Thermal stability of the endohedral complex of [60]fullerene with tetrahedrane, $C_4H_4@C_{60}$

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The structure and stability of the C_4H_4 tetrahedrane molecule encapsulated inside the C_{60} cage was theoretically investigated via both molecular dynamics (MD) and potential energy search (PES) methods. All calculations were performed using the nonorthogonal tight-binding total energy model, originally developed for hydrocarbon compounds [1].

The heat of formation of the $C_4H_4@C_{60}$ complex was found to be 758.0 kcal/mol. The C–C and C–H bond lengths in tetrahedrane are equal to 1.524 A and 1.068 A for isolated C_4H_4 molecule, and 1.499 A and 1.043 A for $C_4H_4@C_{60}$ complex, respectively. The main decomposition channel of the encapsulated tetrahedrane molecule is its isomerisation to cyclobutadiene, as for isolated one. Energy and structure of the saddle configuration, which separates tetrahedrane@C_{60} from cyclobutadiene@C_{60} on the reaction path, was obtained using PES technique. The minimum energy barrier preventing tetrahedrane decay inside the fullerene cage was found to be 0.46 eV, which coincides with the same barrier for isolated C_4H_4 molecule. On the other hand, at the presence of fullerene cage the energy barrier for the reverse reaction significantly changes: it decreases from 3.06 eV to 2.05 eV.

To study the evolution of the excited $C_4H_4@C_{60}$ endohedral complex, we used the MD approach with the same tight-binding potential [1]. At the initial instant of time, all atoms were given random velocities and displacements, such that the momentum and the angular momentum of the whole system were equal to zero. The classical Newton equations of motion were solved numerically using the standard velocity Verlet algorithm. Combining the results of both PES and MD calculations, we derived an activation energy E_a and the frequency factor A in the Arrhenius equation $\tau^{-1}(T) = A\exp(-E_a/k_B T)$, describing temperature dependence of the encapsulated into the [60]fullerene cage tetrahedrane molecule lifetime τ (k_B is the Boltzmann's constant). These values are $E_a = (0.46 \pm 0.01)$ eV and $A = 10^{14.81\pm0.64}$ s^{-1}.

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[1] M.M. Maslov, A.I. Podlivaev and L.A. Openov. *Phys. Lett. A* **373**, 1653 (2009).