Self-organising formation of fullerene molecules from graphene patterns

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Abstract. The self-organised formation of C\textsubscript{60} and C\textsubscript{70} from well designed graphene patterns was proved by molecular dynamic simulations. In this paper graphene nanopattern models are shown which can be used for self-organised formations of larger fullerene molecules. The rules of building of the nanopatterns are defined. Examples are shown for the self-organised processes in case of larger fullerene molecules.

1. Introduction

Lately, more and more researchers are paying attention to the self-organizing behaviour of the graphene patterns. As there is missing only an order of magnitude for obtaining atomic precise nanopatterns [1], theoretical calculations are motivated to simulate the formation of 3D nanostructures from graphene patterns. In general, they are looking for the answers given, regarding the different interventions and interactions, with molecular dynamic simulations, mainly in the form of upwinding (curling) on pre-designed formations.

It was shown that owing to the van der Waals interaction, graphene nano-ribbons wind up, in a self-organizing manner, on nanoscale metallic wires [2]. In case of graphene ribbons, an upwinding, taking place in a self-organizing manner, was also examined where, instead of a straight metal wire, a carbon nanotube was the van der Waals interaction provider factor, triggering the upwinding [3]. The self-organising upwinding was triggered by fullerene molecules [4], as well. The deliberately created defects on the graphene ribbons were examined, with molecular dynamics simulation as well, as factors causing the folding of the graphene ribbons [5]. Cone-like self-organizing folding of slashed graphene platelets was studied [6]. It was proved by molecular dynamics calculations that C\textsubscript{60} and C\textsubscript{70} fullerene molecules can be formed from one graphene pattern [7-9]. It was also proved that zigzag and armchair nanotubes can be obtained from graphene patterns. As the driving force was generated with the help of a half fullerene the final tube was closed with a half fullerene at one end [9]. Creating nanotubes from two nanoribbons is important because in this way open-ended nanotubes can be produced [9]. The production of nanotubes from two parallel nanoribbons is not a trivial task. First the possibility was published [10-11], and after that the topological and energetic conditions for obtaining perfect tubes were published [12-13].

In this paper we show that, similarly to the case of the C\textsubscript{60} and the C\textsubscript{70}, large fullerene molecules can be formed from well-designed graphene patterns by self-organising way.
2. Method
In our molecular dynamics simulations the carbon-carbon interaction were calculated with the help of Density Functional Theory adjusted Tight Binding method (DFT-TB) [14]. The time step was $\Delta t=0.7$ fs and Verlet algorithm [15] was used in calculating velocities of the carbon atoms. The environmental temperature was controlled with the help of Nosé-Hoover thermostat [16-17]. Applying the Nosé-Hoover thermostat made it possible that we could start our simulation with randomly determined initial velocities of the carbon atoms. The corresponding $T_{\text{init}}$ atomic temperature was different of the environmental temperature of the simulation. This initial temperature was taken from the range [1000, 1100] Kelvin.

3. Examples for self-organising formation of fullerene molecules
In this chapter results are shown which were run by molecular dynamics simulations for the self-organising formation of larger fullerene molecules.

In Figure 1 snapshots from the simulation for the case of C$_{88}$ molecule can be seen. The input model (Fig. 1a) consists of 88 carbon atoms, so the graphene pattern has 88 vertices. The 3D structure starts to develop so that several chemical bonds is coming into existence between atom pairs being close to each other at the perimeter, the new bonds compose pentagons with the neighbouring bonds, therefore the pattern starts to curve (Fig. 1b). Subsequently the number of the new bonds and so the new pentagons is rising, therefore the structure is curving more and more largely (Fig. 1c-d), in the end the structure is closing up and the C$_{88}$ molecule is being born (Fig. 1e).

![Figure 1. Snapshots from the simulation of self-organising process of C$_{88}$](image)

The process in the case of the C$_{100}$ molecule is very similar.

In Figure 2 snapshots from the simulation for the case of C$_{100}$ molecule can be seen. The input model (Fig. 2a) consists of 100 carbon atoms, accordingly the graphene pattern has 100 vertices. The 3D structure starts to develop so that several chemical bonds is coming into existence between atom pairs being close to each other at the perimeter, the new bonds compose pentagons with the neighbouring bonds, therefore the pattern starts to curve (Fig. 2b). Subsequently the number of the new bonds and so the new pentagons is rising, therefore the structure is curving more and more largely (Fig. 2c-d), in the end the structure is closing up and the C$_{100}$ molecule is evolving (Fig. 2e).

![Figure 2. Snapshots from the simulation of self-organising process of C$_{100}$](image)
4. Rules of planning graphene patterns for self-organising formations of fullerene molecules

The planning of the graphene patterns (cutting from the graphene sheet) has the following rules:

1. The number of the vertices in the graphene pattern has to be equal to the number of the atoms in the planned fullerene molecule.
2. The pattern has to consist of only hexagons.
3. The pattern has to be contiguous.
4. Curving up the pattern the planned fullerene structure has to recover.

These rules seem to be simple however they can be fulfilled not so simple way. In Figure 3 patterns planned for the formation of $C_{140}$, $C_{240}$ and $C_{540}$ fullerene molecules are shown. It is clear that there are a number of different patterns having e.g. 140 vertices. Only few of them fulfil the condition of 4, that is to say, the structure of $C_{140}$ recovers only in case of few patterns after curving up the pattern. The structure recovering with the curving up of the pattern of Figure 3a is shown in Figure 3d. The patterns shown in Figure 3b-c are planned for $C_{240}$ and $C_{540}$, that is, after curving up them the structure of $C_{240}$ and $C_{540}$ recover.

![Graphene patterns for self-organising formation of $C_{140}$, $C_{240}$, $C_{540}$ (a-c) and the curved up pattern of the $C_{140}$ (d)](image)

Figure 3. Graphene patterns for self-organising formation of $C_{140}$, $C_{240}$, $C_{540}$ (a-c) and the curved up pattern of the $C_{140}$ (d)

5. Conclusions

According to the results of molecular dynamics simulations, similarly to the self-organising formation of $C_{60}$ and $C_{70}$ (which was already shown earlier [7-9]) larger fullerene molecules can develop from pre-designed graphene patterns, as well.
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