Pressure-Driven C-H Activation through Non-Bonding Anagostic Interactions for Petroleum Hydrocarbon Transformation

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Petroleum hydrocarbon contamination is an outstanding environmental issue that causes damage to ecosystems and public health. Activation of carbon-hydrogen (C-H) bonds is central to the chemical transformations of these hydrocarbons. However, C-H activation remains a challenge due to the bond's nonpolar nature and large dissociation energy. This demands the development of new catalysts. Agostic interactions have been studied for C-H activation but are restricted to early transition metals (TMs), limiting catalyst design. On the other hand, anagostic interactions are predominant in late TM compounds. Enabling C-H activation in anagostic complexes will expand the breadth of catalyst design, but these non-bonding interactions are weak at ambient conditions. It is hypothesized that mechanical pressure can enhance the properties of anagostic interactions, leading to C-H activation. Calculations were performed on two compounds containing anagostic complexes, AEMANI and QANQOM. Raman spectroscopy calculations indicated that for AEMANI, vibrational frequency decreased and bond length increased with pressure, a sign of C-H activation. QANQOM showed the opposite pressure response, indicating that the nature of anagostic interactions depends on many factors. Nuclear magnetic resonance calculations reflected a downfield chemical shift for AEMANI, highlighting that the mechanisms of the metal-CH interaction are fundamentally different from agostic bonds. This insight can help establish TM-catalyzed C-H activation, which will allow engineering of mechanically modulated catalytic systems. This will aid in the design of selective, efficient catalysts, allowing the transformation of toxic petroleum hydrocarbons into functional materials, protecting the environment and public health.

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