Faced with depleting fossil carbon sources, the search for alternative energy carriers and energy storage possibilities has become an important issue. Nature utilizes carbon dioxide as starting material for storing sun energy in plant hydrocarbons. A similar approach, storing energy from renewable sources in chemical bonds with CO2 as starting material, may lead to partial recycling of CO2 created by human industrial activities. Unfortunately, currently available routes for the transformation of CO2 involve high temperatures and are often not selective. With the development of more sophisticated methods and better software, theoretical studies have become both increasingly widespread and useful. This concept article summarizes theoretical investigations of the current state of the feasibility of CO2 activation with molecular transition metal catalysts, highlighting the most promising reactions of CO2 with olefins to industrially relevant acrylic acid/acylates, and the insertion of CO2 into metal element bonds, particularly for the synthesis of cyclic carbonates and polymers. Rapidly improving computational power and methods help to increase the importance and accuracy of calculations continuously and make computational chemistry a useful tool helping to solve some of the most important questions for the future.