Density Functional Theory Calculation of Electronic Structure of Fe-N-C, Fe-O-C, Fe-P-C, Fe-S-C Single-Atom Catalysts Systems

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Single atom catalyst (SAC) is an emerging class of catalyst that is able to incorporate the scalability of heterogeneous catalysts and atom economy of homogeneous which exhibit superior catalytic performance in various reaction, i.e., CO oxidation, CO2 conversion, oxygen reduction reaction (ORR), etc. It is challenging to explore electronic structure and catalytic mechanics of SACs when the single metal atom is bonding with different dopants, which influence the catalytic efficiency directly, via experimental methods. In this work, Density Functional Theory (DFT) calculations, an ab initio simulation methods, have been carried out to investigate electronic and catalytic properties of Fe-N-C, Fe-O-C, Fe-P-C, and Fe-S-C with oxo or hydroxyl group. Geometry configurations of Fe-N-C, Fe-O-C, Fe-P-C, Fe-S-C, and corresponding structures bonded with oxo or hydroxyl group are studied. Formation energy of the SAC, oxygen formation energy, and hydrogen atom transfer energy of the four SACs are also studied. In order to investigate electronic structure of the four SACs, I examined the HOMO LUMO gap for each system. Moreover, I plotted the corresponding HOMO and LUMO orbitals of each SACs with oxo or hydroxyl group. Fe-N-C is found to have lower formation energy, higher oxygen formation energy, and higher hydrogen atom transfer energy. The results suggest Fe-N-C model is favorable to synthesis and stable among the four models. My work discloses the microscopic properties of SACs and provide valuable suggestions to experimental scientists. Hence, my catalysts models represent a promising contribution to sustainability and making advances in chemical synthesis.

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