Supporting Information

The Critical Role of Active $[GaH]^{2+}$ Site in the n-Heptane Dehydrocyclization over Ga/H-ZSM-5 Zeolite

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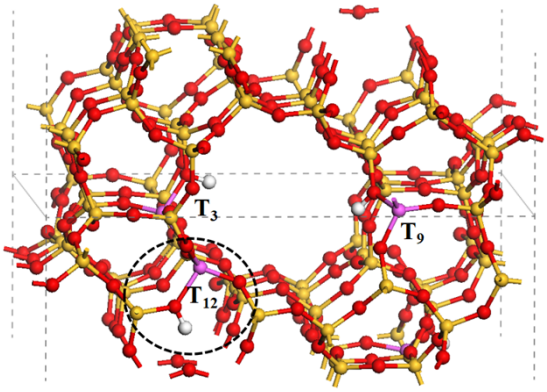


Fig. S1 Optimized structure of H-ZSM-5. The Si, Al, O, and H atoms are represented by yellow, magenta, red and white spheres, respectively.

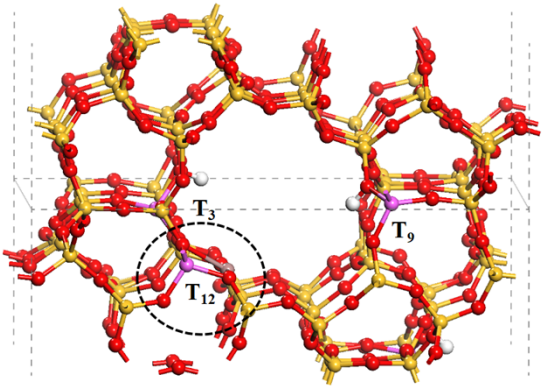


Fig. S2 Optimized structure of Ga/H-ZSM-5. The color scheme in Figure S1 is applied. The Ga atom is represented by the brown sphere.

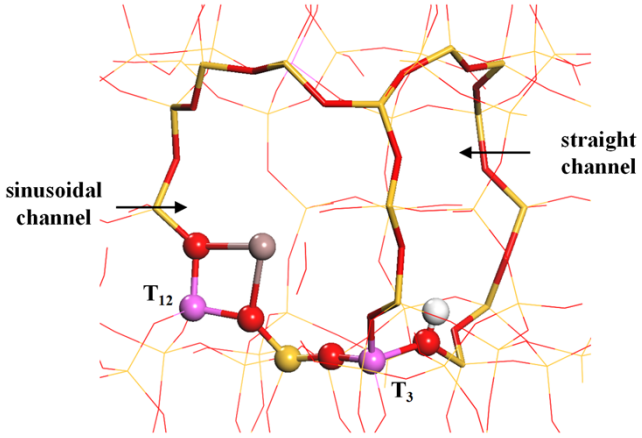


Fig. S3 Optimized structure of Ga/H-ZSM-5. The color scheme in Figure S1 is applied. The Ga atom is represented by the brown sphere.

