Path sampling simulatoins of CH_4 dissociation on Ni(111)

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1. Purpose

My simulations focus on understanding the CH_4 dissociation mechanism. It is an important intermediate in CH_4 reforming which convert the green-house gasses into syngas.

2. Outline

Using deep neural network potential trained with dataset obtained from density functional theory calculation (DFT), I perform path sampling simulations of CH_4 dissociation on Ni(111).

The path sampling works by performing many short molecular dynamics simulations from several interfaces between reactant and product state. By compiling all the samples, one can obtain the kinetic quantities of the reaction.

In my case, I calculate the CH_4 dissociative sticking probability (S_0) on Ni(111). The calculated S_0 is then compared with experimental estimation to validate our simulation setup.

3. Tools

1. DFT: Quantum ESPRESSO (quantum-espresso.org)
2. Molecular dynamics: LAMMPS (lammps.org)
3. Path sampling: infRETIS (github.com/infretis)

4. Computational resource used

SQUID General Purpose GPU nodes
854.87 node-hour
250 GB memory used
1 node parallelize

5. Results

Snapshots of path sampling results are shown in Fig. 1 where CH_4 initially in the gas phase approximate Ni surface. The interaction with the surface caused one of the the C—H bond to elongate. It is then later dissociated resulting the CH_3 and H.

The calculated S_0 in Fig. 2 has a two order of magnitude difference with experiment, while the activation energy obtained from Arrhenius expression is in very good agreement.

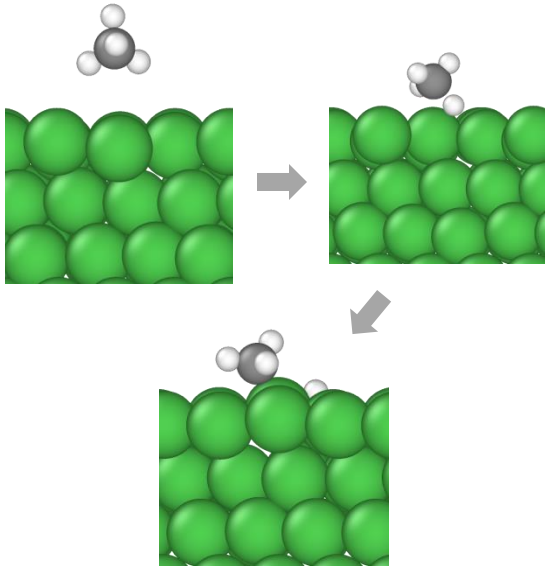

Fig. 1 Snapshoot of pathsampling simulation. Big, medium, and small ball represent Ni, C, and H atoms, respectively.

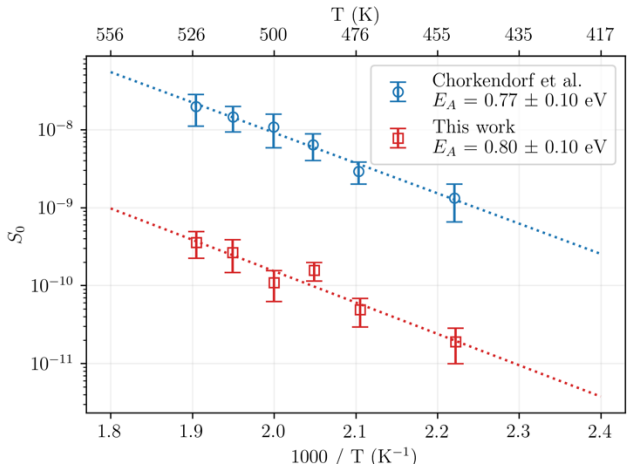

Fig. 2 Dissociation sticking probability (S_0) from experimental estimation (circle) and path sampling calculation in this work (square). The experimental results are obtained from Chorkendorff et al. Surf. Sci. 497, 183 (2002).