News Release

金沢大学
KANAZAWA UNIVERSITY

令和８年５月１２日

各報道機関文教担当記者　様

時間とともに構造が変化する高分子材料を開発
―自然界の「らせん構造」に着想―

金沢大学融合研究域融合科学系の堀優太准教授は、千葉大学、キール大学、静岡大学、立命館大学の共同研究チームの一員として、自然界の DNA やタンパク質に見られる「らせん構造」に着想を得て、**時間の経過とともに自律的に構造を成熟させる新しいクロロフィル由来の超分子ポリマーを開発しました。**

本材料は、形成直後に構造が固定されることが多い従来の合成材料とは異なります。クロロフィル誘導体が水素結合により環状に集合した「ロゼット」が積み重なることで、まず非らせん状の繊維を形成します。その後、数日かけて段階的に構造が変化し、ピッチの異なる 3 種類のらせん構造（HF1：26 nm、HF2：13 nm、HF3：8 nm）へと自発的に移行する特徴的な動的挙動が確認されました。

この構造変化は、ポリマーが一度分解して再集合するのではなく、超分子ポリマー内の一部で生じた構造変化が隣接する領域へと連鎖的に伝わる「協働的」な再組織化によって進行することが、原子間力顕微鏡（AFM）や中性子小角散乱を用いた解析によって明らかになりました。さらに、AFM 観察では、右巻きのらせん構造が確認される一方、円二色性スペクトルによる光学解析では、左巻きに相当する信号が得られました。この一見矛盾する結果について、ロゼットは単に回転しながら積み重なるのでなく、横方向の「ずれ」を伴って積層する「クリーパー・ヘリックス」構造をとることが示され、この構造的な「ずれ」がエキシトン結合の符号反転を引き起こす要因であることが、エキシトン理論に基づく計算シミュレーションによって説明されました。

本研究で示された、**複数のエネルギー状態を段階的に経由させる分子設計の考え方は、時間の経過に応じて光学的・電気的特性をプログラム通りに変化させる、高度な適応型材料の開発に向けた、新たな可能性を示すものです。**

堀准教授は、本研究において、特に理論計算による局所構造探索や光学解析に貢献しました。

本研究成果は、2026 年 4 月 20 日付で国際学術誌『*The Journal of the American Chemical Society*』のオンライン版に掲載されました。

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RESEARCH NEWS STORY

April 27, 2026
Chiba University
Shizuoka University
Ritsumeikan University
Kanazawa University

Mimicking Nature's Twist: Time-Evolving Helicity in a Polymer

A chlorophyll-based polymer shows stepwise evolution from nonhelical to helical structures, offering a new route to adaptive materials

Synthetic materials rarely mimic the dynamic helicity observed in biological systems like DNA and proteins, often forming fixed structures early in assembly. Inspired by this adaptability, researchers at Chiba University, Keele University, Shizuoka University, Kanazawa University, and Ritsumeikan University developed a chlorophyll-based supramolecular polymer that gradually evolves from nonhelical fibers into helical structures through intermediate stages. This stepwise, cooperative transformation offers a new strategy for designing adaptive materials with tunable optical and electronic properties.

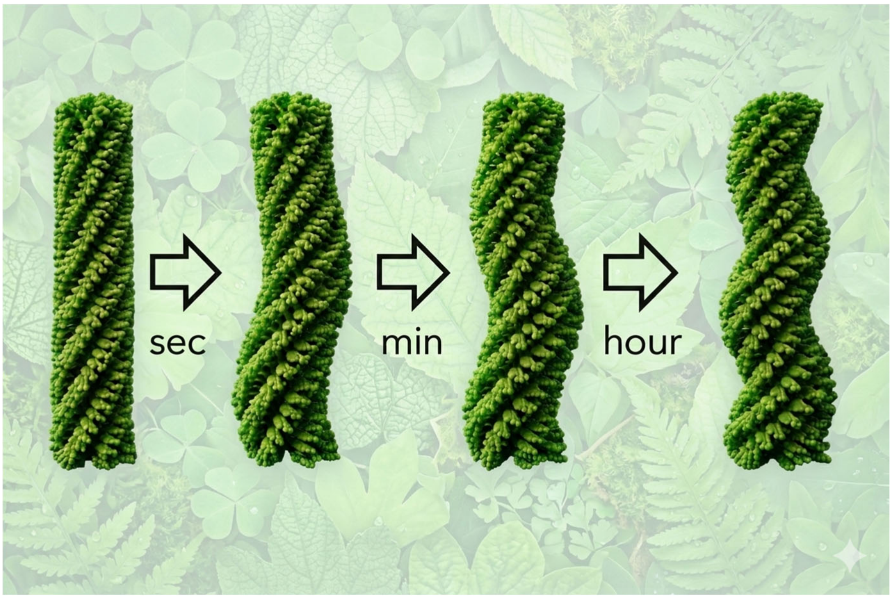

Image title: Time-Evolving Helicity in Chlorophyll-Based Polymers
Image caption: The figure shows how the material evolves from nonhelical fibers into progressively tighter helices over several days. This stepwise transformation demonstrates dynamic helicity in synthetic systems, where structure develops gradually rather than forming instantly.
Image credit: Professor Shiki Yagai from Chiba University, Japan
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Science has long taken inspiration from the natural world, and few natural designs are as iconic as the helical shape that makes life possible. The best-known example of such a molecule is DNA, a double helix that carries the genetic instructions for all living organisms. Similar helical shapes are also found in proteins. This shape is special in that it imparts a certain adaptability to biological molecules. For instance, by changing how tightly they twist or even the direction of their twist, biological systems can respond and adapt to their environment. This helps proteins adjust their shapes to fold correctly and perform essential tasks.

