

Theoretical Insights into Syngas Catalysis

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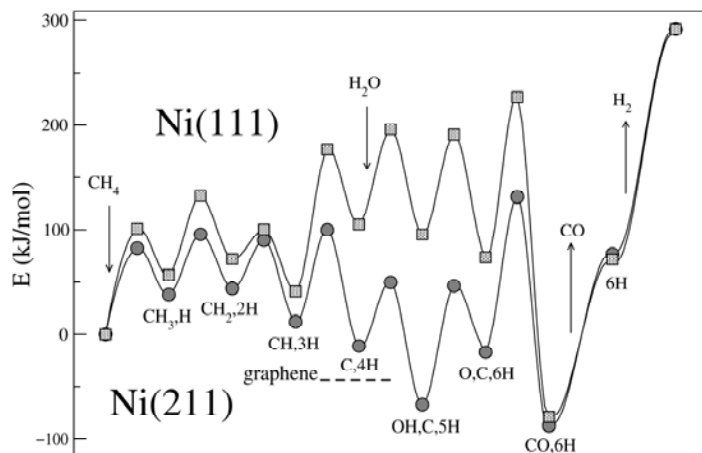
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Electronic structure methods based on density functional theory have reached a level of sophistication where they can be used to describe complete catalytic reactions on transition metal surfaces. This gives an unprecedented insight into these processes, and it allows us to pinpoint the origin of the catalytic activity of a metal in terms of its electronic structure. The new possibilities are illustrated by presenting results for the steam reforming reaction over a number of transition metal surfaces have provided a number of insights into the process.

The full potential energy diagram for the steam reforming reaction, Fig. 1, shows a series of activated processes on Ni surfaces[1]. The stepped Ni(211) surfaces has considerably lower barriers for essentially all the reaction steps, pointing to steps and other open defects as active sites.



Potential energy diagram for methane steam reforming over Ni(111) and (211) surfaces.

The role of the step sites has been confirmed experimentally by showing a scaling between the number of step sites (which is the same as the so-called B₅-sites introduced by v. Hardeveld [2]) and the catalytic activity for a range of catalysts.

The Calculated potential energy diagram also points to step sites as nucleation centers for graphite formation on Ni surfaces, something which can also be observed in detailed *in situ* electron microscopy[3].

The role of promoters and poisons in the steam reforming process will be discussed taking into account the nature of the active sites. Finally, a set of concepts that can be used to rationalize the findings in terms of changes in the electronic properties of metal surfaces[4,5] will be presented.

References

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