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研究課題名（和文）新しい炭素および有機金属フレームワーク材料に関するＤＦＴ計算

研究課題名（英文）Theoretical DFT Study on New Carbon K4 and Metal-Organic Framework Structures

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研究成果の概要（和文）：クラスタ・アプローチの中でIRMOF構造及び新たな炭素K4をスクリーニングしてきたが、様々な吸着相互作用を考案するために理論的なDFTとポストハートリーフォック法が更に適応されました。これらはもっとも人気のあるDFT及びポスト・ハートリーフォックレベルtwo-layer ONIOMアプローチ等で計算されました。これらの材料の中で最もよい構造のものとして選ばれたものは、ガス分子との相互作用を模倣することに応用することができる。

研究成果の概要（英文）：The DFT calculations clearly show that cluster A (C46H28) and cluster B (C144H62) mimicking 3C-3D carbon K4 structures have partially metallic character. A recipe has been proposed to overcome an intrinsic instability of these structures originating from the removal of symmetry where the selected side-end C-H groups are replaced by fixation with more rigid silica or zeolite fragments. The overall decorated structure thus obtained can be a good candidate for hydrogen storage applications due to its huge surface area and large void spaces. The most promising structures for IRMOF materials are obtained by replacement of Be-based metal-oxide connector parts with more complex cubic Mg-Ti oxide parts with an increased unit-cell volume size and the basicity of oxide centres that make them as efficient and potential material for hydrogen storage and related other applications. The binding energy of molecular hydrogen were estimated at two-layer ONIOM (MP2/6-31G* : HF/3-21G) level.

研究分野：理論化学

キーワード：DFT study　Carbon K4　Metal-Organic Framework　Cluster calculations

１．研究開始当初の背景

A remarkable mathematical basis has been reported for the existence of the carbon K4 crystal with its three-coordinated and three-dimensional (3C-3D) structure. This led to the question of the possible existence or the possibility of synthesizing this structure from atoms of any element. A couple of theoretical papers have been then reported. However, they led to drastically contrasting result, the latter one has ruled out even the possibility or the existence of the carbon K4 structure. As a result, the overall question on K4 crystal is left open, representing a highly attractive challenge.

２．研究の目的

The present research is aimed to formulate and apply cluster quantum chemical modeling to accurately and exhaustively investigate the structure and chemical activity of a new metallic carbon K4 core-shell crystal and highly modified isoreticular metal-organic framework (IRMOF) structures. The goal included also the theoretical design of new energetic materials on the basis of the two that would have high efficiency for hydrogen storage, catalysis and for other related applications.

３．研究の方法

Our systematic cluster quantum chemical considerations have been directly extended for the modified carbon K4 crystal structures and IRMOF ones. Our goal was to create and characterize the potentially new materials with the carbon K4 crystal structure motif as well as highly modified IRMOF materials to enhance their applicability as gas (hydrogen) storage or gas separation applications and in catalysis.

４．研究成果

The DFT calculations clearly show that cluster A (C46H28) and cluster B (C144H62) mimicking 3C-3D carbon K4 structures have partially metallic character. A recipe has been then proposed to overcome an intrinsic instability of these structures originating from the removal of symmetry where the selected side-end C-H groups are replaced by fixation with more rigid silica or zeolite fragments. The overall decorated structure thus obtained can be a good candidate for hydrogen storage applications due to its huge surface area and large void spaces. The most promising structures for IRMOF materials are obtained by replacement of Be-based metal-oxide connector parts with more complex cubic Mg-Ti oxide parts with an increased unit-cell volume size and the basicity of oxide centers that make them as efficient and potential materials for hydrogen storage and related other applications. The binding energy of molecular hydrogen were estimated at two-layer ONIOM (MP2/6-31G* : HF/3-21G) level.

５．主な発表論文等

（研究代表者、研究分担者及び連携研究者には下線）

〔雑誌論文〕（計　1　件）

① N.U. Zhanpeisov. Theoretical DFT Study on Structure and Chemical Activity of New Carbon K4 Clusters. *Res. Chem. Intermed.* 39 (2013) 2141-2148.

