Interaction of oxygen with fullerenes: oxidation versus singlet oxygen production

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Photophysical and bactericidal action of fullerenes and its derivatives are believed to originate from the interaction with oxygen in two ways: through electron transfer resulting to the formation of super oxides (and other reactive oxygen species ROS), and secondly through energy transfer to triplet oxygen forming singlet oxygen. The latter process is not understood: it is agreed that the quenching of singlet oxygen occurs via "an unknown mechanism" [1]. While fullerenes dissolved in organic solvent are very efficient singlet oxygen generators, in solid state their bactericidal action decreases but not disappears [2]. Motivated by this fact, we simulated the possible routes of interaction of oxygen with fullerenes.

Singlet oxygen generation was examined on the basis of resonance energy transfer from the first excited triplet state of isolated C_{60} molecule to triplet oxygen. Based on the values of the lifetimes and energies for different processes the probability of the excited state T_1 of C_{60} to excite ground state molecular oxygen to the singlet spin states was estimated.

Ab initio calculations were performed to model the interaction of oxygen with fullerenes. It is shown that besides singlet oxygen generation, many other competing processes may happen. Attachment of the whole O_2 molecule is unfavourable. However, the energy of two $C_{60}O$ molecules is lower than two C_{60} and O_2 molecule. It is energetically beneficial to produce atomic oxygen, and under the action of visible light the attachment of oxygen to fullerene is likely to occur. The stability of the electronic structure of the oxidized products $C_{60}O$ and C_{60}-O-C_{60} in the singlet and triplet spin states were calculated on the basis of the pre- atomization of oxygen by photoexcited C_{60} using DFT/B3LYP method.

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