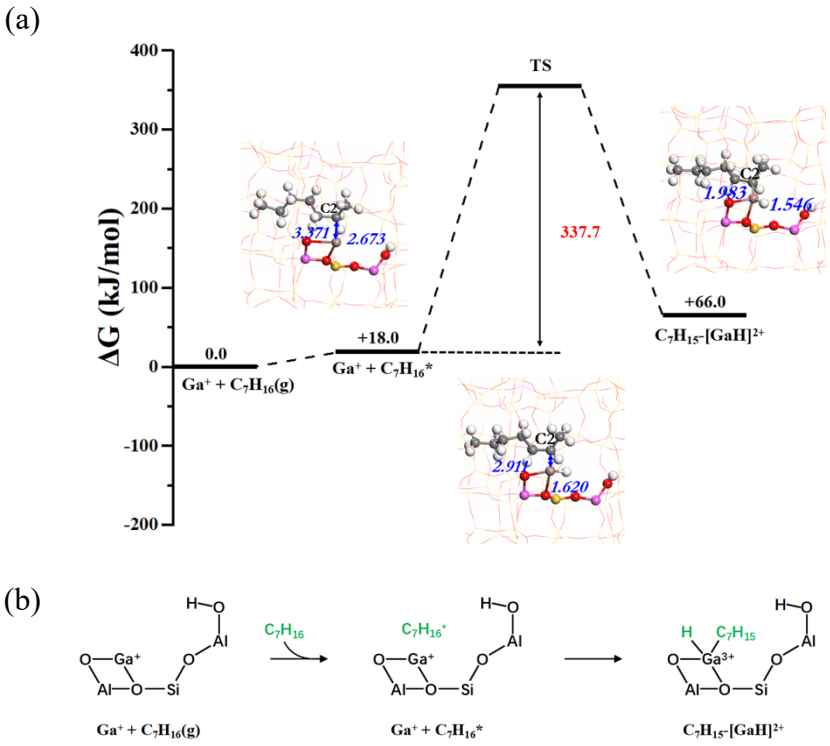


Fig. S4 The dehydrogenation of n-heptane into C_7H_{15}-Ga-H by Ga^+.

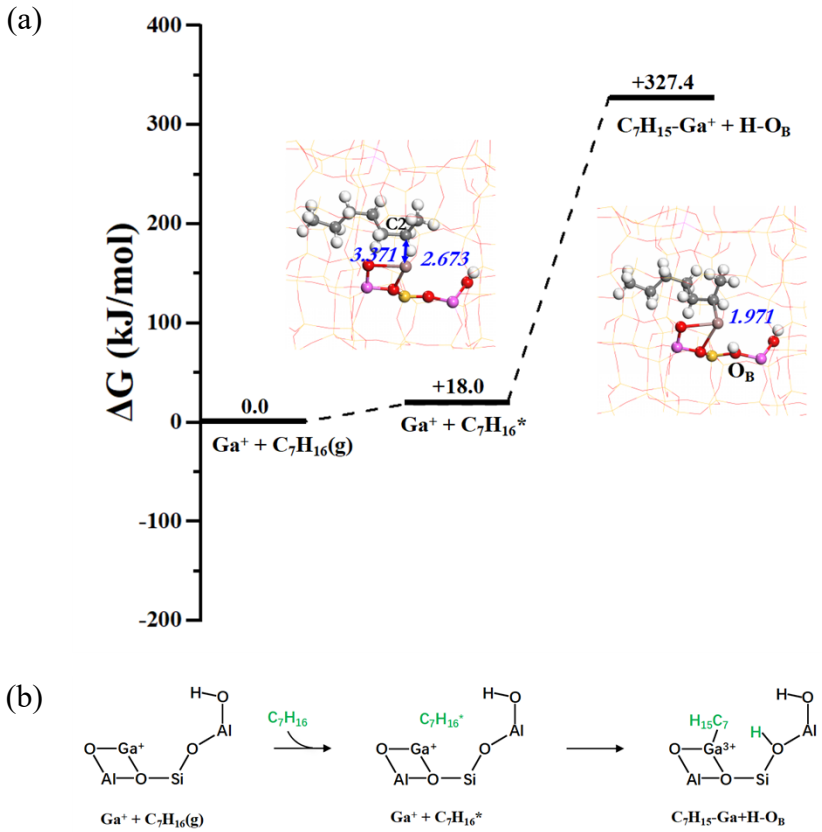


Fig. S5 The dehydrogenation of n-heptane into C_7H_{15}-Ga and H-O_B by Ga^+.

