

Complex molecules that fold like proteins can emerge spontaneously.

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Rok wydania

2019

Czasopismo

Journal of the American
Chemical Society

Numer woluminu

141

Strony

1685-1689

DOI

10.1021/jacs.8b11698

Kolekcja

Naukowa

Język

Angielski

Typ publikacji

Artykuł

Streszczenie

Folding can bestow macromolecules with various properties, as evident from nature's proteins. Until now complex folded molecules are the product either of evolution or of an elaborate process of design and synthesis. We now show that molecules that fold in a well-defined architecture of substantial complexity can emerge autonomously and selectively from a simple precursor. Specifically, we have identified a self-synthesizing macrocyclic foldamer with a complex and unprecedented secondary and tertiary structure that constructs itself highly selectively from 15 identical peptide-nucleobase subunits, using a dynamic combinatorial chemistry approach. Folding of the structure drives its synthesis in 95% yield from a mixture of interconverting molecules of different ring sizes in a one-step process. Single-crystal X-ray crystallography and NMR reveal a folding pattern based on an intricate network of noncovalent interactions involving residues spaced apart widely in the linear sequence. These results establish dynamic combinatorial chemistry as a powerful approach to developing synthetic molecules with folding motifs of a complexity that goes well beyond that accessible with current design approaches. The fact that such molecules can form autonomously implies that they may have played a role in the origin of life at earlier stages than previously thought possible.

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Adres publiczny

<http://dx.doi.org/10.1021/jacs.8b11698>

Strona internetowa wydawcy

<https://www.acs.org/content/acs/en.html>

Plik został wygenerowany dnia 2026-10-06 15:09:05

Adres w repozytorium <https://old.chem.uni.wroc.pl/pl/repozytorium/wb8ZqDf>.