Project Title:

Computational Study on Rare Earth Metal Complexes and Their Catalytic Reactions

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1. Background and purpose of the project

Chemical conversions driven by catalysts are essential to modern society. The ability to predict and modify the reaction rout and rate-determining steps in chemical reactions would be a boon in designing better catalysts. Technical innovations in computer simulations bring that goal closer. Our research is focused on the fundamental study of molecular design for rare earth metal catalysts. As a powerful tool, quantum chemical theory planted into available programs was used to investigate the relationship between electronic structure and properties and hence predict and design new molecules. Since it is hard for experiment to do the same, static quantum chemical calculation and molecular dynamics simulation play an important role in this field.

Noteworthy, hou and his colleagues in RIKEN have recently discovered a serial of novel rare-earth-metal complex catalysts and found that they are highly active and selective to polymerize alkenes. However, the mechanistic aspect is extremely difficult to study by experimental approaches because of high reactivity of the catalyst. With the help of RIKEN Supercomputer System, we may conduct such mechanistic studies and further provide useful information for catalyst design.

2. Specific usage status of the system and calculation method

Only part of the specified computational time was used. The density functional theory method implanted in Gaussian program was mainly utilized.

3. Results

In this project, we plan to computationally investigate mononuclear, binuclear, and polynuclear rare earth complex system. At this stage, we have investigated a mononuclear Sc complex and its catalytic reaction, and positive results have been obtained. In particular, for the first time, we found that the origin of a Sc catalyst system having higher activity toward olefin polymerization is the ease of generating true catalytic species rather than significant lower reaction energy barrier for olefin insertions in comparison with Y analogues.

4. Conclusion

The results obtained in the last fiscal year demonstrate that it is promising to study the project further by using the theoretical method selected. In view of this point, we hope to continue to use RICC for the same project in this fiscal year.

5. If you wish to extend your account, provide usage situation.

We have almost complete calculations of a reaction system catalyzed by a mononuclear rare earth metal complex. Parts of these calculations were carried out on RICC. Due to low speed for file transfer from RICC to local PC, the other parts of the calculations were performed by the computers outside Riken.

6. If no research achievement was made, specify the reason.

At this moment, we have no publications concerning with the usage of RICC. This is partially because the time of less than one year is not enough to publish systematic results. In

addition, although the computational performance is quite good for the current project, the rather slow transfer of resulting files from RICC to our local PC hampered research process.