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Computational Design of Metal-Exchanged ZSM-5 Catalyst For Propane Dehydrogenation

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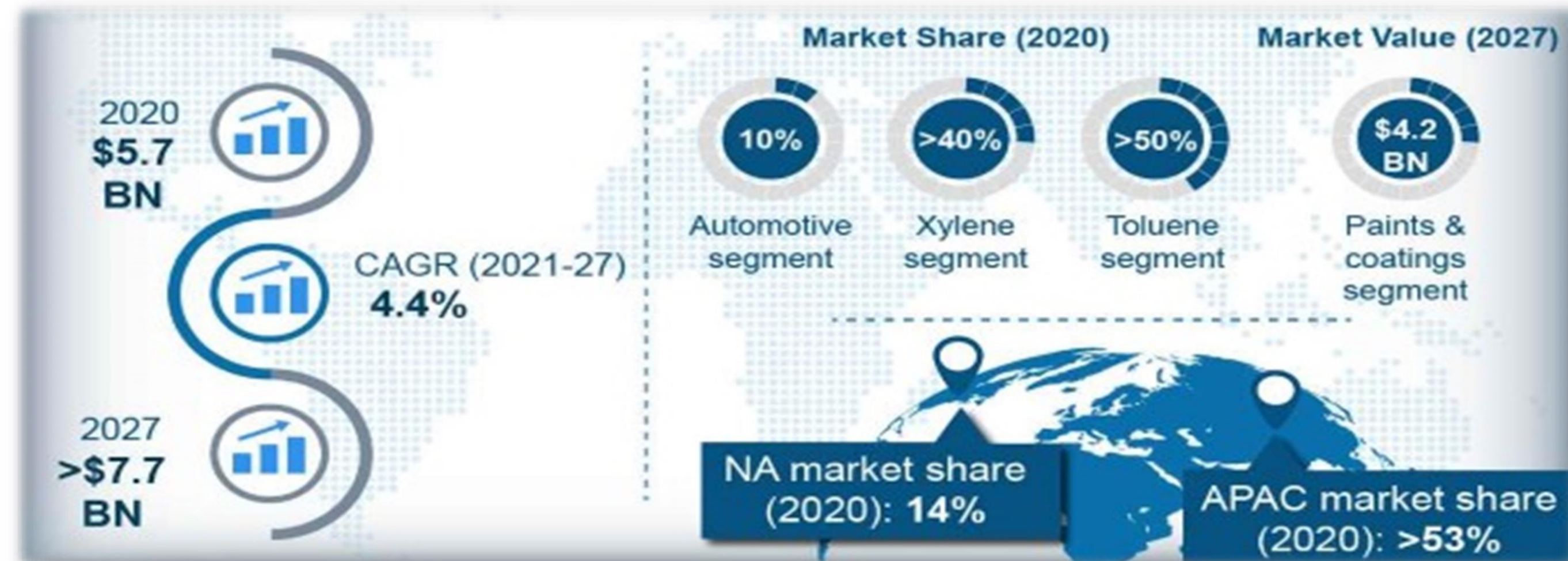
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Introduction

- Dehydrogenation [1,2] is a vital preceding step in the production of aromatics from propane.
- The end-products find application in the petrochemicals [2] and fuel [3] production.



