戦略的国際共同研究プログラム(SICORP)

日本一ベトナム・フィリピン・インドネシア共同研究

終了報告書 概要

- 1. 研究課題名:「東南アジア特有な生体分子とナノカーボン材料の融合による革新的ナノ バイオデバイスの創製」
- 2. 研究期間:令和2年4月~令和5年3月
- 3. 主な参加研究者名: 日本側チーム

	氏名	役職	所属	研究分担		
研究代表者	梅村 和夫	教授	東京理科大学	研究総括		
主たる	平 久夫	講師	北海道教育大学	理論計算		
共同研究者						
研究参加者	宮代 大輔	博士学生	東京理科大学	理論計算		
研究参加者	松川 雄二	博士学生	東京理科大学	ナノ実験		
研究参加者	井出 祐貴	博士学生	東京理科大学	ナノ実験		
研究参加者	Nay San Lin	博士学生	東京理科大学	ナノ実験		
研究	研究期間中の全参加研究者数 11名					

ベトナム側チーム

	氏名	役職	所属	研究分担
研究代表者	Trong-Tan Do	Director	Ministry of Science and Technology, National Centre for Technological Progress	PI, nano fabrication
主たる 共同研究者	Anh-Tuan Mai	Associate Professor	VNU University of Science and Technology	Co-PI, nano fabrication
主たる 共同研究者	Thi-Xuan Chu	Researcher, Lecturer	Hanoi University of Science and Technology	Nano fabrications
研究参加者	Thi-Luyen Tran	Researcher, Lecturer	Hanoi University of Science and Technology	Nano fabrications
研究参加者	Binh-Duong Le	Researcher	Ministry of Science and Technology, National Centre for Technological Progress	Nano fabrications
研究参加者	Minh Hieu Nguyen	Lecturer	VNU University of Science	Nano characterizations
研	究期間中の全参	於加研究者数	6名	

フィリピン側チーム

	こり例グ			
	氏名	役職	所属	研究分担
研究代表者	Leo Cristobal II Ambolode	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose's and hybrid's electronic properties simulation
主たる	ARNOLD	Professor	Mindanao State	Cellulose
	ALGUNO		University – Iligan	synthesis and

共同研究者			Institute of Technology (MSU-IIT)	characterization
主たる共同研究者	Eli Christopher Enobio	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Device prototype and characterization
主たる共同研究者	Rosario L. Reserva	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose's and hybrid's mechanical properties simulation
主たる 共同研究者	Rolando T. Candidato, Jr.	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose and hybrid synthesis and characterization
主たる共同研究者	Rey Y. Capangpan gan	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose and hybrid synthesis and characterization
研究期間中の全参加研究者数 20名				

インドネシア側チーム

	氏名	役職	所属	研究分担
研究代表者	Eko Siswoyo	Associate Professor	Islamic University of Indonesia (UII), Environmental Engineering	As a leader of the research group in Indonesia, he organizes the research activity from the beginning until finish and also preparing manuscript for publication in the international journal.
主たる共同研究者	Awaluddin Nurmiyanto	Assistant Professor	Islamic University of Indonesia (UII), Environmental Engineering	research and analyze data together with all members.
主たる 共同研究者	Nur Aini Iswati Hasanah	Lecturer	Islamic University of Indonesia (UII), Environmental Engineering	Conduct research and analyze data together with all members.
研究参加者	Lutfia Isna Ardhayanti	Researcher	Islamic University of Indonesia (UII), Environmental Engineering	Conduct research and analyze data together with all members.
研	究期間中の全参	加研究者数	4 名	

4. 国際共同研究の概要

パパインなどの東南アジア特有の生体分子と、カーボンナノチューブなどのナノカーボンとのハイブリッド化を行い、高温で安定のあるなどの特徴を持つ新規なナノ・マイクロバイオデバイスを提案すると同時に、微量バイオセンシングに応用する。

5. 国際共同研究の成果

5-1 国際共同研究の学術成果および実施内容

パパイン、天然セルロース、天然界面活性剤、ココナツハスクなどを用いたバイオ・ナノカーボン複合体を多種類試作し、ナノカーボンの近赤外発光現象を用いて生体分子の種類を識別するなどの応用を行った。また、原子間力顕微鏡などのナノ計測実験、および計算科学からのアプローチで生体物質とナノカーボンの相互作用について知見を得た。これらにより3年間のプロジェクトにおいて46の学会報告、および21報の原著論文を発表した。

