様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Colloidal-rods for examining polymer dynamics in complex flows

研究課題名（英文）Colloidal-rods for examining polymer dynamics in complex flows

研究代表者
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研究者番号：４０８９５４１３

交付決定額（研究期間全体）：（直接経費）　　2,600,000円

研究成果の概要（和文）：プロジェクトのこの 2 年間で、候補者はポリマー溶液中のコロイドロッドの整列の動力学の基本的な理解を進めました。この候補者は、比較的小さなコロイドロッドの流れによる整列の開始が、周囲のポリマーの緩和力学と結びついていることを示した。逆に、比較的大きなコロイドロッドの流れによる整列の開始は、ポリマー溶液のバルク粘度によって支配されます。この理解は、提案された研究プロジェクトの主な目的にとって重要な結論をもたらします。

研究成果の学術的意義や社会的意義
The results obtained with this project indicate that colloidal rod alignment in polymer solutions can be predicted on the basis of the critical shear rate at which polymer coils are deformed by the flow, aiding the synthesis and design of anisotropic materials.

研究成果の概要（英文）：During these two years of the project, the candidate advanced the fundamental understanding of the dynamics of colloidal-rods alignment in polymeric solutions. The candidate has shown that the onset of flow-induced alignment of relatively small colloidal-rods is coupled with the relaxation dynamics of the surrounding polymers. On the contrary, the onset of flow-induced alignment of relatively large colloidal-rods is govern by the bulk viscosity of the polymer solution. This understanding has important conclusions for the primary purpose of the proposed research project. Specifically, this understanding can lead to the development of a technique based on the alignment of colloidal rods, referred to as colloidal-rods alignment micro-rheology (CRAM), to provide quantitative and spatially resolved structural properties of polymeric fluids in complex flows.

研究分野：polymer

キーワード：polymers　colloids　rheology　microfluidics

様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９（共通）

１．研究開始当初の背景

Rheological and structural properties of viscoelastic fluids are conventionally characterized under homogeneous flow conditions, comprising simple shearing or extensional deformations. However, non-homogeneous, complex flows comprising mixed contribution of shearing and extensional deformations are ubiquitously encountered in “real-life” industrial (e.g. fibre spinning, spraying) and biological processes (e.g. spermatozoa beating, flagellar locomotion in bacteria). Therefore, in situ characterization methods that provide spatial and temporal resolution of structural and dynamical properties of polymeric fluids are vital.
Methodologies that probe the structural conformations of polymers under complex flows are limited, and rely on specific polymer properties, such as the high polarizability of the monomer unit. Therefore, the general understanding of polymeric fluids under complex flows is restricted to specific polymer types.

２．研究の目的

The proposed idea is the use of tracer colloidal-rods, with sub-micron length scales, to probe the structure and dynamics of polymeric solutions in microfluidic flows. The core idea is based on two propositions: (a) colloidal-rods suspended in a fluid progressively orient with the increasing deformation rate, inducing structural order and causing double diffraction of the light and (b), the surrounding polymer crowding dictates the alignment of colloidal-rods. The project aims at retrieving structural parameters of the polymer surrounding the colloidal-rods through the birefringence derived from the colloidal-rod alignment.
Understanding the hydrodynamic alignment of colloidal rods in polymer solutions is pivotal for manufacturing structurally ordered materials. How polymer crowding influences the flow-induced alignment of suspended colloidal rods remains unclear when rods and polymers share similar length scales. In this project the candidate tackles this problem by analyzing the alignment of colloidal rods suspended in crowded polymer solutions and comparing that to the case where crowding is provided by additional colloidal rods in a pure solvent.

