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Excited states, stability and polyradicaloid character of periacenes and modified periacenes

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Abstract

The n-acenes are a class of polyaromatic hydrocarbons (PAHs) composed of linearly condensed benzene rings, resembling a quasi-1D graphene strip with zigzag boundaries. The two-dimensional structures are known as periacenes. The acenes with more than five linear benzene rings, are characterized by having a singlet open shell wave function in the ground state, showing that these systems endue an elevated radical nature. Because of their small band gaps and high charge-carrier mobilities these PAHs have attracted strong interest since they can be used in many applications such as organic semiconductors, in organic light-emitting diodes (OLEDs), in singlet fission processes, in nonlinear optics and as energy storage devices. The characterization of the ground and excited states electronic properties of the pristine and modified structures by means of quantum chemistry methods provide important information to build new materials. However, due to their radicaloid character at least two main conclusions are well-known from the literature: (1) the multireference character of the ground state electronic configuration increases with increasing number of fused benzene rings and (2) doubly excited configurations contribute to the wave functions of the low-lying excited states. Therefore, for a completely satisfactory treatment of the ground and excited states of these systems multireference methodology are preferentially requested. In this presentation, the results of the ground and excited states using multireference methods are reported for different sets of acenes and periacenes, as well as for the modified systems with B/N substitution to carbons, which proved to be a powerful tool to tune the polyradical character of PAHs. As descriptors for the biradicaloid character natural orbital occupations, number of unpaired electrons and singlet/triplet splitting are used. These benchmark data are compared with density functional theory (DFT) results in terms of spin contamination and the fractional occupation number weighted density (FOD). Examples for B/N modification of acenes to find optimal singlet fission compounds will be given.

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