別紙様式３

学業及び研究等の進捗状況等報告書
Report of Research Progress and Future Research Plan

氏名 Name	CHEN Pengru	学　年 Year	D3
所属学院等名 Graduate School	Graduate School of Chemical Sciences and Engineering		
所 属 専 攻 名 Major	Molecular Chemistry and Engineering Course		
所属研究室等名 Laboratory	Catalytic Transformation (Fukuoka Lab)		
指導教員職・氏名 Supervisor	Prof. Atsushi Fukuoka		

１．研究テーマ名 Research theme

Carbon-catalyzed hydrolysis of cellulose to cello-oligosaccharides

２．研究等の進捗状況等 Research progress, etc.

研究の概要、独創性、状況等を含めて具体的に記入のこと。
※研究成果の発表･公表実績がある場合については学会名、掲載紙等の情報を含め詳細を記載すること
In detail, including the outline, originality and so on.
*Please state the name of academic conferences, journals or transactions if you have presented your research or your research was published.

- Summary

Cellulose, a polymer composed of repeated anhydro-glucose units linked by β-1,4-glycosidic bonds, is the most abundant component in lignocellulosic biomass. Cello-oligosaccharides are short chain linear polymers that can be produced by partial depolymerization of cellulose. They exhibit biological activity that can benefit the growth and health of plants, animals, and humans, and thus provide multiple benefits to healthcare and agriculture industries. Commercial application of cello-oligosaccharides is limited due to the high-cost of their synthesis. Therefore, cost-effective production of cello-oligosaccharides from cellulose is attractive. During the hydrolysis of cellulose cello-oligosaccharides are formed as intermediates. Selective synthesis of cello-oligosaccharides is difficult because they are water soluble and undergo hydrolysis at a faster rate in comparison to cellulose in the presence of a homogeneous acid catalyst, resulting in formation of glucose. However, when cellulose is adsorbed on heterogeneous carbon catalyst, its hydrolysis is faster than the subsequent hydrolysis of cello-oligosaccharides. Given this, I chose carbon materials as catalysts to synthesize cello-oligosaccharides by hydrolysis of cellulose. An innovative semi-flow reactor was developed for the reaction in which the products could be rapidly removed from the reaction system, preventing their successive hydrolysis. In addition, a kinetic study was performed to understand the underlying mechanism of cellulose hydrolysis.

72 % yield of cello-oligosaccharides was obtained after hydrolysis at 473 K for 40 min over AC-Air, prepared by oxidation of activated carbon, in the semi-flow reactor with limited yield of dimer and monomer. Cello-oligosaccharides with a degree of polymerization (DP) up to 13 were observed with cellohexaose and celloheptaose obtained as the major products at a space velocity of 70 h^{-1}. Then, a kinetic analysis was performed to understand the mechanism for hydrolysis of β-1,4-glycosidic bonds in cellulose. Only in the presence of carbon catalyst, the rate of hydrolysis of individual cello-oligosaccharides was strongly dependent on their chain length. Larger cello-oligosaccharides underwent hydrolysis at a much faster rate. A plausible mechanism was proposed that larger molecules experience a greater degree of conformational distortion during their adsorption within the micropores of carbons. This leads to reduction in activation energy required to cleave the glycosidic bond and enhances the rate of reaction.

- Journal publications

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Soluble cello-oligosaccharides produced by carbon-catalyzed hydrolysis of cellulose. ChemSusChem, 2019, 12, 2576-2580.

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Unraveling the hydrolysis of β-1,4-glycosidic bonds in cello-oligosaccharides over carbon catalysts. Catalysis Science & Technology, 2020, 10, 4593-4601.

- Conference Contributions

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, The 125th Catalysis Society of Japan, Tokyo, Japan, March 27, 2020 (Oral)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, The 124th Catalysis Society of Japan, Nagasaki, Japan, September 18, 2019 (Oral)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, The 14th European Congress on Catalysis, EuropaCat 2019, Aachen, Germany, August 19, 2019. (Oral)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, The 122th Catalysis Society of Japan, Hakodate, Japan, September 27, 2018. (Oral)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, ICAT-BIC Students' workshop, Sapporo, Japan, November 21, 2018. (Oral)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, IRCCS the 3rd Joint International Symposium, Nagoya, Japan, January 31, 2020. (Poster)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, IRCCS The 5th Symposium, Sapporo, Japan, November 11, 2019. (Poster)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, IRCCS the 2rd Joint International Symposium, Kyoto, Japan, January 25, 2019. (Poster)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, The 14th Hokkaido University-Nanjing University-NIMS/FMCU Joint Symposium, Sapporo, Japan, December 07, 2018. (Poster)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Hokkaido University-National Central University Joint Symposium on Materials Chemistry and Physics, Sapporo, Japan, November 15, 2018. (Poster)