３．研究の方法

For the completion of the project a multitude of techniques and methods have been used. The primary experimental setup employed in this project was flow induced birefringence (FIB) coupled with in-house built microfluidic devices fabricated with selective laser-induced etching (SLE) technique. Microfluidic devices are versatile platforms to study the flow-induced alignment of colloidal rods in different flow fields and for the retrieval of relevant time- and length-scales associated with the colloidal rods. For instance, microfluidic devices can be designed to generate two-dimensional (2D) flows that provide, to a good approximation, a uniform flow through the channel height (e.g., microfluidics with rectangular cross sections and relatively high aspect ratio). 2D flows are convenient for flow visualization and for techniques aimed at probing the structural properties of the fluid, such as FIB and small-angle scattering (SAS). Alternatively, three-dimensional (3D) flows, generated in microfluidic devices with symmetric and/or low aspect ratio cross-sections, are more representative of processing condition under which naturally derived CR may be employed (e.g., fiber spinning). Most importantly, microfluidics can be designed to generate not only shearing flows (the flow type generated by rotational rheometers) but also extensional flows and mixed flows comprising both shearing and extensional deformations. Thus, coupling microfluidics with flow visualization techniques, pressure sensors, FIB, and/or SAS techniques provides a comprehensive fingerprint of the structure–property relationship of colloidal rods in unique flow scenarios. Moreover, the small length scales adopted in microfluidic devices allow relatively high deformation rates while preserving creeping flow conditions.
In this project, the polymeric fluids were routinely tested using classical shear rheometry. To gauge the fluid response in extensional-dominated flows, the polymeric fluids were also tested using techniques based on the capillary-driven self-thinning of polymeric fluids (e.g., Capillary Breakup Extensional Rheometry CaBER).

４．研究成果

During these two years of the project, the candidate advanced the fundamental understanding of the dynamics of colloidal-rods alignment in polymeric solutions. The candidate has shown that the onset of flow-induced alignment of relatively small colloidal-rods is coupled with the relaxation dynamics of the surrounding polymers. On the contrary, the onset of flow-induced alignment of relatively large colloidal-rods is govern by the bulk viscosity of the polymer solution. This understanding has important conclusions for the primary purpose of the proposed research project. Specifically, this understanding can lead to the development of a technique based on the alignment of colloidal rods, referred to as colloidal-rods alignment micro-rheology (CRAM), to provide quantitative and spatially resolved structural properties of polymeric fluids in complex flows.

５．主な発表論文等

〔雑誌論文〕　計6件（うち査読付論文　0件／うち国際共著　6件／うちオープンアクセス　6件）

１．著者名 Calabrese Vincenzo、Porto Santos Tatiana、Lopez Carlos G.、Lettinga Minne Paul、Haward Simon J.、Shen Amy Q.	４．巻 6
２．論文標題 Extensibility governs the flow-induced alignment of polymers and rod-like colloids	５．発行年 2024年
３．雑誌名 Physical Review Research	６．最初と最後の頁 1-6
掲載論文のDOI（デジタルオブジェクト識別子） 10.1103/PhysRevResearch.6.L012042	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Detert Marvin、Santos Tatiana Porto、Shen Amy Q.、Calabrese Vincenzo	４．巻 24
２．論文標題 Alignment?Rheology Relationship of Biosourced Rod-Like Colloids and Polymers under Flow	５．発行年 2023年
３．雑誌名 Biomacromolecules	６．最初と最後の頁 3304～3312
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.biomac.3c00347	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Calabrese Vincenzo、Shen Amy Q.、Haward Simon J.	４．巻 17
２．論文標題 Naturally derived colloidal rods in microfluidic flows	５．発行年 2023年
３．雑誌名 Biomicrofluidics	６．最初と最後の頁 021301～021301
掲載論文のDOI（デジタルオブジェクト識別子） 10.1063/5.0142867	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Santos Tatiana P.、Calabrese Vincenzo、Boehm Michael W.、Baier Stefan K.、Shen Amy Q.	４．巻 638
２．論文標題 Flow-induced alignment of protein nanofibril dispersions	５．発行年 2023年
３．雑誌名 Journal of Colloid and Interface Science	６．最初と最後の頁 487～497
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.jcis.2023.01.105	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

