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Kinetics and Mechanism of Proton Transfer in Molten Lithium Carbonate: Insights from Static and Dynamic DFT Studies XUELING LEI, Benedict College, KEVIN HUANG, University of South Carolina, CHANGYONG QIN, Benedict College — Using static and dynamic DFT methods and a cluster model, the mechanism and kinetics of proton transfer in lithium molten carbonate (MC) were investigated. The migration of proton prefers an inter-carbonate pathway with an energy barrier of 8.0 kcal/mol. At TS, a linkage of O—H—O involving O 2p and H 1s orbitals is formed between two carbonate ions. It is noticeable that the solvation of proton in an ionic liquid is beyond the capacity of a simple cluster model and that the FPMD method is more suitable for such a molecular system. Corrections on the calculated energies using an extracted cluster were performed and the results displayed good consistency with the value of 7.6 kcal/mol and 7.8 kcal/mol from experiment and FPMD simulation, respectively. The calculated trajectory of H indicates that proton has a good mobility in MC, while both carbon and oxygen only move slightly to facilitate the proton migration. Small geometric variations were observed on all involved ions, not just on the local structure where the proton transfer occurs, implying a synergetic process. A better description of this synergetic step can be displayed in the Lewis diagram. Overall, the results indicate that the combination of the static and dynamic DFT methods is of great advantages in treating such ionic liquid systems and can improve the reliability of the calculated results.

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