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Supplementary information

Self-assembly of emulsion droplets through programmable folding

In the format provided by the authors and unedited

Supplementary Information

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Description of supplementary videos

The supplementary videos 1 - 7 are representative experimental videos of the $N = 6$ 'triangle,' 'chevron,' and 'ladder', the $N = 7$ 'rocket #2,' the $N = 8$ 'hourglass', the $N = 9$ 'poodle,' and the $N = 10$ 'crown' foldamers. Each movie was captured using its respective protocol listed in Figure 3(a) in the main text. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. Videos are sped up 100x from real time. All droplets in videos 1-7 are approximately 2.8 μm in diameter. The supplementary video 8 is a representative simulation video of the $N = 10$ 'crown' foldamer. Supplementary videos 9 and 10 show a 3D polytetrahedron foldamer in experiments and simulation, respectively.

Supplementary video 1 demonstrates the step-wise folding of a $N = 6$ polymer into the 'triangle' configuration. The blue-blue interaction is turned on first (at 40° C), then the blue-yellow interaction is switched on (at 32° C) to complete the triangle. Supplementary video 1 also demonstrates the unfolding of the polymer after the temperature is raised after successful folding. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 2 demonstrates the step-wise folding of a $N = 6$ polymer into the 'chevron' configuration. The blue-yellow interaction is turned on first (at 32° C), then the yellow-yellow interaction (at 27° C) is switched on to complete the chevron. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 3 demonstrates the folding of a $N = 6$ polymer into the 'ladder' configuration. The blue-blue interaction is turned on first (at 40° C), then the yellow-yellow interaction is switched on (at 27° C) to complete the ladder. The order of interactions for this protocol only changes the fold of the ladder. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 4 demonstrates the step-wise folding of a $N = 7$ polymer into the 'rocket' configuration. First, the blue-blue interaction is turned on (at 40° C). The blue-yellow interaction is switched on (at 32° C) to form a structure that resembles a floppy Christmas tree. Finally, the yellow-yellow interaction is switched on (at 27° C) to complete the blue-core fold into the 'rocket' configuration. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 5 demonstrates the folding of a $N = 9$ polymer into the 'poodle' configuration. Only the blue-blue interaction is turned on (at 40° C) and the polymer folds into the 'poodle' configuration. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 6 demonstrates the folding of a $N = 8$ polymer into the 'hourglass' configuration. Only the blue-blue interaction is turned on (at 40° C) and then the blue-yellow interaction is switched on (at 32° C) and the polymer folds into the 'hourglass' configuration. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 7 demonstrates the step-wise folding of a $N = 10$ polymer into the 'crown' configuration. First, the blue-blue interaction is turned on (at 40° C), then the blue-yellow interaction is switched on (at 32° C), and finally the yellow-yellow interaction is switched on (at 27° C) to complete the fold into the 'crown' configuration. The duration of each video frame is 10 seconds, with a frame rate of 10 frames per second. The video is sped up 100x from real time. Scale bar is 5 μm.

Supplementary video 8 shows the molecular dynamics simulations of the folding of a decamer chain following two different scenarios, during the three step protocol for the 'crown' (only two steps are shown in the video). On the video to the left (pure downhill), the blue-blue interaction is turned on infinitely fast. Bonds form irreversibly, and the chain gets quickly trapped in a local minimum. In contrast, the video to the right shows a slow temperature quench (thermalization) where bond rearrangements are possible. This favours the collapse of the chain into a blue core, allowing it to further fold into the correct 'crown' geometry. The waiting time on the blue-blue interaction is $\tau = 1000$ time units.

Supplementary video 9 An alternating sequence hexamer can be folded into a 3D polytetrahedron foldamer. Starting from a 1D chain, both our experiments and simulations measure a 100% yield of the polytetrahedron, when the following protocol is used: first the blue-blue interaction is turned on, then blue-yellow, and finally yellow-yellow. The duration of each video frame is 5 seconds, with a frame rate of 10 frames per second. The video is sped up 50x from real time. Scale bar is 5 μm.

Supplementary video 10 A polytetrahedron foldamer with an alternating sequence realized in simulations. Interactions are turned on every $\tau = 500$ simulation time units.