〔学会発表〕（計　14　件）

① N.U. Zhanpeisov. Theoretical Insights into the Structure and Chemical Activity of Complex Modified Catalysts. The 7th Asia-Pacific Conference on Theoretical Computational Chemistry (7th APCTCC), Kaohsiung, Taiwan. January 25-28, 2016 (Invited Lecture)

② N.U. Zhanpeisov. Structure and Chemical Activity of Complex Modified Catalysts. The 10th International Conference of Asian Consortium on Computational Materials Science (10th ACCMS-VO), Sendai, Japan. November 1-3, 2015 (Invited Lecture)

③ N.U. Zhanpeisov. Structure and Chemical Activity of Complex Modified Catalysts. A Theoretical DFT Study. The 6th Japan-Checj-Slovak International Symposium on Theoretical Chemistry (6th JCS), Smolenice, Slovakia. October 11-15, 2015 (Invited Lecture)

④ N.U. Zhanpeisov. Theoretical DFT Study on Structure and Chemical Activity of Complex Modified Catalysts. The International Conference on Functional Materials for Frontier

Energy Issues (ICFMFEI 2015), Novosibirsk, Russia. October 1-5, 2015 (Invited Lecture)

⑤ N.U. Zhanpeisov. Cluster Approach to New Materials Design. International Catalysis Symposium, Osaka Prefecture University, Osaka, Japan. June 27th, 2015 (Invited Lecture)

⑥ N.U. Zhanpeisov. Modified Metal-Organic Framework and Metallic Carbon K4 Structures. A Theoretical DFT Study. The 15th International Congress of Quantum Chemistry (15th ICQC), Beijing, China. June 8-13, 2015. (Poster)

⑦ N.U. Zhanpeisov. Computational New Materials Design Based on Cluster Approach. The Joint Session of Physics, Chemistry and Engineering of Azerbaijan National Academy of Sciences. March 5-6, 2015, Baku, Azerbaijan.

⑧ N.U. Zhanpeisov. Theoretical DFT Study on Structure and Chemical Activity of Complex Modified Structures. The 9th Asian Consortium on Computational Materials Science – Virtual Organization (ACCMS-VO9). December 20-22, 2014, Okinawa, Japan (Invited talk).

⑨ N.U. Zhanpeisov. Theoretical DFT Study on New Carbon K4 and Metal-Organic Framework Structures. The 50th Symposium on Theoretical Chemistry, September 14-18, 2014, Vienna, Austria. (Oral talk)

⑩ N.U. Zhanpeisov. Modified New Carbon K4 and Metal-Organic Framework Structures: A Theoretical DFT Study. The Asian Consortium on Computational Materials Science – Working Group Meeting – 2014 (ACCMS-WGM-2014). June 4-7, 2014, Astana, Kazakhstan. (Invited talk).

⑪ N.U. Zhanpeisov. New Materials Design Based on DFT Cluster Approach. The Symposium "Toward to Future Chemistry Based on Quantum Chemistry". February 11th, 2014, Kyoto, Japan. (Invited talk).

⑫ N.U. Zhanpeisov. Theoretical DFT Study on New Carbon K4 and Metal-Organic Framework Structures. The 5th JCS International Symposium on Theoretical Chemistry. December 2-6, 2013, Nara, Japan. (Invited talk)

⑬ N.U. Zhanpeisov. Modified New Carbon K4 and Metal-Organic Framework Structures. A Theoretical DFT Study. The 8th Congress of the International Society of Theoretical Chemical Physics. August 25-31, 2013, Budapest, Hungary (Poster)

⑭ N.U. Zhanpeisov. Theoretical DFT Study on New Carbon K4 and Metal-Organic Framework Structures. The 7th Conference of the Asian Consortium on Computational Materials Science (ACCMS7). July 23-28, 2013, Nakhon Ratchasima, Thailand. (Oral talk).

〔図書〕（計　0　件）

〔産業財産権〕
○出願状況（計　0　件）

名称：
発明者：
権利者：
種類：
番号：
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○取得状況（計　0　件）

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〔その他〕
ホームページ等

６．研究組織

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