Informatics study on Methane Activation on binary alloy

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In the current steam reforming process, the surface of nickel is mainly used as a catalyst due to its high reactivity and low cost. As shown in figure 1, Blaylock et al. used DFT calculations to examine the C-H bond cleavage of methane on the Ni(111) surface.[1] The nickel surface can cleave the strong C-H bond of methane, and further cleaving the C-H bonds produces CH*, the most energetically stable adsorbate of CH_n* (n=1-4). By using heterogeneous catalyst that stabilize CH_3* rather than CH*, it is expected that the intermediate CH_3* will have a longer lifetime and coking will be suppressed. Based on this idea, we explored various binary alloys that enables the direct conversion of methane by a combination of DFT computation[2] and catalyst informatics.

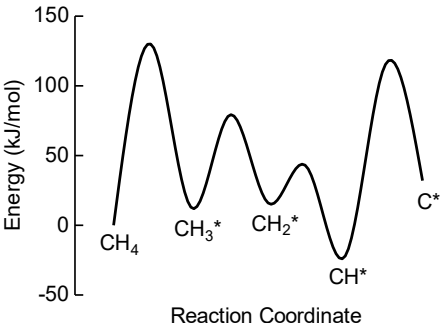

Figure 1. Energy diagram for the C–H bond cleavage of methane on the Ni(111) surface. Data were taken from ref. 1.

The structure of each alloy was obtained from AFLOW[3], the calculation database based on the Vienna ab initio simulation package (VASP). The surface energies of the low index planes of all alloys in the database were calculated by using CASTEP, and the lowest-energy surface was used for subsequent calculations. VASP was used for geometrical optimization of adsorption structures of CH_3* and CH*. We calculated the adsorption energy of CH_3* and CH* on each alloy surface, and evaluated the stability of CH_3* versus CH*. The genetic algorithm-based partial least squares (GA-PLS) regression is used to search effective catalyst.

310 alloys were retrieved from the AFLOW database. The stability of CH_3* against CH* for 88 alloys was calculated. The GA-PLS regression model which predicts the reaction enthalpy ($CH_4 \rightarrow CH_3$*+H*) and the stability of CH_3* versus CH *, was built with 88 training data. The coefficient of determination obtained by LOO-CV was 0.625 for the former and 0.622 for the latter. The model predicted that 26 of the 222 alloys not included in the training dataset would satisfy both conditions. (shown in Figure 2)

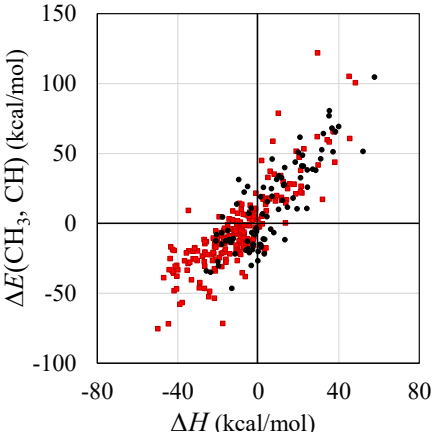

Figure 2: Predicted values of ΔE(CH_3, CH) and ΔH by PLS regression. ● means predicted values and ■ means data values contained in the training dataset.

1) D. W. Blaylock, T. Ogura, W. H. Green, G. J. O. Beran, *J. Phys. Chem. C,* **113**, 4898 (2009). 2) T. Kamachi, et al. *J. Comput. Chem. Jpn.*, **16**, 147 (2017). 3) Automatic FLOW for Materials Discovery Home Page. http://aflowlib.org/