【Grant-in-Aid for Scientific Research (S)】

Development of a quantum chemistry based chemical reaction discovery method for heterogeneous catalysis, polymer growth, and two-dimensional material formation

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Purpose and Background of the Research

●Outline of the Research

In this study, we aim to develop a computational method (computer simulation technology) that can predict complex chemical reactions, such as those occurring on supported catalysts, polymer growth, and two-dimensional material formation.

The project leader and his team have pioneered the development of a computational method (the AFIR method) that takes the information in the chemical formula shown in (a) as input, repeatedly calculates the potential energy surface shown in (b) based on density functional theory (DFT) calculations, and obtains the reaction pathway network representation in (c) of the input chemical reaction. By calculating complex reaction pathway network representations, it is possible to identify the kinetically most feasible reaction pathway among the various reaction pathways, and to predict the chemical reaction which is most likely to occur in reality, i.e., the chemical reaction which actually occurs in the flask, from among the many possibilities.

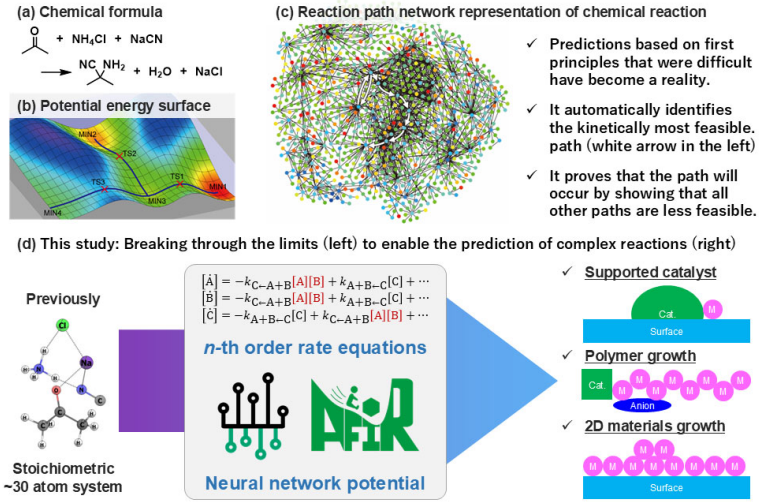

Figure 1: (a) Chemical formula (shown here as an example of the Strecker reaction). (b) Potential energy surface (PES) of a chemical reaction. The energy minimum (MIN) corresponds to the stable structure of the molecule, and the first-order saddle point (TS) at the top of the pass corresponds to the transition state of the chemical reaction. The line connecting MIN-TS-MIN is called the reaction pathway. (c) Reaction path network representation of a chemical reaction (shown here as an example of the Strecker reaction in (a)). Each point (node) on the network corresponds to a MIN on the PES, and each line (edge) corresponds to a reaction pathway on the PES. The white arrows indicate the actual reaction path of the Strecker reaction. (d) Overview of this study. By combining our AFIR method with an algorithm that applies arbitrary order rate equations and the neural network potential technology, we will enable the prediction of chemical reactions of complex systems, such as heterogeneous catalysis, polymer growth, and two-dimensional material formation.

On the other hand, due to the limitations of the algorithm, the reaction pathway network representation was calculated only for stoichiometric reactions, where the molar ratio of all reagents is equal (1:1:1...). In addition, it was necessary to repeat DFT calculations several million times to obtain a reaction pathway network representation. A single DFT calculation takes several seconds to several minutes even for a simple system of about 30 atoms, so long computation times were required for the entire network calculation. Furthermore, for a system with more than 100 atoms, a single DFT calculation takes several hours to several days, so such systems could not be considered as computational targets.

In this study, as shown in Figure 1(d), we will combine the AFIR method with an algorithm that extracts arbitrary order rate equations from reaction path networks and the neural network potential technology that accurately and highly efficiently estimates the results of DFT calculations, thereby overcoming these limitations. This will make it possible to predict chemical reactions in complex systems.

Expected Research Achievements

● Forming Higher-order Rate Equations

Since the rate equations obtained from the reaction pathway network are first-order, they can only handle stoichiometric reactions. Therefore, we will develop an algorithm to extract rate equations of arbitrary order from the reaction pathway network. This will allow simulations that take into account experimental conditions such as concentration and coverage, and will make it possible to apply the method to complex systems such as catalysis and structure growth, where the coverage of the catalyst and the reagent concentration control the reaction efficiency.

● Neural Network Potential Technology

Neural network potential is an ultrafast energy calculation technology that has attracted worldwide attention in recent years. A prediction model is created by learning the relationship between molecular structures and energy values (and the forces acting on atomic nuclei) obtained through DFT calculations, and energy values are predicted using the prediction model without DFT calculations. A well-trained prediction model can predict energy values with almost the same accuracy as DFT calculations in an instant. In other words, by utilizing neural network potential technology, the problem of the computational cost of DFT calculations in complex systems can be greatly alleviated. In this study, we will solve the problem of computational cost in reaction path search using the AFIR method by utilizing the neural network potential technology.

● Target Applications

The application targets are heterogeneous catalysis, polymer growth, and two-dimensional material formation, as shown in the title and Figure 1(d). In small molecule conversion (e.g., steam reforming) using supported nanocatalysts, it has been reported that the reaction is promoted at the catalyst/support interface. In this study, we will clarify the whole picture of the complex catalytic process, including changes in the catalyst structure at the catalyst/support interface. For polymer growth, we will elucidate the branching and tacticity of the polymer chains through accurate kinetic simulations. For two-dimensional material formation, we will elucidate the whole picture, including defect formation and additive effects on the growth rate.

In the second half of this study, based on the applicability established in the first half, we plan to conduct collaborative research with experimental groups who are actually developing cutting-edge chemical reactions.

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