95	0.81
	65 sp^3 (linear)	−14.17	2.53	1.27
	65 sp^3 (L)	−14.14	2.51	1.31
	pentagon (linear)	−11.06	2.96	0.94
C_{183}	66 sp^2/sp^3 (linear)	−18.44	1.79	0.64
	65 sp^2/sp^3 (linear)	−21.30	2.51	1.27
C_{188}	65 sp^2/sp^3 $(1, 7)$	−56.32	0.80 (2.51)	1.14
	65 sp^2 $(2, 6)$	−56.30	1.16 (2.72)	1.04
	65 sp^2 $(3, 5)$	−55.70	0.90 (1.05)	1.23
	65 sp^2 $(4, 4)$	−56.30	1.61	0.98

3.3 Trimers

Trimers are made up of three C_{60} cages connected by two bridges. We consider only the same kind of joints in one isomer, and the 65/65 sp^3 is shortened as 65 sp^3 in the tables and text, following reference [12]. Since there are two C–C_{60} joints on the middle cage of the C_{60} trimer, one may ask the question whether the two joints on one C_{60} cage affect each other or not. First, let us take a look at the shape dependence of the total energies. The centers of mass of the three C_{60} cages can either form a linear or an "L" shape (Fig. 1c). From Table 2 we learn that both linear shape and "L" shape C_{182} isomers have similar total energies, dissociation energies and HOMO-LUMO gaps. This is similar to the situation in [2+2] cycloaddition C_{180} clusters [7]. Thus the energetics of the C_{182} isomers are independent of the molecule shape (*i.e.*, the relative position of the C–C_{60} joints on the central cage). Secondly, let us analyze the dependence of trimer's energy on its components. Intuitively, the trimers can be viewed as being made up of two dimers. From Tables 1 and 2 we know that the total energy of the trimers is the summation of two independent constituent dimers, while the dissociation energy and HOMO-LUMO gap are the smaller one of the two constituent dimers. Therefore, based on our tight-binding calculations, the trimers can be viewed as being made up of two independent or weakly interacting dimers as long as the two C–C_{60} joints on the central cage are far apart from each other. For trimer C_{188}, there are eight bridge carbon atoms in total. The two bridges may contain one and seven, two and six, three and five, four and four carbon atoms, respectively. Again, the total energy is the summation of the independent constituent dimers. Since the longer chain dimers are less stable, the C_{188} isomers dissociate by severing the joint between the longer chain and a C_{60}.

3.4 Tetramers and larger clusters

The tetramers and larger clusters that are connected by only one kind of sp^2 or sp^3 joints in each isomer are considered. Several possible configurations for tetramers was given in Figure 4 of reference [8]. However, the purpose of this work is not to check these structures one by one, but is to find out the general rule for these structures by observing the interactions between the different joints on any single cage.

While several C_{60} cages are connected to a single cage by carbon atoms, the centers of mass of the cages form a "star" (Fig. 1d). We call this kind of structures star-like clusters. We have calculated star-like C_{60} clusters with four C_{60} cages connected to a central cage (*e.g.*, C_{304}, see Tab. 3). The carbon-C_{60} joints are separated from each other by at least two pentagon or hexagon rings. Table 3 shows that linear shape clusters and star-like clusters with the same kind of joints have similar energies, just like the case in trimers. Furthermore, the energies of tetramers and larger clusters are also the summation of constituent dimers. For example, the total energies of linear 65 sp^3 C_{243} and C_{304} isomers are approximately three and four times that of the 65 sp^3 C_{121} dimer. Therefore, tetramers and larger clusters can also be regarded as being made up of independent or weakly interacting dimers, as long as the C–C_{60} joints on a single cage are not too close to each other. Considering the different arrangement of the joints, there may be a large variety of different isomers with similar stability, which is consistent with the experimental observations [8].

The situation is quite similar for larger clusters. The dissociation energy, HOMO-LUMO gap of linear-shape, 65 sp^3 $(C_{60})_5C_4$ are quite close to those of 65 sp^3 $(C_{60})_2C_1$. Thus the larger cluster has similar stability to its constituent dimers. For linear-shape $(C_{60})_5C_5$ and

Table 3. Relative total energies, dissociation energies and HOMO-LUMO gaps of tetramers and larger clusters. The shape of the clusters is denoted in the parentheses in the connection column.

Molecule	Connection	Total energy (eV)	Dissociation energy (eV)	HOMO-LUMO (eV)
C_{243}	65 sp^3 (linear)	−21.26	2.50	1.25
	65 sp^3 (star)	−21.00	2.36	1.24
	66 sp^3 (linear)	−17.00	1.73	0.66
	66 sp^3 (star)	−17.04	1.66	0.80
C_{244}	65 sp^3 (square)	−27.67	4.24	1.17
	66 sp^3 (square)	−23.20	3.76	0.82
C_{304}	65 sp^3 (linear)	−28.30	2.45	1.25
	65 sp^3 (star)	−27.88	2.28	1.15
	66 sp^3 (linear)	−22.64	1.73	0.63
	66 sp^3 (star)	−22.67	1.55	0.51
C_{305}	65 sp^2/sp^3 (linear)	−35.37	2.28	1.24
C_{366}	65 sp^2/sp^3 (linear)	−42.31	2.28	1.25

$(C_{60})_6C_6$, the results are similar. There is no sign for the dissociation energies and HOMO-LUMO gaps to decrease with increasing number of C_{60} cages. It is thus possible to form quite long C_{60} polymers with one or two carbon atoms in each bridge. Besides the chain-like and star-like clusters, a square-like tetramer C_{244}, which is constructed by an “L” shape trimer and a monomer, is also stable. The square-like C_{244} has a closed structure; it needs to break two joints to evaporate one C_{60} cage from the C_{244} molecule. The dissociation energy is approximately twice that of linear clusters.

4 Summary

In this paper we have studied the structures and energetics of carbon bridged C_{60} clusters $(C_{60})_nC_m$ using a simulated annealing technique of tight-binding molecular dynamics. A pentagon-linked C_{121} isomer and a hexagon-linked C_{122} isomer are found stable. For the ball-and-chain dimers, the dissociation energies and HOMO-LUMO gaps decrease with increasing number of C_2 units in the chain. Notably, the structures and energies of ball-and-chain dimers exhibit odd-even alternations over the number of chain atoms. The two joint planes are approximately either perpendicular or parallel to each other depending on whether the number of chain atoms is odd or even; the dimers with even chain atoms are more stable against dissociation than their immediate neighbors with odd chain atoms, while the HOMO-LUMO gaps of the former are smaller. Based on our tight-binding calculations, trimers and larger clusters can be regarded as being made up of independent or weakly interacting dimers, if the C–C_{60} joints on a single cage are not too close to each other (farther than two hexagon or pentagon rings). The relative total energies (excluding the part of the cages) are summation of the independent constituent dimers; the dissociation energy of the larger cluster is just the smallest one among those of the constituent dimers. Considering the stability of large clusters, we have calculated the energies of linear-shape sp^2 or sp^3 $(C_{60})_nC_m$ $(m = n - 1$ or $n)$ with “n” up to “$n = 6$”. There is no noticeable decrease of the dissociation energies and HOMO-LUMO gaps with increasing number of C_{60} cages, indicating the possibility to form stable C_{60}-carbon polymers.

We thank Prof. K.M. Ho and Dr. A.A. Shvartsburg for helpful discussions and comments on the results. D.K. Yu would like to thank Profs. D.S. Wang and E.G. Wang for helpful discussions.

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