Inspired by this design, researchers from Chiba University, Shizuoka University, Keele University, Kanazawa University, and Ritsumeikan University, Japan, have developed a chlorophyll-based supramolecular polymer that can gradually transform from nonhelical fibers into well-defined helical structures over time.

The study was led by Professor Shiki Yagai at the Institute for Advanced Academic Research, Chiba University, along with Balaraman Vedhanarayanan and Ryoma Tsuchida from the Graduate School of Engineering, Chiba University; Shinnosuke Kawai from Shizuoka University; and Martin J. Hollamby at Keele University, UK. The study was published online in the *Journal of the American Chemical Society* on April 20, 2026.

"Examples of synthetic supramolecular polymers in which multiple helicity arises dynamically from kinetically trapped, nonhelical structures are rare," says Prof. Yagai.

The developed molecule overcomes this limitation. Instead of forming a helix immediately, it evolves step by step, passing through several intermediate stages before reaching its final helical form. It begins as a nonhelical fiber, then develops two loose helices, and finally tightens into a more twisted structure.

The researchers synthesized a chlorophyll derivative functionalized with barbituric acid groups and long alkyl chains. These molecules assemble into ring-like structures called rosettes through hydrogen bonding. In low-polarity solvents, the rosettes stack into long, one-dimensional fibers. The large and complex structure of each chlorophyll unit prevents the rosettes from immediately arranging into a stable configuration. As a result, the system first forms nonhelical fibers, which gradually reorganize into helices with tighter twists over time.

Using atomic force microscopy, the team identified four distinct fiber types: a nonhelical form (NF), in which rosettes are stacked directly without offset, and three helical forms (HF1, HF2, and HF3) that arise from slight translational shifts between stacked rosettes, resulting in twisted structures. All three helices are right-handed but differ in pitch: 26 nm for HF1, 13 nm for HF2, and 8 nm for HF3.

Using advanced imaging techniques, the team then tracked how these structures evolved over time. Starting with a solution dominated by nonhelical fibers, they observed a gradual transformation into helical structures over the course of several days. Within the first 30 minutes, most nonhelical fibers disappeared, giving way to HF1 and HF2. Over the next few hours, HF1 was converted almost entirely into HF2. The final transformation, from HF2 to the most tightly twisted form, HF3, occurred much more slowly, taking several days.

The researchers also found that this transformation occurs cooperatively. Once a small region of a fiber adopts a more stable helical structure, it promotes similar changes in neighboring regions, allowing the transformation to spread along the polymer.

"We demonstrate that helicity in a one-dimensional supramolecular polymer can emerge and mature through discrete, cooperative reorganizations occurring within the polymer backbone across a rugged energy landscape, representing a rare behavior," says Prof. Yagai.

These findings also point to a blueprint for designing dynamic helical structures. By creating molecular building blocks that can adopt multiple stable arrangements with only small energy differences, it may be possible to design materials that change their structure over time in a controlled way.

Looking ahead, the team notes that an important question remains: whether these structural changes occur randomly along the fibers or propagate in a directional manner from specific starting points. Understanding this process could help scientists design materials that more closely mimic the adaptability seen in nature.

To see more news from Chiba University, click here.

About Professor Shiki Yagai

Professor Shiki Yagai is a well-established researcher at Chiba University, Japan. In 2002, he received his Ph.D. from Ritsumeikan University, Japan. He joined Chiba University as an Assistant Professor and was promoted to full-time professor in 2017. He has authored over 200 publications in the fields of molecular self-assembly, supramolecular polymers, and functional dyes, with a focus on nanostructural control. He leads the Grant-in-Aid for Transformative Research Areas (A) project titled "Materials Science of Meso-Hierarchy," which spans from 2023 to 2027. This project focuses on exploring the hierarchical structures of materials to develop innovative functional materials at the mesoscopic scale.

Funding:

This work was supported by the Japan Society for the Promotion of Science (JSPS) KAKENHI (grant numbers: 22H00331, 23K19237, and 23H04873) under the Grant-in-Aid for Transformative Research Areas "Materials Science of Meso-Hierarchy." It also used the SasView application, originally developed under NSF award DMR-0520547 and further supported by the European Union's Horizon 2020 research and innovation program through the SINE2020 project (grant number: 654000).

Reference:

Title of original paper: Sequential, Multistep, and Cooperative Helicity Evolution in Supramolecular Polymers of Chlorophyll Rosettes

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Journal: *Journal of the American Chemical Society*
DOI: 10.1021/jacs.6c03125

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