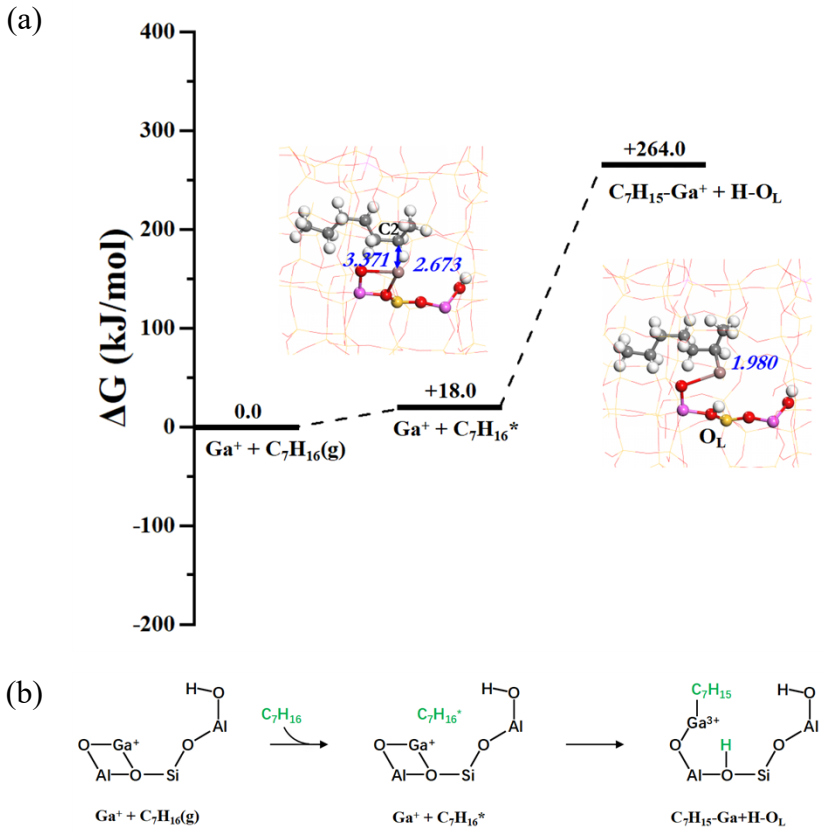


Fig. S6 The dehydrogenation of n-heptane into C_7H_{15}-Ga and H-O_L by Ga^+.

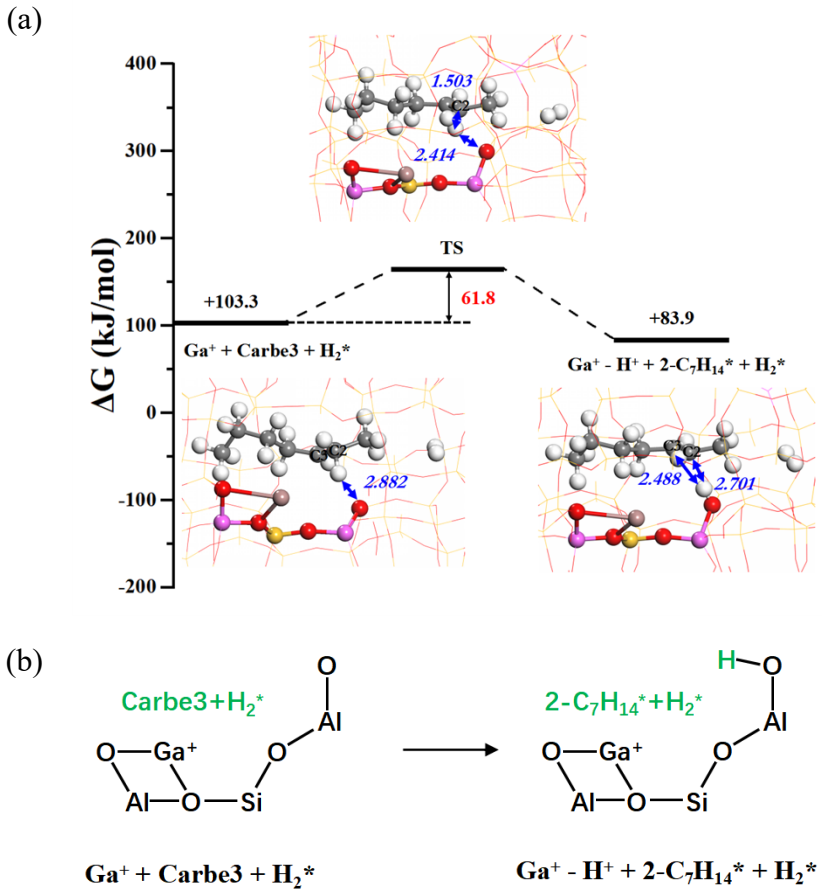


Fig. S7 The dehydrogenation of Carbe3 into 2-C_7H_{14} accompanied by the BAS recovery.