Global Market Insight for Aromatics Solvent Market

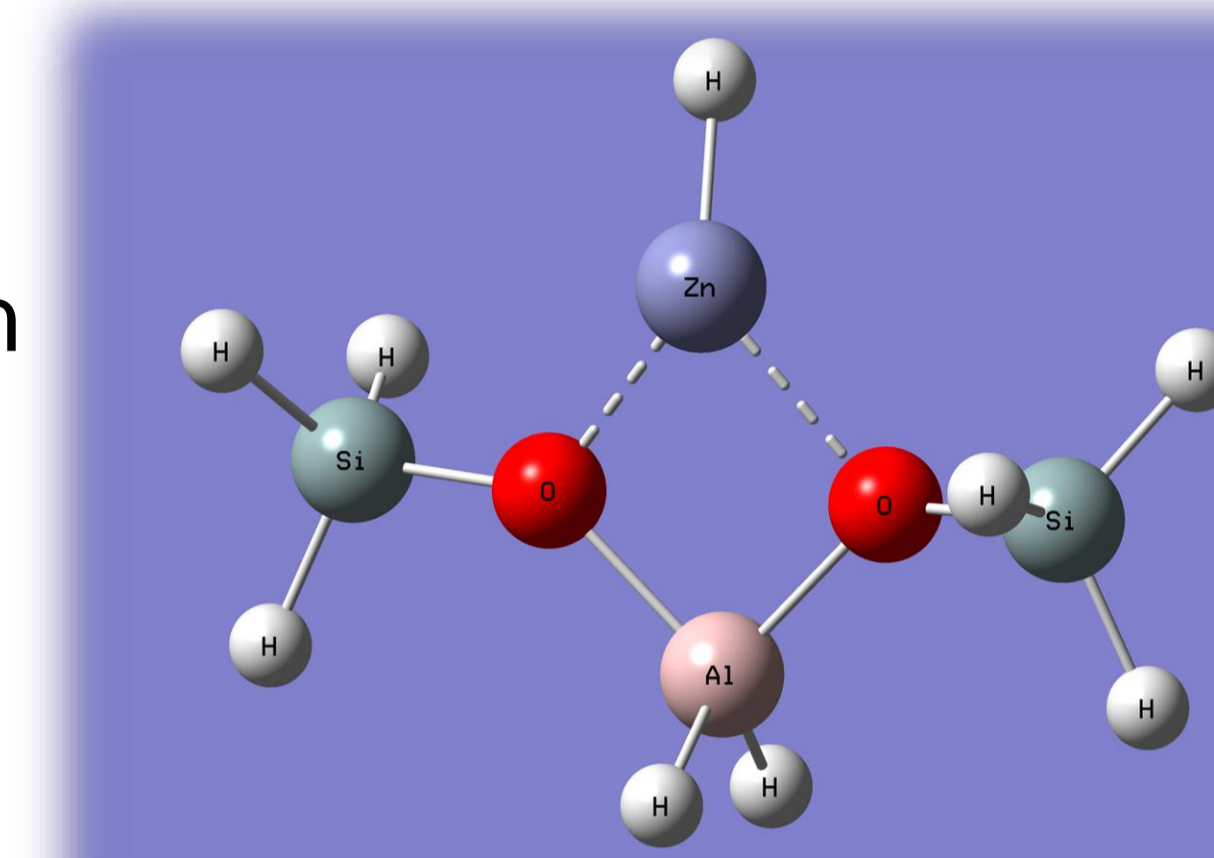
- The demand for aromatics [3] and olefins [4] is significantly rising in the petrochemicals market.
- Existing studies [1-4] have attempted to re-design the catalyst for the purpose of improving catalyst selectivity and production yield.

Objective

We elucidated details of the reaction mechanisms and identified the most kinetically relevant steps in the dehydrogenation of propane over a Ga and Zn hydride exchanged ZSM-5.