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Pre-TOCAT 8, Sapporo, Japan, August 03, 2018. (Poster)

３．今後の研究計画等 Future research plan

現在までの進捗状況等を踏まえ、今後の研究発表等を含めて具体的に記入のこと。
In detail, based on current progress, including a future research presentation plan.

This work investigates the transformation of cellulose to versatile cello-oligosaccharides and elucidates the mechanistic insights β-1,4-glycosidic bond hydrolysis over carbon catalyst. Future work should focus on reducing the requirement of pretreatment to enhance the contact between cellulose and carbon catalyst. Current technology relies on mix milling to improve the contact and adsorb cellulose on carbon surface, which requires a substantial amount of energy. Development of a novel catalyst that can remove the requirement of mix milling and directly depolymerize crystalline cellulose to cello-oligosaccharides is promising approach for the large-scale application in industries. In addition, detailed study on the unique behavior of preferential hydrolysis of terminal glycosidic bonds over carbon catalyst is necessary to understand the similarities between enzymes and heterogeneous carbon catalysts during cellulose hydrolysis.

- Journal publications in preparation

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Carbon catalyst with high density of vicinal carboxyl groups for the hydrolysis of β-1,4-glycosidic bonds in cellulose.

Pengru Chen, Abhijit Shrotri and Atsushi Fukuoka, Catalytic synthesis of oligosaccharides from cellulose.

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(https://www.global.hokudai.ac.jp/admissions/scholarships/scholarships-for-prospective-students/)

氏名 Name	Hla Myet CHEL	学　年 Year	2020
所属学院等名 Graduate School	Graduate School of Infectious Disease, Faculty of Veterinary Medicine		
所 属 専 攻 名 Major	Parasitology		
所属研究室等名 Laboratory	Laboratory of Parasitology		
指導教員職・氏名 Supervisor	Prof. Nariaki NONAKA		

１．研究テーマ名 Research theme

Study on parasites in Asian elephants from Myanmar

There is little information about the parasites infested in Asian elephants of Myanmar and no genetic information of those parasites is available. Thus, morphological and molecular identification of parasites in Asian elephants was studied.

2．研究等の進捗状況等 Research progress, etc

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※研究成果の発表・公表実績がある場合については学会名、掲載紙等の情報を含め詳細を記載すること

In Myanmar, there are approximately 3000 captive Asian elephants, *Elephas maximus*, in which cyathostomine gastrointestinal nematodes and stomach bot fly cause enteritis and death especially in younger animals. Despite the importance as the causative agents, there is no study on the prevalence of those parasites in Myanmar and there are few studies with the photomicrographs or molecular aspects so far. In this study, 47 nematodes and a bot fly larva were obtained from the faeces of Asian elephants in Myanmar after anthelmintic treatment and subjected to morphological identification and molecular analysis. In the results, five cyathostomine nematode species, *Murshidia falcifera* (n=3), *Murshidia indica* (1), *Murshidia neveulemairei* (10), *Quilonia renniei* (29) and *Quilonia travancra* (4), and one bot fly species, *Cobboldia elephantis* (1), were identified by morphology with providing the photomicrographs of key structures for morphological diagnosis. For molecular study, the partial sequences of the *COI* gene were determined for each species. Phylogenetic analysis revealed that *Murshidia indica, Quilonia renniei* and *Cobboldia elephantis* have close relationship to *Murshidia africana, Quilonia africana* and *Cobboldia loxodontis* reported from African elephants, respectively. It was also suggested that *Murshidia falcifera* and *Murshidia neveulemairei* constructs a clade with *Murshidia linstowi* and *Murshidia longicaudata* reported from African elephants. This clade is divided into three sub-clades, one by *Murshidia falcifera*, one by *Murshidia neveulemairei* and the last by *Murshidia linstowi* and *Murshidia longicaudata*. *Quilonia travancra* makes one separate clade. This study was the first report of the prevalence of five cyathostomine nematode species and one species of stomach bot fly in Asian elephants in Myanmar. This study could provide the photomicrographs of key structures for morphological identification of three *Murshidia*, two *Quilonia* and one *Cobboldia* species 100 years after the original drawings. The *COI* gene sequences of *M. falcifera, M. indica, M. neveulemairei, Q. renniei, Q. travancra,* and *C. elephantis* were deposited as the first time for genetic information in Asian elephants and the phylogenetic analysis was conducted with the parasite species found in African elephants.