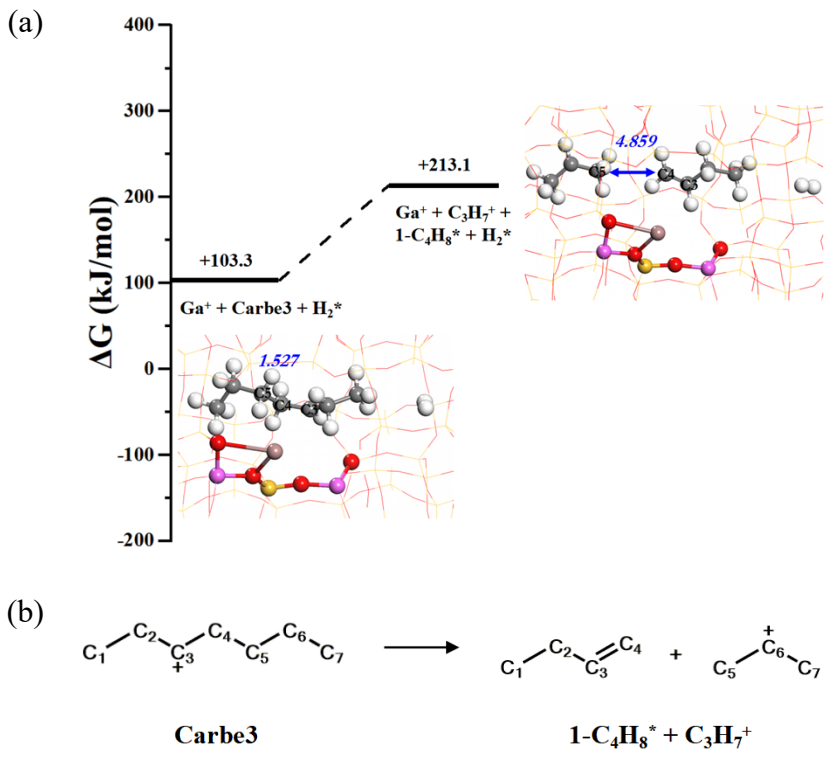
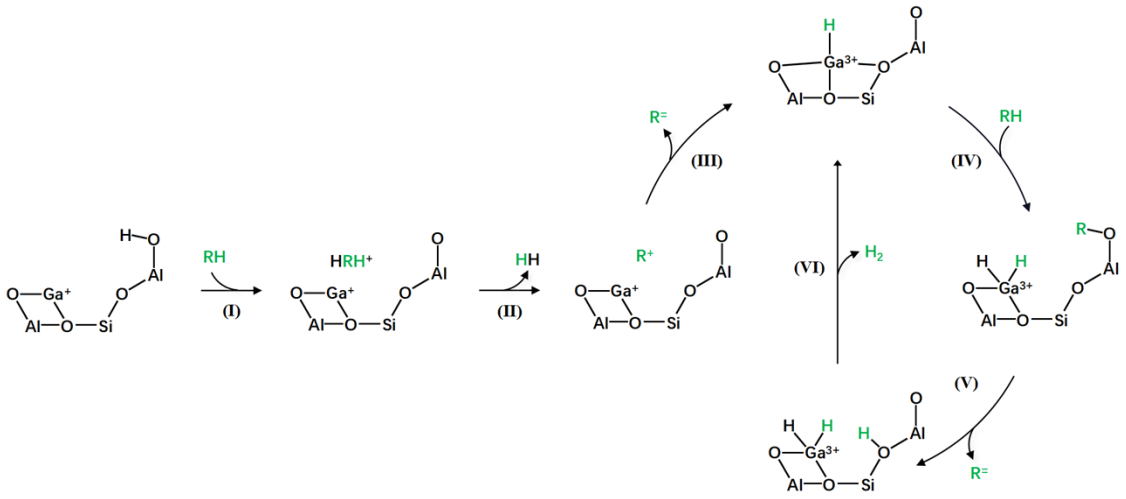


Fig. S8 The C-C bond cracking of Carbe3 into $C_3H_7^+$ and 1-C_4H_8.



Scheme S1. Schematic illustration of generation and regeneration of $[GaH]^{2+}$ in the catalytic cycle of alkane dehydrogenation via a carbenium mechanism.