5-2 国際共同研究による相乗効果

21 報の原著論文のうち 4 報は国際共同研究による国際共著論文を刊行することができた。COVID-19 による障害はプロジェクト期間全般に及んだが、計算科学的なアプローチを行い、当初計画に拘泥せず稼働できる研究設備での実施を行うなどの工夫を行った。人材育成・交流については 2 名の国費留学生と 1 名の研究員を日本チームに迎え、最終年度には日本チームのメンバーが各国に出向いて共同実験、シンポジウム参加・研究打ち合わせを行うことができた。また、各年度秋に開催したオープンなシンポジウムでは 100 名を超える聴講者を得て研究成果の普及に努めた。

5-3 国際共同研究成果の波及効果と今後の展望

本プロジェクトの研究成果が社会に与える波及効果について、第一にナノカーボンのキラリティに起因する近赤外発光を用いて生体分子を識別する手法は、今後さらに洗練することで新規な微量バイオセンシング技術として確立できるものと考えられる。第二に、本研究で試作した高温で攪拌なしに高い活性を示すパパイン酵素デバイスは、安全な材料によるマイクロバイオデバイスとして活用が期待される。また、第一の成果と合わせることで、酵素反応をモニターできると考えられる。第三に、本プロジェクトで作製した多種類の東南アジア特有な生体分子とナノカーボンの複合体は、計算科学からの検証も合わせて、高温で安定なナノバイオデバイスなどとして多方面への利用が期待される。

Strategic International Collaborative Research Program (SICORP) Japan-Vietnam, Philippines, Indonesia Joint Research Program Executive Summary of Final Report

- 1. Project title: Development of innovative nano-biodevices based on hybrid materials by combination of endemic South Asian biomolecules and nanocarbons.
- 2. Research period : April 2020 \sim March 2023
- 3. Main participants:

Japan-side

<u> </u>	Japan-Side					
	Name	Title	Affiliation	Role in the		
				research project		
PI	Kazuo	Professor	Tokyo University of	Leader PI		
	UMEMURA		Science			
Co-PI	Hisao Taira	Junior	Hokkaido Education	Theoretical		
		Associate	University	approach		
		Professor				
Collaborator	Daisuke	Ph.D.	Tokyo University of	Theoretical		
	Miyashiro	student	Science	approach		
Collaborator	Yuji	Ph.D.	Tokyo University of	Nano		
	Matsukawa	student	Science	experiments		
Collaborator	Yuki Ide	Ph.D.	Tokyo University of	Nano		
		student	Science	experiments		
Collaborator	Nay San Lin	Ph.D.	Tokyo University of	Nano		
	-	student	Science	experiments		
Tota	al number of partic	cipants through	out the research perior	d: 11		

Vietnam-side

	Name	Title	Affiliation	Role in the research project		
PI	Trong-Tan Do	Director	Ministry of Science and Technology, National Centre for Technological Progress	PI, nano fabrications		
Co-PI	Anh-Tuan Mai	Associate Professor	VNU University of Science and Technology	Co-PI, nano fabrications		
Co-PI	Thi-Xuan Chu	Researcher, Lecturer	Hanoi University of Science and Technology	Nano fabrications		
Collaborator	Thi-Luyen Tran	Researcher, Lecturer	Hanoi University of Science and Technology	Nano fabrications		
Collaborator	Binh-Duong Le	Researcher	Ministry of Science and Technology, National Centre for Technological Progress	Nano fabrications		
Collaborator	Minh Hieu Nguyen	Lecturer	VNU University of Science	Nano characterizations		
Tot	Total number of participants throughout the research period: 6					

Philippines -side

гішррі	Philippines -side					
	Name	Title	Affiliation	Role in the		
				research project		
PI	Leo Cristobal II Ambolode	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Leader PI Cellulose's and hybrid's electronic properties simulation		
Co-PI	ARNOLD ALGUNO	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose synthesis and characterization		
Co-PI	Eli Christopher Enobio	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Device prototype and characterization		
Co-PI	Rosario L. Reserva	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose's and hybrid's mechanical properties simulation		
Co-PI	Rolando T. Candidato, Jr.	Professor	Mindanao State University – Iligan Institute of Technology (MSU-IIT)	Cellulose and hybrid synthesis and characterization		
Co-PI	Rey Y. Capangpangan	Professor	Mindanao State University – Naawan	Cellulose and hybrid synthesis and characterization od: 20		
Total number of participants throughout the research period: 20						

Indonesia-side

	Name	Title	Affiliation	Role in the research project
PI	Eko Siswoyo	Associate Professor	Islamic University of Indonesia (UII), Environmental Engineering	As a leader of the research group in Indonesia, he organizes the research activity from the beginning until finish and also preparing manuscript for publication in the international journal.
Co-PI	Awaluddin Nurmiyanto	Assistant Professor	Islamic University of Indonesia (UII), Environmental Engineering	Conduct research and analyze data together with all members.