Computational Methodology Details

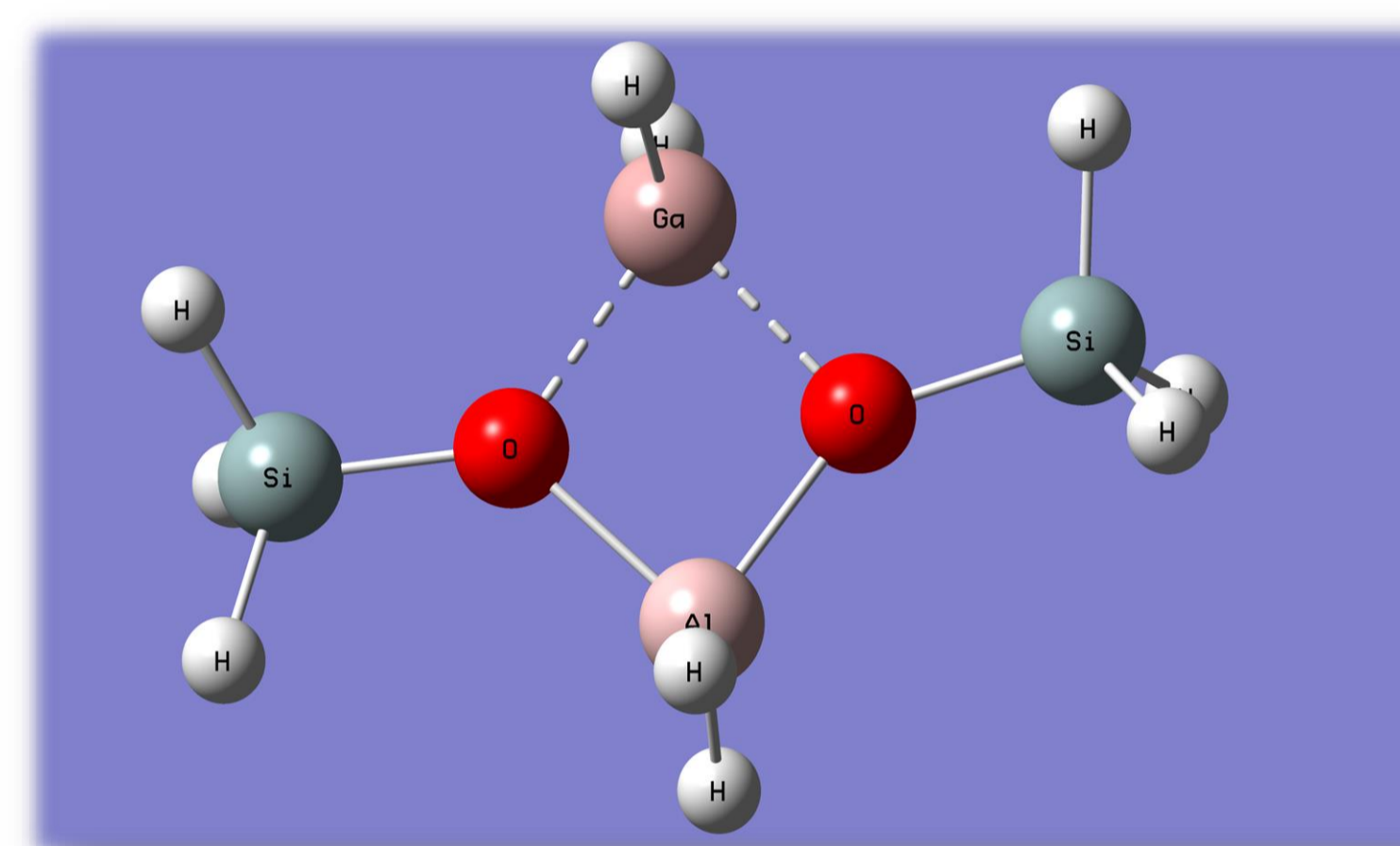
- A similar reaction mechanism to Frash & van Santen [1] was employed in this study. The cluster model (3 tetrahedral atoms, 3T) was used for the ZSM-5.



ZnH [AlZnH₃O₂Si₂]

- The quantum mechanics (QM) calculation was run on Gaussian98. The model theory is used as DFT with a 6-31G(d) basis set.

- The single point energy of the optimized molecule was corrected with a Zero Point Energy, in order to obtain the most stable structure at 0K.



