

**Electronic Supplementary Information for the paper:  
Machine learning thermodynamic perturbation theory offers accurate activation free  
energies at the RPA level for alkene isomerization in zeolites**

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## SI. PERFORMANCE OF HYBRID FUNCTIONALS

Incorporation of exact-exchange in calculations via hybrid DFT [S1] can be expected to reduce the self-interaction error and thus improve the description of ionic states and reaction barriers [S2]. Such calculations should, in principle, be more affordable than RPA calculations presented in the main text. Unfortunately, the MLPT with PBE+D2 production and hybrid DFT methods B3LYP [S3, S4] and HSE06 [S5–S7] yields unreliable results because of very poor overlap in configuration spaces sampled ( $I_w < 0.001$ , which is far less than the recommended minimum of 0.05 [S8]). We note that a similar observation was made also in our previous work [S9]. To obtain at least a qualitative estimate of the effect of using hybrid DFT on computed activation energies, we performed static calculations and quantified the deviation with respect to PBE+D2. The results, provided in the Fig. S1, suggest that hybrid functionals do not lead to reliable results with respect to the RPA reference level.

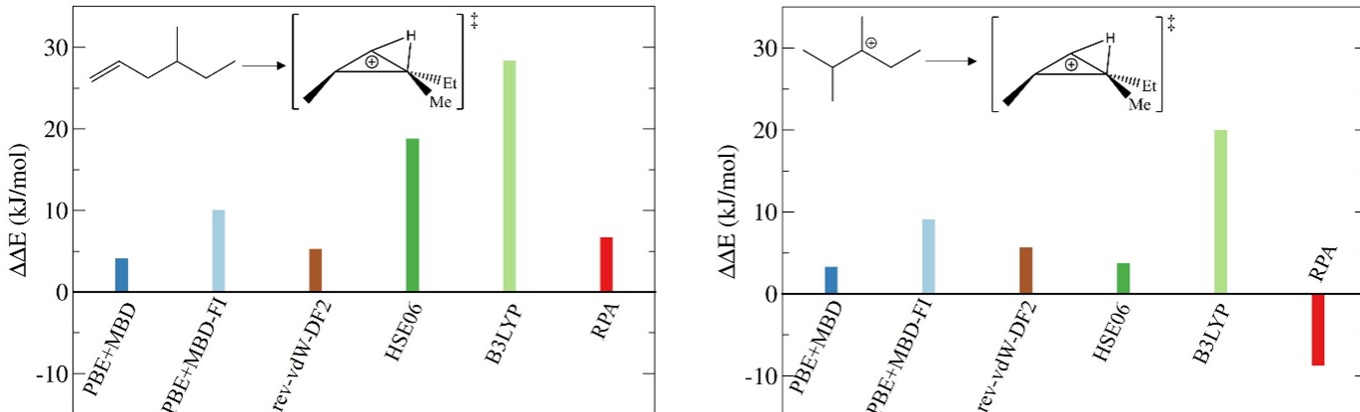


FIG. S1. Differences in potential energy barriers between different electronic structure methods and PBE+D2 determined for the forward (left) and the backward (right) modes of the isomerization of 4-methyl-hex-1-ene in protonated chabazite. Note that differences were determined in single-point calculations using geometries of stationary states relaxed at the PBE+D2 level.

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