1．著者名 Calabrese Vincenzo、Gy?rgy Csilla、Haward Simon J.、Neal Thomas J.、Armes Steven P.、Shen Amy Q.	4．巻 55
2．論文標題 Microstructural Dynamics and Rheology of Worm-like Diblock Copolymer Nanoparticle Dispersions under a Simple Shear and a Planar Extensional Flow	5．発行年 2022年
3．雑誌名 Macromolecules	6．最初と最後の頁 10031～10042
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.macromol.2c01314	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

1．著者名 Calabrese Vincenzo、Varchanis Stylianos、Haward Simon J.、Shen Amy Q.	4．巻 55
2．論文標題 Alignment of Colloidal Rods in Crowded Environments	5．発行年 2022年
3．雑誌名 Macromolecules	6．最初と最後の頁 5610～5620
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acs.macromol.2c00769	査読の有無 無
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計9件（うち招待講演　0件／うち国際学会　7件）

1．発表者名 Calabrese Vincenzo、Shen Amy Q, Haward Simon J.
2．発表標題 How do polymers stretch in capillary-driven extensional flows?
3．学会等名 Colloidal, Macromolecular and Polyelectrolyte Solutions（国際学会）
4．発表年 2024年

1．発表者名 Calabrese Vincenzo、Porto Santos Tatiana、Lopez Carlos G.、Lettinga Minne Paul、Haward Simon J.、Shen Amy Q.
2．発表標題 Extensibility governs the flow-induced alignment of polymers and rod-like colloids
3．学会等名 Colloidal, Macromolecular and Polyelectrolyte Solutions（国際学会）
4．発表年 2024年

１．発表者名
Calabrese Vincenzo、Porto Santos Tatiana、Lopez Carlos G.、Lettinga Minne Paul、Haward Simon J.、Shen Amy Q.

２．発表標題
Extensibility governs the flow-induced alignment of polymers and rod-like colloids

３．学会等名
7th International Soft Matter Conference (ISMC2023), Osaka (JP).（国際学会）

４．発表年
2023年

１．発表者名
Calabrese Vincenzo、Porto Santos Tatiana、Lopez Carlos G.、Lettinga Minne Paul、Haward Simon J.、Shen Amy Q.

２．発表標題
A unified framework to describe shear- and extension-induced alignment of macromolecules of various flexibility

３．学会等名
International congress on Rheology（国際学会）

４．発表年
2023年

１．発表者名
Calabrese Vincenzo, Santos Tatiana P, Haward Simon J.、Shen Amy Q.

２．発表標題
A unified framework to describe shear- and extension-induced alignment of macromolecules of various flexibility

３．学会等名
Workshop on Recent Trends in Microrheology and Microfluidics

４．発表年
2022年

１．発表者名
Santos Tatiana P.、Calabrese Vincenzo、Boehm Michael W.、Baier Stefan K.、Shen Amy Q.

２．発表標題
Flow-induced alignment of protein nanofibril dispersions

３．学会等名
Workshop on Recent Trends in Microrheology and Microfluidics

４．発表年
2022年

１．発表者名 Calabrese Vincenzo、Varchanis Stylianos、Haward Simon J.、Shen Amy Q.
２．発表標題 Alignment of colloidal rods in crowded environments
３．学会等名 Annual Meeting of The Society of Rheology (SOR), Chicago, USA（国際学会）
４．発表年 2022年

１．発表者名 Santos Tatiana P.、Calabrese Vincenzo、Boehm Michael W.、Baier Stefan K.、Shen Amy Q.
２．発表標題 Flow-induced alignment of protein nanofibril dispersions
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１．発表者名 Calabrese Vincenzo、Varchanis Stylianos、Haward Simon J.、Shen Amy Q.
２．発表標題 Alignment of colloidal rods in crowded environments
３．学会等名 nnual European rheology conference (AERC) 2022, Seville, Spain.（国際学会）
４．発表年 2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
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６．研究組織（つづき）

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
研究協力者	サイモン　ハワード (Haward Simon)		

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関