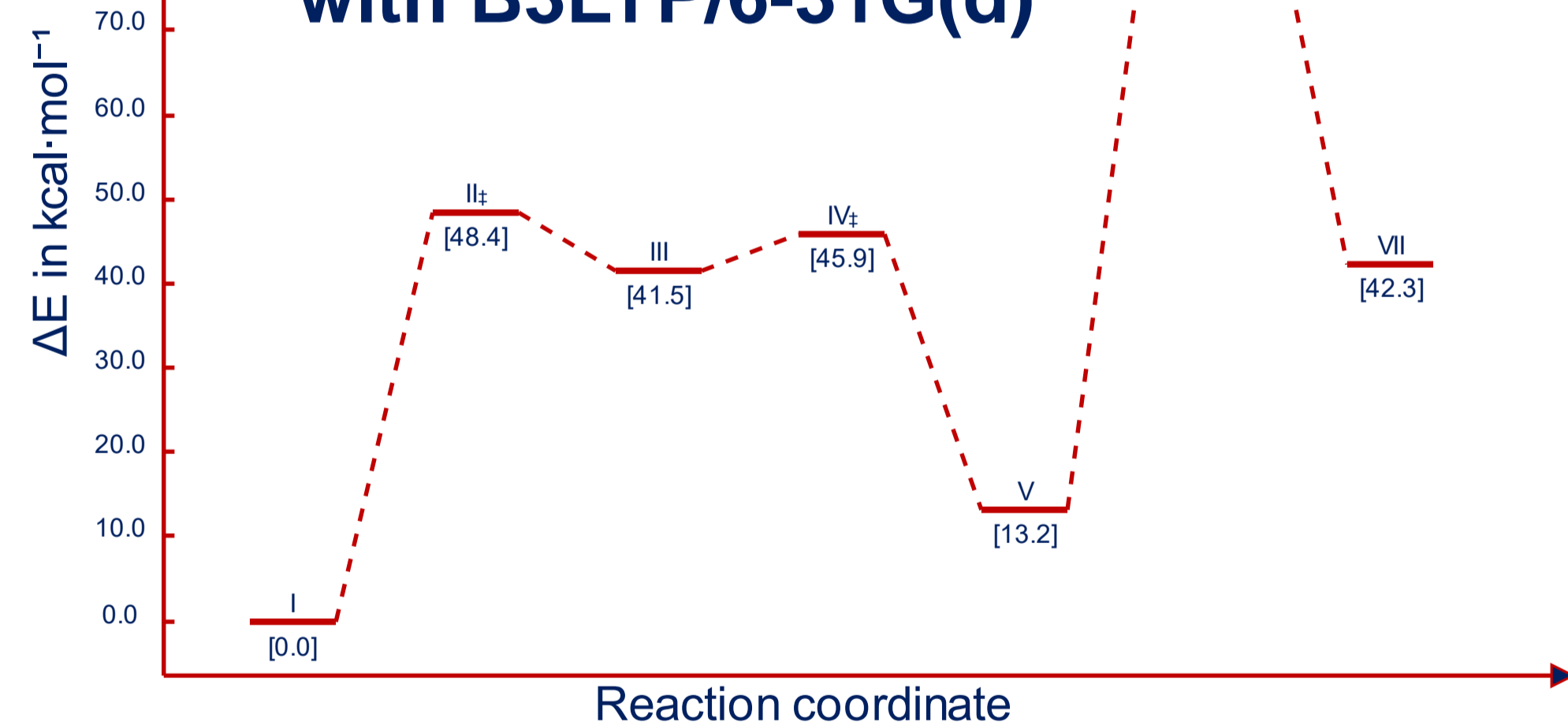
GaH2 [AlGaH₁₀O₂Si₂]

- The transition state (TS) search was done using Quadratic Synchronous Transit 3, QST3.

