化学学报 1998 56 (11): 1081-1086 ISSN: 0567-7351 CN: 31-1320/06

生物体系中电子转移机理的理论研究Ⅰ:一种计算电子转移 的方法及在螺环化 合物中的应用

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收稿日期 修回日期 网络版发布日期 接受日期

摘要 应用"相应轨道变换"和"广义"密度矩阵的方法,向MOPAC程序包中加入了新的功能,
使其能处理电子转移反应中的部分参数。然后用此程序包中AM1方法对具有螺环结构的分子进行处理,
计算了化合物在不同外加电场强度下的势能面、反应热ΔE,重组能λ及电子转移矩阵元V~A~B,
结果表明,λ,V~A~B受外加电场的影响较小,而ΔE则与之成正比。对标题化合物1
的计算结果也同abinitio法的结果进行了比较,发现其变化趋势完全一致,
这说明本方法在计算电子转移方面是可靠的。与abinitio方法相比,本程序不仅适用于计算较大体系
(如生物大分子),而且还具有速度快,耗时少的优点。

关键词 电子转移反应 电子矩阵元 重组能 AM1方法 偶极矩 总能量 螺环化合物

分类号 0621.16

Theoretical study on the electron transfer in biological systems Ⅰ: A method for calculating the electron transfer and its application in spiro π-electron system

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Abstract We have added a new function to the MOPAC program package by employing the methods of corresponding orbital transformation and generalized density matrix and developed a new program called MOPAC- ET, which can deal with the electron transfer reactions. We have also calculated a molecule with spiro π-electronic structure by use of this method and obtained its potential energy surface, exothermicity ΔE, reorganization energy λ and electron transfer matrix elements V~A~B. The results indicate that V~A~B and λ are not obviously affected by the outer electric field while the ΔE is directly proportional to it. The calculated results of title compound 1 were also compared with that from ab initio method. It was found that our results are consistent with that calculated by ab initio method. This approved the reliability of the program MOPAC-ET. In addition to this, the program has advantages of faster speed and less time consuming over the ab initio methods, thereby can be used to simulate the electron transfer for the large systems such as biological macromolecules.

Key words ELECTRON TRANSFER REACTION ELECTRON MATRIX ELEMENT AM1 METHOD DIPOLE MOMENTS TOTAL ENERGY SPIRO COMPOUNDS

DOI:

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