Publications

Chel, H.M, Nakao, R., Ohsawa, N., Oo, Z.M., Nonaka, N., Katakura, K. First record and analysis of the *COI* gene of *Cobboldia elephantis* obtained from a captive Asian elephant from Myanmar. Parasitol. Int. 75: 102035, 2020.

Chel, H.M., Iwaki, T., Hmoon, M.M., Thaw, Y.N., Soe, N.C., Win, S.Y., Bawm, S., Htun, L.L., Win, M.M., Oo, Z.M., Masum, M.A., Ichii, O., Nakao, R., Nonaka, N., Katakura, K. Morphological and molecular identification of cyathostomine gastrointestinal nematodes of *Murshidia* and *Quilonia* species from Asian elephants in Myanmar. Int J Parasitol Parasites Wild. 11: 294-301, 2020.

Conference presentations

Chel, H.M., Thu, M.J., Htun, L.L., Bawm, S., Nakao, R. and Katakura, K. Detection of two types of Strongylidea parasites from Asian elephants in Myanmar. 161st Meeting of the Japanese Society of Veterinary Science, Tsukuba, September 11-13, 2018.

Chel, H.M., Thu, M.J., Htun, L.L., Bawm, S., Nakao, R. and Katakura, K. Morphological and molecular characterization of strongylid nematodes in Asian elephants of Myanmar. 88th Annual Meeting for the Japanese Society of Parasitology, Nagasaki, March 15-16, 2019.

3．今後の研究計画等 Future research plan

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In detail, based on current progress, including a future research presentation plan.

1. Scanning electron microscopy will be conducted to study the variation in morphology of parasites.
2. The whole mitochondrial genome of the nematode parasites will be determined for understanding genetic relationship among *Quilonia* and *Murshidia* species.
3. Morphological and molecular identification of trematode parasites will be studied for more information of parasitic infection in Asian elephants of Myanmar.
4. Genetic relationship between the third stage larvae and adult worms will be clarified.

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(https://www.global.hokudai.ac.jp/admissions/scholarships/scholarships-for-prospective-students/)

氏名 Name	ZHENG XIN	学　年 Year	博士３年
所属学院等名 Graduate School	環境科学院		
所 属 専 攻 名 Major	環境物質科学専攻		
所属研究室等名 Laboratory	野呂研究室		
指導教員職・氏名 Supervisor	野呂　真一郎		

１．研究テーマ名 Research theme

Studies on phase/structure transformable copper coordination polymers using bis(trifluoromethanesulfonyl)imide anion
（ビス（トリフルオロメタンスルホニル）イミドアニオンを用いた相転移あるいは構造変化できる銅配位高分子に関する研究）

2．研究等の進捗状況等 Research progress, etc.

研究の概要

Phase/structure transformations are almost everywhere in people's lives and have been well investigated from the melting of ice to the origin of superconductivity. Coordination polymers (CPs), a kind of crystalline solid constructed from metal ions and organic ligands, own a high diversity and flexibility of structures, which makes CPs good candidates for functional phase/structure transformable materials. However, designing phase/structure transformable CPs remains a challenge. In my research, I tried to synthesize a variety of phase/structure transformable CPs with a bulky and flexible fluorinated anion, bis(trifluoromethanesulfonyl)imide (NTf_2^-), which is one of common anions in ionic liquids (ILs), as a key building block. It was expected that the delocalized negative charges, low electric polarizability, and flexible structure with two revolving axes in the NTf_2^- anion contribute to weak intermolecular interactions and a variety of structures to form target transformable CPs.

研究成果の発表・公表実績

1. 論文

Xin Zheng, Hiroyasu Sato, Kiyonori Takåhashi, Shin-ichiro Noro, and Takayoshi Nakamura: "A Synchronous Change in Fluid Space and Encapsulated Anions in a Crystalline Polymethylene Unit Containing Metal–Organic Framework" *Cryst. Growth Des.*, 2020, *20*, 3596–3600.

