[P4] Theoretical study on methane activation on binary alloys: An informatics approach to catalyst design

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Ni is mainly used in the methane steam reforming processes because of its high reactivity and low cost. The C-H bond cleavage of methane on the Ni(111) surface was investigated by Blaylock et al. with a DFT calculation.[1] The strong C-H bond of methane can be cleaved on the Ni surface, and further cleavage of C-H bonds leads to energetically stable CH* with low activation barriers (see Figure 1). CH* is converted into CO using H_2O in the conventional process. If CH_3* and CH_2* are below CH* on the potential energy surface (PES) of the methane conversion by a new rationally designed catalyst, the direct conversion of methane into methanol, ethylene, etc. can be achieved because the lifetime of the CH_3* and CH_2* intermediates is expected to become longer. Based on this idea, we screened various binary alloys with DFT computations.

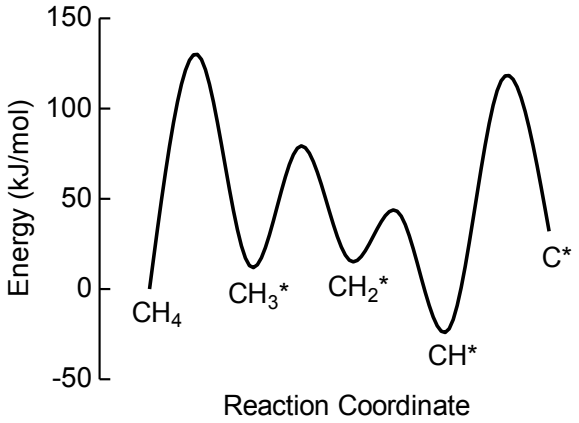

Figure 1. Energy diagram for the C–H bond cleavage of methane on the Ni(111) surface. Data were taken from ref. 1.

The structures of alloys were obtained from AFLOW[2], which is a database of VASP computations. 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. The adsorption structures of CH_3* and CH* on the alloy surfaces were optimized by using VASP. We calculated the adsorption energy of CH_3* and CH* on each alloy surface, and evaluated the stability of CH_3* versus CH*.

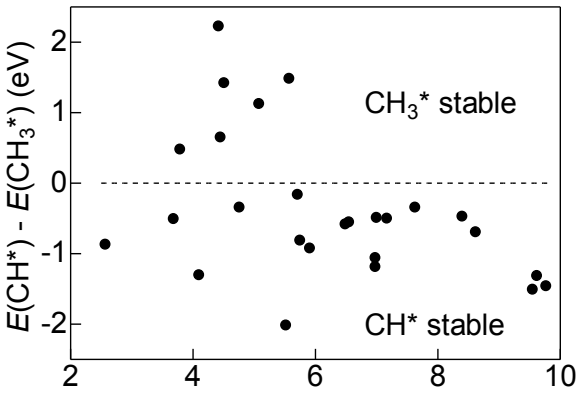

Figure 2. Energy difference between CH* and CH_3* on the stable surface of 26 binary alloys.

We have found 6 binary alloys on which CH_3* is expected to be more stable than CH* (see Figure 2). We are working on the calculation of the thorough energy profile of the reaction on the alloy surfaces.

Bibliography :

[1] D. W. Blaylock; T. Ogura; W. H. Green; G. J. O. Beran ; J. Phys. Chem. C, 113 (2009) 4898.
[2] Automatic FLOW for Materials Discovery Home Page. http://aflowlib.org/