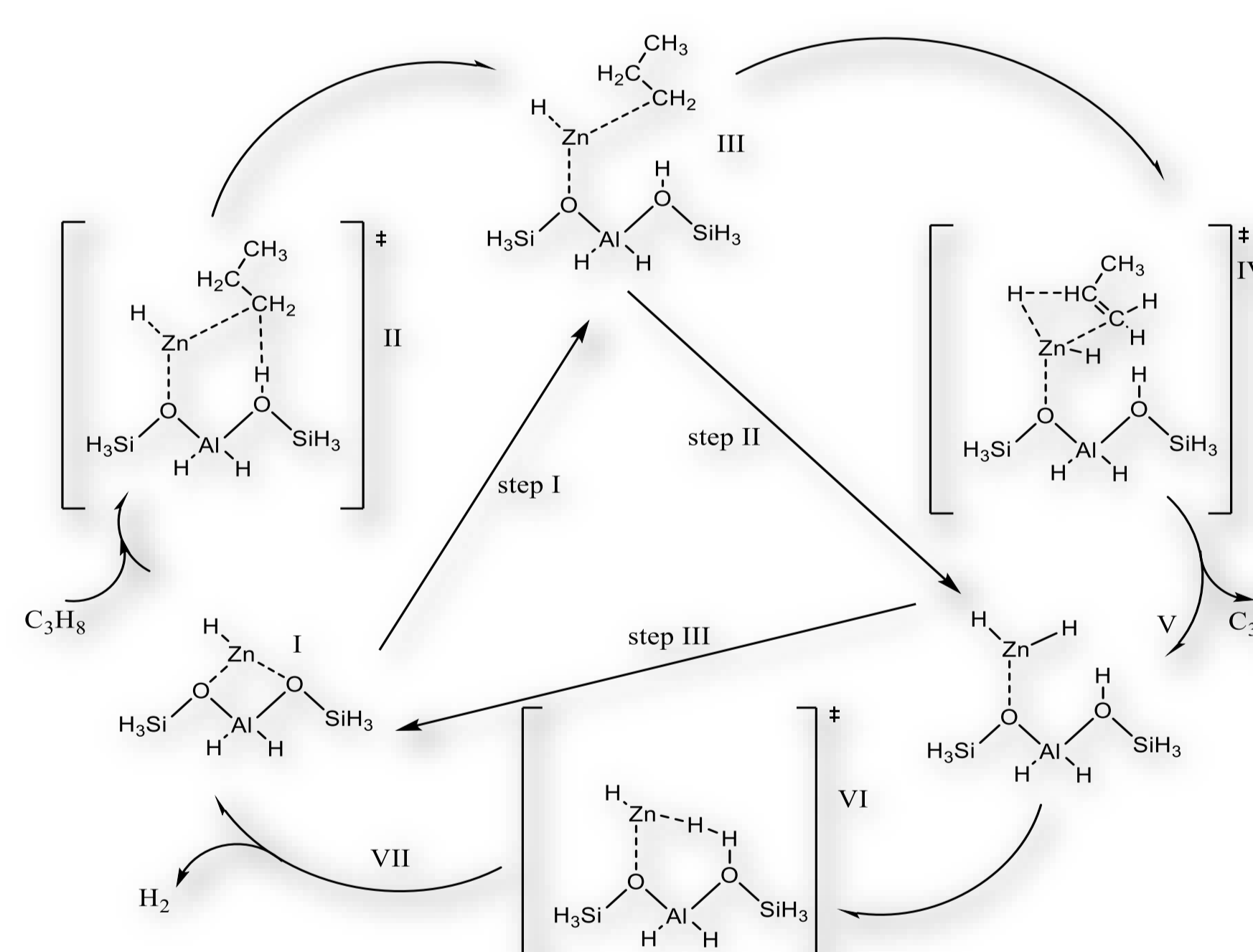
- Intrinsic reaction coordinates were used to validate the TS. And the post-QM calculation was done using MS-Excel & mechaSVG.