Co-PI	Nur Aini Iswati	Lecturer	Islamic University	Conduct research
	Hasanah		of Indonesia (UII),	and analyze data
			Environmental	together with all
			Engineering	members.
Collaborator	Lutfia Isna	Researcher	Islamic University	Conduct research
	Ardhayanti		of Indonesia (UII),	and analyze data
			Environmental	together with all
			Engineering	members.
Total number of participants throughout the research period: 4				

4. Summary of the international joint research

Southeast Asian specific biomolecules such as papain and nanocarbons such as carbon nanotubes will be hybridized in order to propose novel nano/micro biodevices which reveal specific advantages such as sufficient stability at high temperatures. The fabricated hybrids will be applied to bio-sensing at nano/micro scales.

5. Outcomes of the international joint research

5-1 Scientific outputs and implemented activities of the joint research

We fabricated many types of bio-nanocarbon composites using papain, natural cellulose, natural surfactants, and coconut husks. One of the fabricated hybrids were applied to the near-infrared emission phenomenon of nanocarbons to identify types of biomolecules. Nanoscale evaluation of the hybrids such as atomic force microscopy and computational approaches have provided insights into the interactions between biomaterials and nanocarbons. 44 conference presentations and 21 original papers were published during the three-year project.

5-2 Synergistic effects of the joint research

Four of the 21 original papers were published as international collaborated papers as a result of this international joint project. Although the COVID-19 problem provided various difficulties throughout the project period, we took a computational approach and demonstrated experimental researches by possible implementation methods. In terms of human resource development and international communications, two students were invited with government scholarship (MEXT). A researcher was also invited. In the final year, members of the Japan team were able to visit counter parts to conduct joint experiments, participate in symposiums, and hold research meetings. In addition, open symposiums held in the fall of each fiscal year attracted more than 100 attendees every time.

5-3 Scientific, industrial or societal impacts/effects of the outputs

Regarding the ripple effects of the research results of this project on society, first, the method of identifying biomolecules using near-infrared emission based on the specific optical responses of different chiralities of nanocarbons could be established as a novel micro biosensing techniques through further refinement in the future. Second, the papain enzyme device prototyped in this study, which exhibits high activity at high temperatures without stirring, is expected to be utilized as an effective microbiodevice made of natural materials. Combining the first and second results, the enzymatic reaction could be monitored without dyes. Third, the composites of nanocarbons with many kinds of Southeast Asian specific biomolecules fabricated in this project, together with verification from computational science, are expected to be used in many fields as advantageous nanobiodevices, including biodevices which are stable at high temperatures.