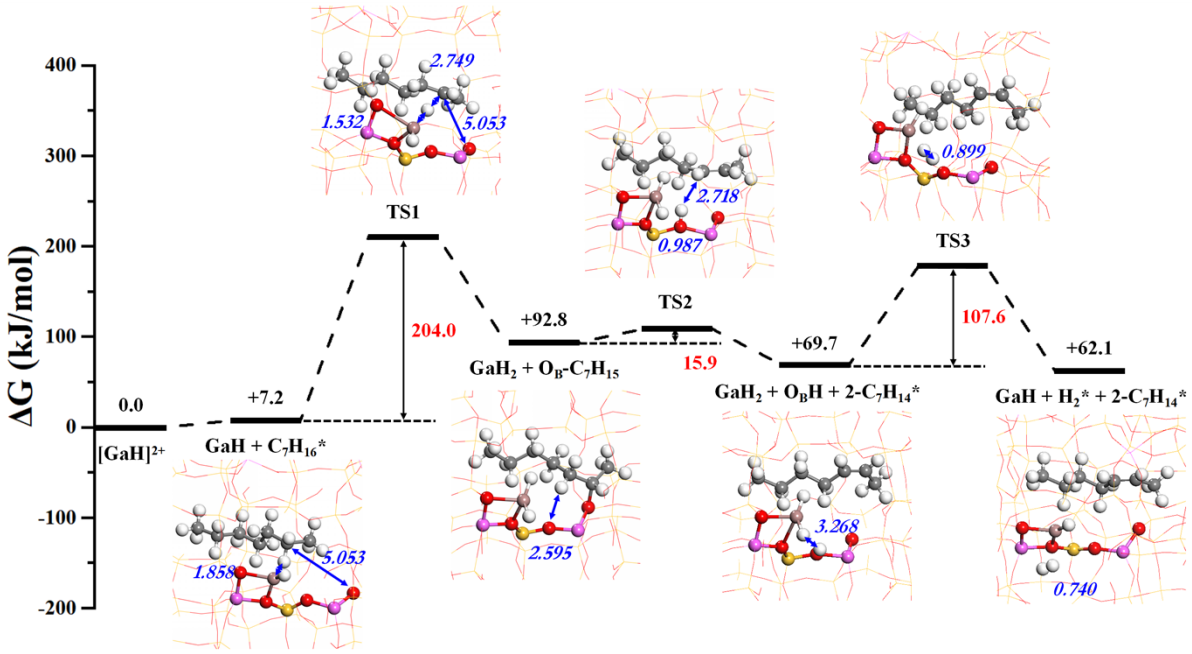


Fig. S9 The Gibbs free energy profile of the n-heptane to 2-heptene process over the $[GaH]^{2+}$ site via a carbenium mechanism.