2. 発表

(1) Xin Zheng, Shin-ichiro Noro, Kiyonori Takahashi, Takayoshi Nakamura, A series of copper coordination polymers constructed by bis(imidazole) ligands and bulky fluorinated anions: synthesis, structures, and physical properties, 錯体化学会 第68回討論会 (2018年7月)

(2) Xin Zheng, Shin-ichiro Noro, Kiyonori Takahashi, Takayoshi Nakamura, A series of copper coordination polymers constructed by bis(imidazole) ligands and bulky fluorinated anions: synthesis, structures, and physical properties, 第6回リーディングプログラム国際シンポジウム (2018年10月)

(3) Xin Zheng, Kiyonori Takahashi, Ichiro Hisaki, Takayoshi Nakamura, Shin-ichiro Noro, Effect of Uptake/Release of Coordinated Molecules on Gate-Opening Adsorption Behavior in Coordination Polymers, 北大・清華大学合同シンポジウム (2019年7月)

(4) Xin Zheng, Kiyonori Takahashi, Ichiro Hisaki, Takayoshi Nakamura, Shin-ichiro Noro, Effect of Uptake/Release of Coordinated Molecules on Gate-Opening Adsorption Behavior in Coordination Polymers, 錯体化学会 第69回討論会 (2019年9月)

(5) Xin Zheng, Kiyonori Takahashi, Ichiro Hisaki, Takayoshi Nakamura, Shin-ichiro Noro, Melting Coordination Polymers with Components Similar to Ionic Liquids, Japan-Germany Bilateral Symposium on Surface-Attached Metal-Organic Framework (2019年10月)

３．今後の研究計画等 Future research plan

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Although the bulky fluorinated anion, NTf_2^- has been widely used in ILs since the ILs including NTf_2^- anions usually have high stability and low viscosity, it is rarely used in CPs as the building block. In my research, NTf_2^- anion plays the key for diverse phase/structure transformations in CPs due to its flexible structure and ability to weaken intermolecular interactions. The strategy of introducing such bulky and flexible fluorinated anions to CPs will break with the stereotype images of crystalline CPs and open up a way to make hybrid amorphous materials. In the future, diverse kinds of bulky and flexible fluorinated anions will be adopted for the construction of phase/structure transformable CPs. I will explore and develop more novel physical properties of such kind CPs and attend the domestic and international conferences to present my research next year.

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氏名 Name	CHOU HUNG-WEI	学　年 Year	Ｄ３
所属学院等名 Graduate School	大学院環境科学院		
所 属 専 攻 名 Major	地球圏科学専攻		
所属研究室等名 Laboratory	三寺研究室		
指導教員職・氏名 Supervisor	三寺史夫　教授		

１．研究テーマ名 Research theme

Tidally-modified western boundary current drives interbasin exchange:
the dynamics of water exchange between the Sea of Okhotsk and the North Pacific through Kuril Straits by tidal forcing.

２．研究等の進捗状況等 Research progress, etc.

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Outlines, originality of our study:

The Sea of Okhotsk, as being the source of North Pacific Intermediate Water and North Pacific's nutrient origination, its interbasin exchange is crucial for bio-productivity prediction and overturning circulation in the North Pacific. However, the high-resolution models exhibit unrealistic extreme small transport except adding the tidal forcing. We are focusing on the problem of how the tidal forcing could improve the representative interbasin exchange in respect with ocean dynamics.

We found out that a tidally-induced small-scale current system is important to modify the western boundary current system, and consequently changes the large-scale flow pattern. The interbasin exchange is determined by the pathway of East Kamchatka Current, a western boundary current in the subarctic North Pacific. In the model without tidal forcing, the East Kamchatka Current overshoots the deep straits where the transport occurred. Once the tidal forcing is included, it deflects into the deep strait and eliminates the blocking of the interbasin transport. It is because a tidally-induced seamount trapped waves above a submarine bank upstream causes cross-slope upwelling, steering the western boundary current's pathway. This tidal/boundary current interaction dynamics can be described as a potential vorticity source for current's pathway through the

Joint-effect-of-baroclinicity-and-relief (JEBAR).

The dynamics of tides is often ignored in large-scale ocean simulations. Our study reveals not only the crucial dynamics of tidal forcing in high-resolution model, but also the question that what is the source of JEBAR. Conventionally, JEBAR is treated as a term reflecting the changes of baroclinicity by flows in potential vorticity equation instead of an external forcing. However, in this study, we addressed the source of JEBAR clearly. It is from the small-scale tidally induced feather described above, and it suggests that JEBAR can thus be a source for large-scale motion.