国際共同研究における主要な研究成果リスト

1. 論文発表等

- *原著論文(相手側研究チームとの共著論文)発表件数:計5件
- ・査読有り:発表件数:計5件
- Daisuke Miyashiro, Hisao Taira, Ryo Hamano, Rosario L. Reserva and Kazuo Umemura, Mechanical vibration of single-walled carbon nanotubes at different lengths by modal analysis method, *Composites Part C: Open Access*, 2020, Volume 2, 100028 DOI: 10.1016/j.jcomc.2020.100028
- Kazuo Umemura, Ryo Hamano, Hiroaki Komatsu, Takashi Ikuno, and Eko Siswoyo, Dispersion of Carbon Nanotubes with "Green" Detergents, Molecules 2021, 26(10), 2908 DOI: 10.3390/molecules26102908
- Makoto Saito, Masaki Kitamura, Yuki Ide, Minh Hieu Nguyen, Binh Duong Le, Anh Tuan Mai, Daisuke Miyashiro, Shigeki Mayama, Kazuo Umemura, An Efficient Method of Observing Diatom Frustules via Digital Holographic Microscopy, Microscopy and Microanalysis, 2022, 28, 2172–2176 DOI:10.1017/S1431927622012508
- 4. Kota Hirayama, Masaki Kitamura, Nay San Lin, Minh Hieu Nguyen, Binh Duong Le, Anh Tuan Mai, Shigeki Mayama, and Kazuo Umemura, Attachment of DNA-Wrapped Single-Walled Carbon Nanotubes (SWNTs) for a Micron-Sized Biosensor, ACS Omega, 2022, 7, 50, 47148–47155 DOI: 10.1021/acsomega.2c06278
- Eko Siswoyo, Rahmalina Nur Zahra, Nurul Hardina A. Mai, Awaluddin Nurmiyanto, Kazuo Umemura, Thomas Boving, Chitosan of blood cockle shell (Anadara granosa) as a natural coagulant for removal of total suspended solids (TSS) and turbidity of well-water, The Egyptian Journal of Aquatic Research, 2023, in press, DOI: 10.1016/j.ejar.2023.04.004
- ・査読無し:発表件数:計0件該当なし。
- *原著論文(相手側研究チームを含まない日本側研究チームの論文):発表件数:計16件・査読有り:発表件数:計16件
- 1. Yuhei Horii, Shusuke Ohura, Kazuo Umemura, An efficient method to quantitatively detect competitive adsorption of DNA on single-walled carbon nanotube surfaces, Analytical Biochemistry, **2020**, 601, 113776 DOI:10.1016/j.ab.2020.113776
- Ryo Hamano, Daisuke Miyashiro, Kazuo Umemura, Study on optical response sensitivity in hybrid of single-walled carbon nanotubes mixed with double-stranded DNA and carboxymethylcellulose, Optical Materials, 2020, 109, 110386 DOI: 10.1016/j.optmat.2020.110386
- Daisuke Miyashiro, Ryo Hamano, HisaoTaira, Kazuo Umemura, Analysis of vibration behavior in single strand DNA-wrapped single-walled carbon nanotubes adhered to lipid membranes, Forces in Mechanics, 2020, 2, J100008 DOI: 10.1016/j.finmec.2020.100008
- Yuji Matsukawa, Kazuo Umemura, Chirality luminescent properties of single-walled carbon nanotubes during redox reactions, Optical Materials, 2020, 112, 110748 DOI: 10.1016/j.optmat.2020.110748
- Yuji Matsukawa, Kazuo Umemura, Optical Response Characteristics of Single-Walled Carbon Nanotube Chirality Exposed to Oxidants with Different Oxidizing Power, Molecules, 2021, 26, 4, 1091 DOI: 10.3390/molecules26041091
- Ryo Hamano, Shingo Shoumura, Yuto Takeda, Tokio Yamazaki, Kota Hirayama, Yasutaka Hanada, Shiegki Mayama, Masaharu Takemura, Han-Jia Lin and Kazuo Umemura, Sinking of Four Species of Living Diatom Cells Directly Observed by a "Tumbled" Optical Microscope, Microscopy and Microanalysis, 2021, 27, 5, 1154-1160 DOI:10.1017/S1431927621012150
- 7. Kota Hirayama, Masaki Kitamura, Ryo Hamano, and Kazuo Umemura, Stable

- Near-Infrared Photoluminescence of Single-Walled Carbon Nanotubes Dispersed Using a Coconut-Based Natural Detergent, ACS Omega, **2021**, 6, 45, 30708–30715 DOI: 10.1021/acsomega.1c04615
- 8. Ryo Hamano, Shigeki Mayama, and Kazuo Umemura, Localization analysis of intercellular materials of living diatom cells studied by tomographic phase microscopy, Applied Physics Letters, **2022**, 120, 133701 DOI: 10.1063/5.0086165
- Satoru Takahama, Masaki Kitamura, Yuki Ide, Kazuo Umemura, Variation in the responses of carbon quantum dots (CQDs) synthesized from native coconut husk and coconut husk-derived charcoal, Optical Materials, 2022, 131, 112739 DOI: 10.1016/j.optmat.2022.112739
- Nay San Lin, Masaki Kitamura, Makoto Saito, Kota Hirayama, Yuki Ide, and Kazuo Umemura, Distinguishing Antioxidant Molecules with Near-Infrared Photoluminescence of DNA-Wrapped Single-Walled Carbon Nanotubes, ACS Omega, 2022, 7, 33, 28896–28903 DOI: 10.1021/acsomega.2c02038
- Naysan Lin, Masaki Kitamura, Kota Hirayama, Ryo Hamano, Kazuo Umemura, Antioxidant Properties of Asian Originated Biomaterials Detected with DNA-wrapped Single Walled Carbon Nanotubes, The ACM International Conference Proceeding Series (ICPS), 2023, 115–119 DOI: 10.1145/3571532.3571550
- 12. Masaki Kitamura, Kazuo Umemura, Hybridization of papain molecules and DNA-wrapped single-walled carbon nanotubes evaluated by atomic force microscopy in fluids, Scientific Reports, **2023**, 13, 4833 DOI: 10.1038/s41598-023-31927-8
- 13. Kota Hirayama, Masaki Kitamura, Shigeki Mayama, and Kazuo Umemura, Chirality-specific near-infrared photoluminescent responses of single-walled carbon nanotubes to detect papain adsorption assisted by the use of DNA and frustules, Chemistry Letters, in press, DOI: 10.1246/cl.230068
- 14. Hisao Taira. Daisuke Miyashiro, Kazuo Umemura, Numerical Analysis for Light Absorption Spectra of the Base of DNA-Wrapped Single-Walled Carbon Nanotubes, Molecules, 2023, 28(6), 2719, DOI: 10.3390/molecules28062719
- 15. Shuaib Edakkaparamban , Masaki Kitamura, Yuki Ide, Kazuo Umemura, Asuka Ishizawa, Photocatalytic degradation study of methylene blue using carbon quantum dots synthesized from coconut husk, Materials Letters, **2023**, in press
- 16. Lin, Nay; Hirayama, Kota; Kitamura, Masaki; Koide, Shinji; Kitajima, Hiromasa; Harada, Takunori; Mayama, Shigeki; Umemura, Kazuo, Fabrication of a Floatable Micron-sized Enzyme Device Using Diatom Frustules, ACS Omega, in press
- ・査読無し:発表件数:計0件該当なし。
- *その他の著作物(相手側研究チームとの共著総説、書籍など):発表件数:計1件
- Kazuo Umemura, Nirmal Mazumder (Editor), Richard Gordon (Editor), Diatom Microscopy, Chapter 2, Page 33-56, Nanobioscience Studies of Living Diatoms Using Unique Optical Microscopy Systems, July 2022 368 Pages, John Wiley & Sons, Inc. ISBN: 978-1-119-71153-7