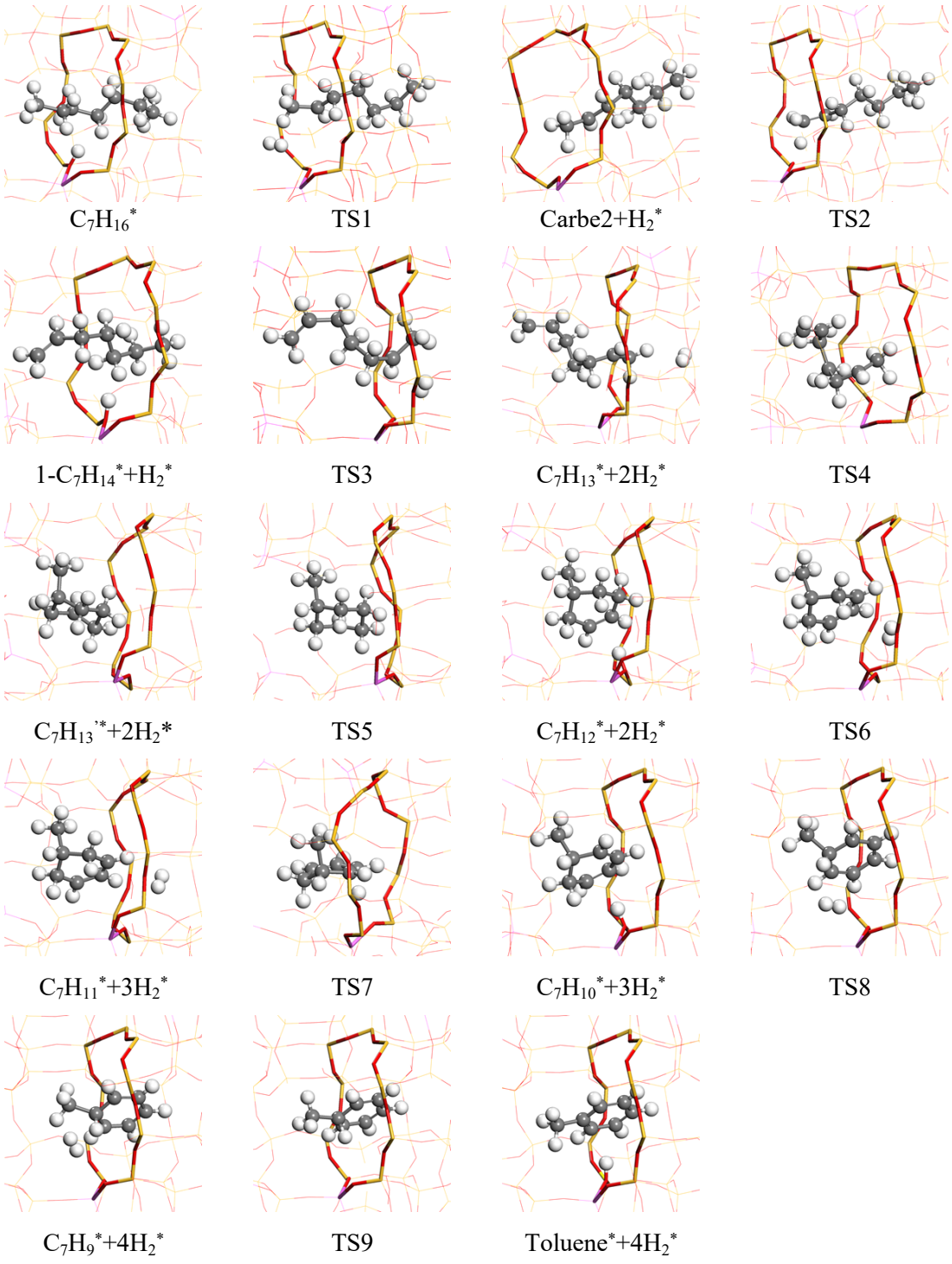


Fig. S10 The corresponding optimized structures of the n-heptane to toluene process over H-ZSM-5.

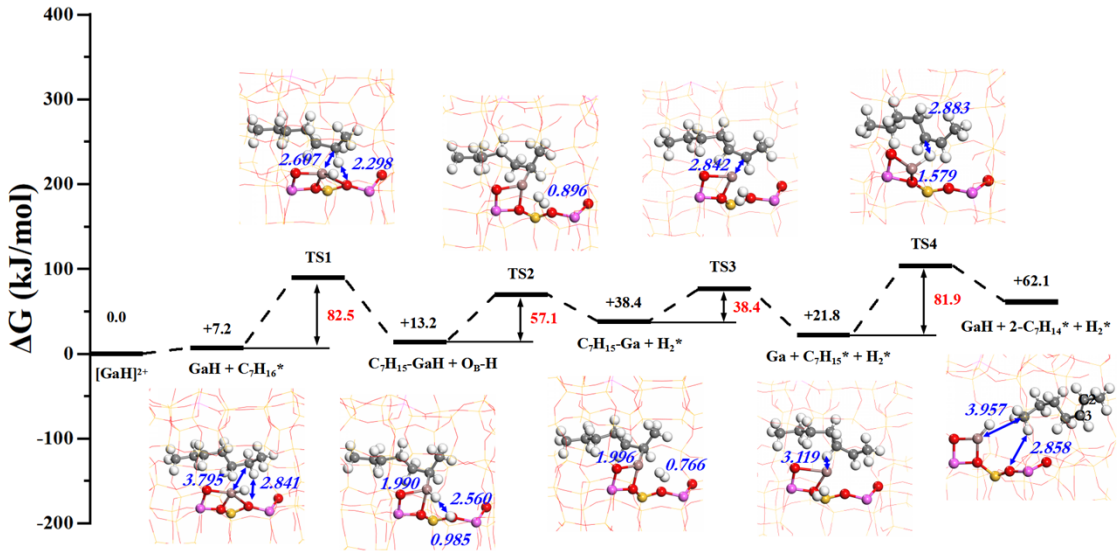


Fig. S11 The Gibbs free energy profile of the n-heptane to 2-heptene process over the $[GaH]^{2+}$ site.