Academic presentations:

In 2017

1. OMIX (Ocean Mixing Processes summer conference),Source and Routes of Deep Water Masses East of Taiwan, Poster. Pebbu, KyuShu, Japan. 2017/09/14
2. オホーツク海と相互に影響を及ぼしあうグローカル大気海洋諸現象,Water Exchange between the Sea of Okhotsk and Pacifc in the point of view of Tides, Oral, Institute of Low Temperature Science, Hokkaido University, 2017/12/22

In 2018

3. Monbetsu Symposium, Hypothesis of Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects. Oral. Monbetsu, Hokkaido, Japan, 2018/02/20, **International.**
4. 新学術領域「新海洋混合学」全体会議, Dynamics of Barotropic water exchange between the sea of Okhotsk and the Pacific from a point of view of tidal effects,poster, Chiba, Japan, 2018/03/17
5. JPGU, Hypothesis of Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects, poster, Chiba, Japan, 2018/05/22
6. 宗谷暖流研究集会, Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects, Oral, Hokkaido, Japan, 2018/07/06
7. OMIX (Ocean Mixing Processes) summer school, Dynamics of Barotropic Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects, Oral, Chiba, Japan, 2018/10/05
8. Japan Ocean society meeting, Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects, Oral, Tokyo, Japan, 2018/09/26
9. OMIX 国際シンポジウム, Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific from a point of view of Tidal Effects, poster, Chiba, Japan, 2018/11/04

In 2019

10. 4th International Joint Workshop on Computationally Intensive Modeling of the Climate System and 9th OFES International Workshop, Improvement and Dynamics of Water Exchange between the Sea of Okhotsk and the Pacific through tidal effects in OGCM, Oral, Kyushu, Japan, 2019/03/01, **International**.
11. annual Poster Presentation Session, Hokkaido University, 2019/03/06, Poster presentation.
12. OMIX annual meeting, Impact of Tidal Vertical Mixing to Barotropic Water Exchange between the Sea of Okhotsk and Pacific through Diffusive Adjustment with Turbulent Closure Scheme, poster, Sapporo, Japan, 2019/03/13

13. JpGU, Impact of Tidal Vertical Mixing to Barotropic Water Exchange between the Sea of Okhotsk and Pacific through Diffusive Adjustment with Turbulent Closure Scheme: Can Vertical Diffusivity Vale represent the Tidal Impact in OGCM?, poster, Chiba, Japan, 2019/05/24
14. OMIX summer school, the Dynamics of the exchange between the Sea of Okhotsk and the Pacific through Tidal forcing, Oral, Shizuoka, Japan, 2019/8/26
15. 海洋システムの統合的理解に向けた新時代の力学理論の構築, the Dynamics of the exchange between the Sea of Okhotsk and the Pacific through Tidal forcing, Oral, Sapporo, Japan, 2019/09/5
16. 海洋若手会 Summer School, Finding the Upwelling Region of NPIW, poster, Chiba, Japan, 2019/09/8

In 2020

17. Ocean Science Meeting 2020, Reveal of Dynamics of Barotropic Exchange between the Sea of Okhotsk and North Pacific through Tidal forcing in High-Resolution Ocean General Circulation Model: the Modification of the Western Boundary Current Pathway by Tidal Rectification, Oral, San Diego, USA, 21th Feb 2020. **International.**
18. Japan Oceanography Society Fall Meeting 2020, Tidally-modified western boundary current drives interbasin exchange, Oral, Online, Japan, 27th Nov 2020. Domestic.

Awards:

1. Aota Masaaki Award in Physisc, Monbetsu Symposium, 2018/02/21
2. Research Excellent Award of Environmental College, Hokkaido University, 2019/03/06

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Progress:

The manuscript of present study (Tidally-modified western boundary current drives interbasin exchang) is prepared and will submit soon.

Future plan:

We further want to know that the impacts of difference of interbasin exchange to North Pacific circulations numerically. The identical isopycnal layer of North Pacific Intermediate Water is 26.8σ. By historical hydrographic data from National Ocean Database Center, the 26.8σ indicates a cold water belt through Tenno Seamount Chain from the Sea of Okhotsk. However, the phenomia cannot be found in the COCO model without tidal forcing. It implies that the interbasin exchange simulation did influence the large scale circulation over whole North Pacific. The understanding of this influence will be our next study.