Results

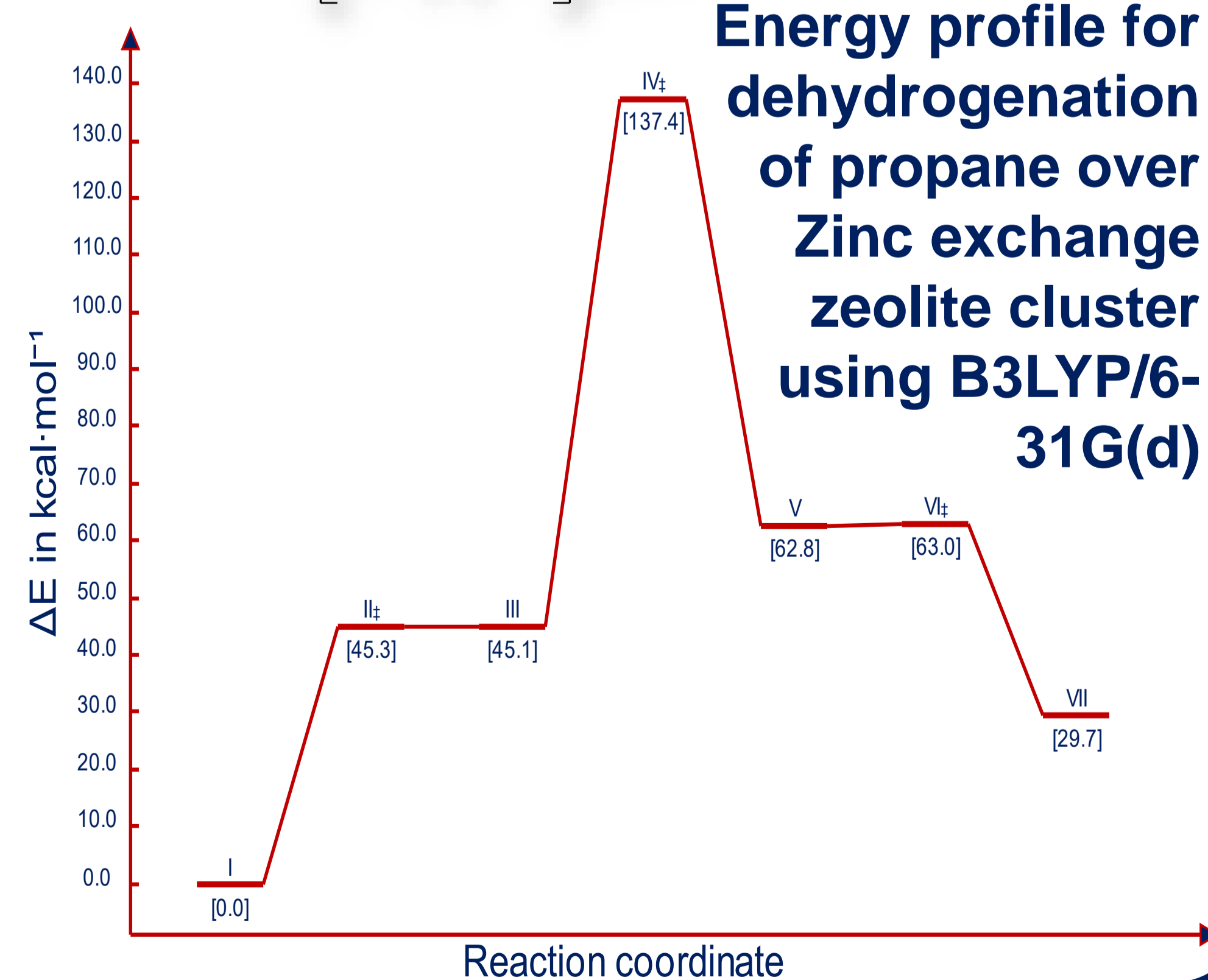
Computational minimum energy pathway for propane dehydrogenation over Gallium dihydride exchange Zeolite cluster with B3LYP/6-31G(d)