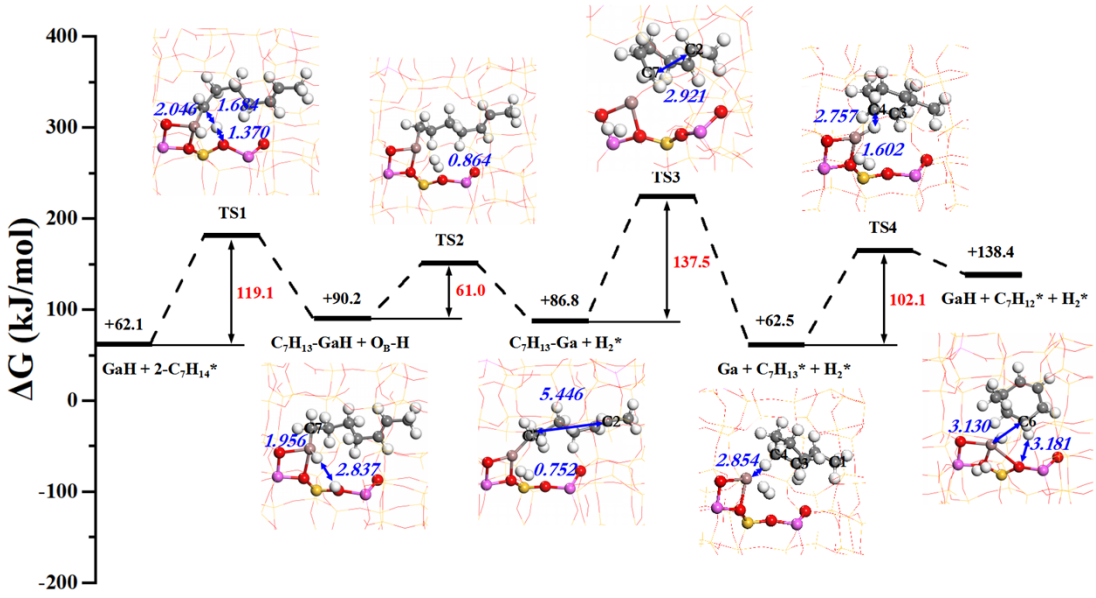


Fig. S12 The Gibbs free energy profile of 2-heptene to methylcyclohexene process over the $[GaH]^{2+}$ site.

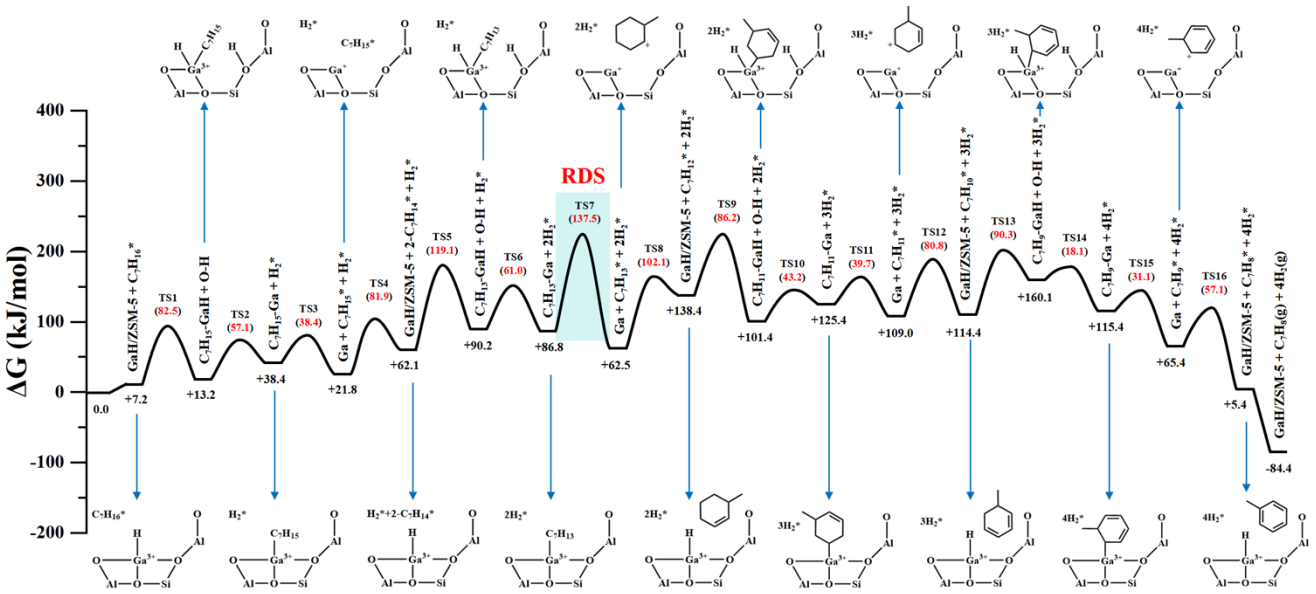


Fig. S13 The Gibbs free energy profiles for the aromatization of n-heptane to toluene via C2-C7 cyclization over the $[GaH]^{2+}$ site.