*その他の著作物(相手側研究チームを含まない日本側研究チームの総説、書籍など):発表件数:計0件 該当なし。

2. 学会発表

*ロ頭発表(相手側研究チームとの連名発表) 発表件数:計4件(うち招待講演:1件)

*口頭発表(相手側研究チームを含まない日本側研究チームの発表)

発表件数:計42件(うち招待講演:10件)

*ポスター発表(相手側研究チームとの連名発表)

発表件数:計0件

*ポスター発表(相手側研究チームを含まない日本側研究チームの発表)

発表件数:計7件

3. 主催したワークショップ・セミナー・シンポジウム等の開催

- 1. 22nd SPVM National Physics Conference: ConVIRTUALisation, Leo Cristobal C. Ambolode II (Mindanao State University Iligan Institute of Technology (MSU-IIT), Professor), Online, 17th, 24th, and 31st, Oct. 2020, 400 名。
- 2. e-ASIA Kick-Off meeting, Kazuo Umemura (Tokyo University of Science, Professor), Online, 31st Aug. 2020, 10 名。
- 3. International Symposium on Nanoscience and its Applications (ISNA), Leo Cristobal C. Ambolode II (Mindanao State University Iligan Institute of Technology (MSU-IIT), Professor), Online, 2nd-3rd, Dec. 2021, 80 名。
- 4. 2022 International Conference on Nanoscience and Nanotechnology, Leo Cristobal C. Ambolode II (Mindanao State University Iligan Institute of Technology (MSU-IIT), Professor), Butuan, Philippines, 8th-9th, Dec., 100 名。
- 5. e-ASIA Closing meeting, Kazuo Umemura (Tokyo University of Science, Professor), Online, 3rd Mar. 2023, 10 名。

4. 研究交流の実績(主要な実績)

【講義】

・2021年2月24日、日本側チームの博士後期課程3年学生がオンラインでミンダナオ州立大学イリガン工科校の学生に向けてデータ解析の講義を行った。

【学生・研究者の派遣、受入】

- ・2022年9月~10月:フィリピンからの研究員を日本側チームに受け入れた。
- ・2023年1月:日本側教員と学生がベトナム訪問し、生体分子とナノカーボン複合体を持参するとともに評価実験を行った。
- ・2023年2月:日本側学生がインドネシア訪問し、インドネシア側が開発している吸着剤にナノカーボンを添加して強化する共同実験を行った。
- ・2021年7月:e-ASIA 枠にて文部科学省国費留学生をミャンマーから受け入れた。
- ・2022年10月:一般枠にて文部科学省国費留学生をフィリピンから受け入れた。

5. 特許出願

研究期間累積出願件数:0件

6. 受賞·新聞報道等

・Best Oral Presentation Runner Up を東京理科大学の博士後期課程 2 年が受賞。 国際会議 RSC Interest Group Chemical Nanoscience and Nanotechnology Annual Symposium, The Royal Society of Chemistry にて。

7. その他

・2022年8月:東京理科大学オープンキャンパスで研究紹介を行った。