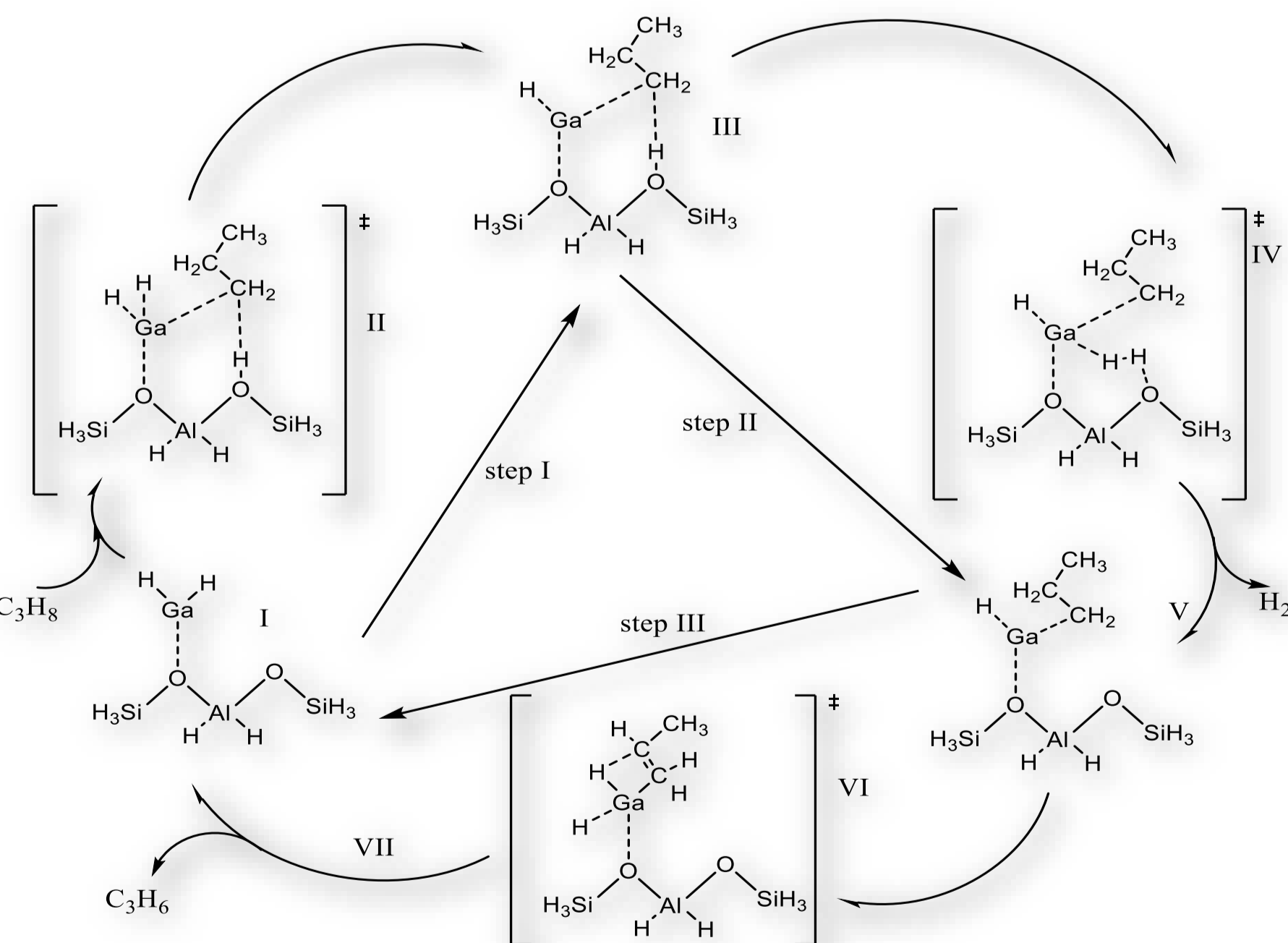
Catalytic cycle for the dehydrogenation of propane on Zinc hydride exchange - Zeolite cluster



Energy profile for dehydrogenation of propane over Zinc exchange zeolite cluster using B3LYP/6-31G(d)



Catalytic cycle for the dehydrogenation of propane on Gallium dihydride exchange - Zeolite cluster



Summary of Findings

- Hydrogen evolved first, followed by propene when GaH₂/T₃ was used, whereas the reverse was the case with ZnH/T₃.

- The activation energy (48.4 kcal/mol) and the heat of reaction (29.1 kcal/mol) of propane dehydrogenation over GaH₂/T₃ using hybrid DFT, B3LYP with 6-31G(d) basis set agrees with the experimental data, 47.8 and 29.7 kcal/mol respectively.

- The computed activation energy and heat of reaction with the ZnH/T₃ catalyst were 45.3 kcal/mol and 29.7 kcal/mol.

- The rate-determining step was observed to change with the reaction conditions, 400-800 K and 101.3 kPa, using transition state theory, TST.

- Findings made agreed with the experimental report [5] and other studies [1,6] where gallium oxide outperforms zinc oxide at a reaction temperature of 600-700 K.

References

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Acknowledgement

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