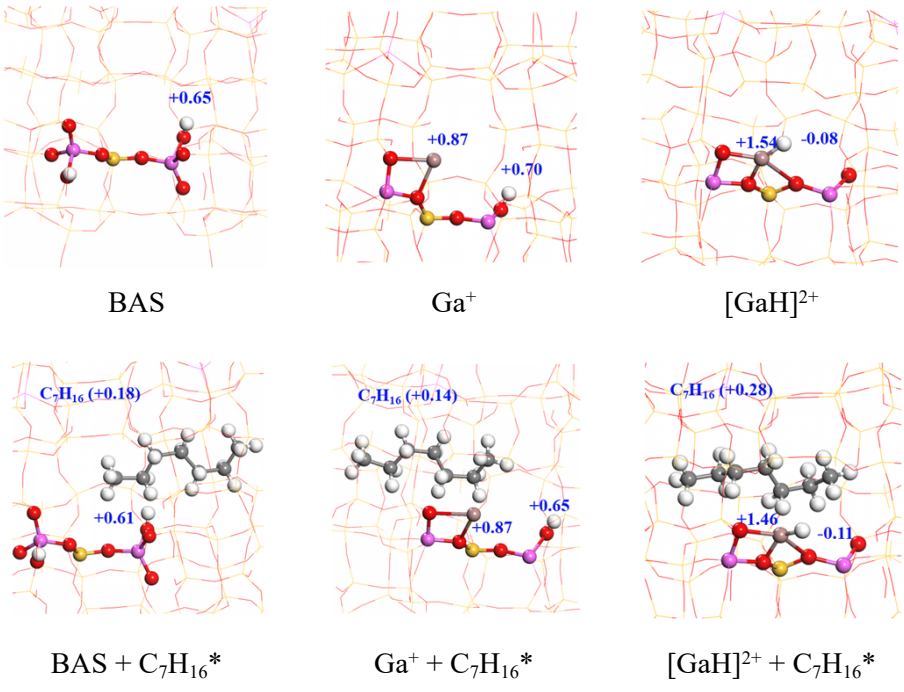
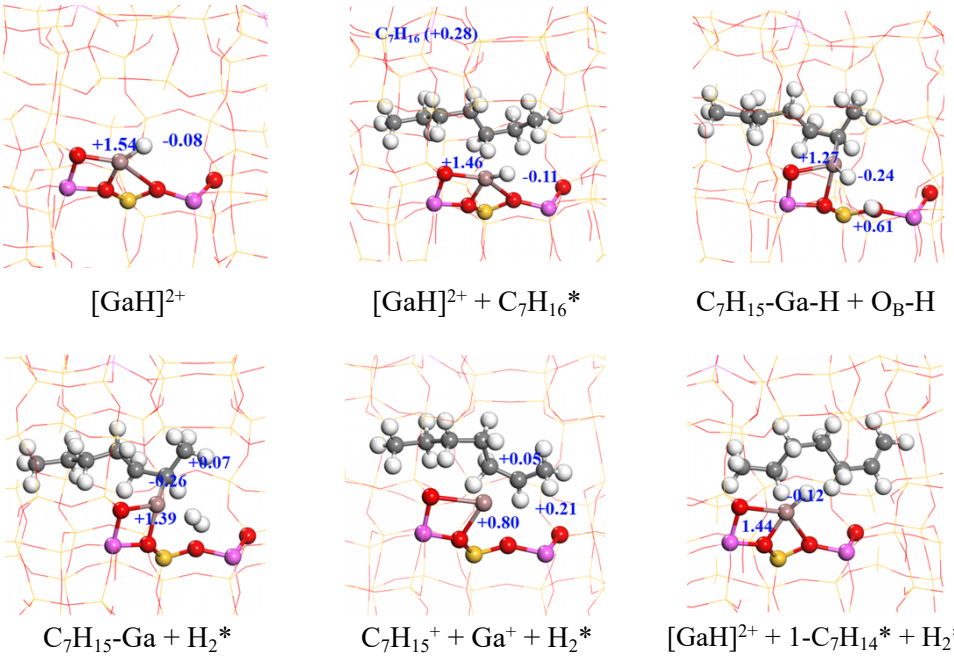


Fig. S14 Bader charges of H_B, Ga, and C_7H_{16}* in (a-b) BAS, (c-d) Ga^+ and (e-f) $[GaH]^{2+}$ site.



$[GaH]^{2+}$ $[GaH]^{2+} + C_7H_{16}$* C_7H_{15}-Ga-H + O_B-H

C_7H_{15}-Ga + H_2* $C_7H_{15}^{+} + Ga^{+} + H_2$* $[GaH]^{2+}$ + 1-C_7H_{14}* + H_2*

Fig. S15 Calculated Bader charges for n-heptane dehydrogenation to 1-heptene and H_2 at the $[GaH]^{2+}$ active site.