MOLECULAR DYNAMICS OF CATENANES AND MOLECULAR MACHINES OF NONBIOLOGICAL NATURE

Ye. V. Tourley*, K. V. Shaitan
Moscow 119899, Vorobjovy Gory, MSU, Faculty of Biology, Dept. of Biophysics

Molecular machines mentioned below are meant to be such molecular systems that use conformational mobility for functioning. Unlike fluctuations of valent bond lengths and valent angles conformational movements are concerned with hindered rotation around chemical bonds and with molecular construction deformations with formation and breakage of nonvalent bonds. Components of molecular machines move mainly by means of restricted diffusion [1]. As an example of molecular machines of nonbiological nature catenanes (compounds with two interlocked molecular rings) could be proposed. Thus, for example, model catenane – (2)-(cyclo-bis(paraquat-p-phenylene))-(1(2,6)-tetrathiafulvalena-16(1,5)naphtalena-3, 6, 9, 12, 15, 17, 20, 23, 26, 29-decaoxatriacontaphane)-catenane – changes its redox status when being applied an electric field, and rotation of the rings takes place. It occurs with a fixation at certain moments of the influence [2]. To find out characteristic properties of rings movements under various external conditions (temperature, magnitude and frequency of an imposed electric field) computer simulation of the molecule dynamics was carried out. Three cationic forms of the catenane were subjected to geometrical optimization and quantum chemical calculation of the molecule charge distribution. Restricted Hartree-Fock method with open shells was used, and the optimization was carried out in STO-3G basis. The partial charges were calculated according to Loewdin. Whole quantum chemical set of calculations was executed in software package GAMESS© [3]. Molecular dynamics calculation was carried out with use of the original software developed at the Moscow State University. The force field used for calculation is modified Amber99© proceeded from results of quantum chemical calculations.

The calculated time dependences of rotation angles, angular velocities and dipole moments of rings allowed to find out two types of movements – large-scale rotation of one of the constituent rings with times about 30 ns at 2000 K and stochastic rotary movements of the rings with times about several nanoseconds. The latter times are of the same order as the system dielectric relaxation time. The leading role in coordination of mutual rotation is attributed to strongly charged cyclophane ring in which no rotation with angle more than 180° occurs. Distribution of charges in rings exerts strong influence upon dynamics of the catenane. The obtained results do not conflict with known experimental data.

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E-mail: yegr@moldyn.ru, egry@biophys.msu.ru