AFRL-AFOSR-VA-TR-2019-0199

Crumpled and Prescriptively Folded Polymer Films for Advanced Lightweight Materials

Andrew Croll
NORTH DAKOTA STATE UNIVERSITY

08/01/2018
Final Report

DISTRIBUTION A: Distribution approved for public release.

Air Force Research Laboratory
AF Office Of Scientific Research (AFOSR)/ RTA1
Arlington, Virginia 22203
Air Force Materiel Command

DISTRIBUTION A: Distribution approved for public release.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing the burden, to Department of Defense, Executive Services, Directorate (0704-0188). Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.
PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ORGANIZATION.

1. REPORT DATE *(DD-MM-YYYY)*	2. REPORT TYPE	3. DATES COVERED *(From - To)*
23-07-2019	Final Performance	01 May 2015 to 30 Apr 2018

4. TITLE AND SUBTITLE
Crumpled and Prescriptively Folded Polymer Films for Advanced Lightweight Materials

5a. CONTRACT NUMBER

5b. GRANT NUMBER
FA9550-15-1-0168

5c. PROGRAM ELEMENT NUMBER
61102F

6. AUTHOR(S)
Andrew Croll

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
NORTH DAKOTA STATE UNIVERSITY
1919 UNIVERSITY DR N, STE 102
FARGO, ND 58102-1843 US

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AF Office of Scientific Research
875 N. Randolph St. Room 3112
Arlington, VA 22203

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/AFOSR RTA1

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-AFOSR-VA-TR-2019-0199

12. DISTRIBUTION/AVAILABILITY STATEMENT
A DISTRIBUTION UNLIMITED: PB Public Release

13. SUPPLEMENTARY NOTES

14. ABSTRACT
Over the course of this grant we performed mechanical experiments designed to uncover fundamental relations between materials and geometry in light-weight structures created from thin polymer films. Our work has lead to new understanding of the role of defects in bent structures, how fluids can deform thin solids and how the strength of crumpled films can be understood. The latter point will be of considerable importance, because we have supplied quantitative structure property relations for crumpled matter for the first time. Such relations are a necessary prerequisite for their use in real world applications.

15. SUBJECT TERMS
Folded Polymer Films, Crumpled and Folded Composite Reinforcements

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			TILEY, JAIMIE
Unclassified	Unclassified	Unclassified	UU		19b. TELEPHONE NUMBER *(Include area code)* 703-588-8316

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. Z39.18

DISTRIBUTION A: Distribution approved for public release.

Contract/Grant Title: Crumpled and Prescriptively Folded Polymer Films
Contract/Grant #: FA9550-15-1-0168
Reporting Period: 1 May 2015 to 30 May 2018

Over the past 3 years we have used the YIP funding granted by the AFOSR to create a basic of physical understanding of how thin, bent polymer films interact mechanically with their environment. The work has led to several important insights, including the importance of adhesion(friction) between sheets in complex folded structures, the plausibility of a reductionist approach to design, and a more detailed understanding of localized lengthscales in thin films. We have one paper published, one under review, one more submitted, 4 currently being written and 3 more in a rough stage of completion. The grant allowed 1 Ph.D. student to complete his degree, contributed to training another Ph.D. student, allowed one Masters student to get her degree and move to the Ph.D. level, and contributed to 3 undergraduate research careers. In total the work supported 17 talks at national and international settings. Below we outline our progress towards the goals set forth in the grant.

The first work we proposed was examining the basic material properties of the polymer films used in our work by developing 'origami' friendly mechanical tests. The test we employed is intentionally trivial; a film is simply bent between two parallel walls and force/displacement and force/time curves are recorded. It is essentially a direct measure of the material's bending modulus, the dominant material property of slender structures. We measured the modulus of 3 materials in this manner, polystyrene (PS), polycarbonate (PC) and Sylgard 184 (polydimethylsiloxane, PDMS), and noted a logarithmic time dependence of the force relaxation in PDMS and PC. The logarithmic time dependence was found to be thickness dependent, in agreement with a diffusive stress relaxation mechanism (in agreement with the literature). This simple characterization is important because the relaxation of ***any origami structure*** will include this basic, thin-film contribution which is rarely noted in the literature.

To add complexity, we introduce a second bend to the material and test in a similar manner. The second bend is orthogonal to the first, creating a single localized structure known as a developable cone (d-cone). This new structure can be seen when one folds a sheet of paper twice, such that its projection is now a quarter size of the original sheet. The d-cone is the point where the two folds meet in the center of the sheet. In this case we have found that, remarkably, the d-cone is completely ignorable – it contributes only a vanishing energy to the continued collapse of the sheet, or its force relaxation. Microscopic investigation of the different materials, however, shows important differences in the resulting structure. PDMS films show smooth elastic behavior at the d-cone core, PC shows irreversible plastic flow, and PS films fracture at the d-cone core resulting in a hole through the film (a massive topological change). These first observations form the basis of the successful Masters thesis, and are currently being written up by the student.

One facet of this set of experiments required exploring different methods of assembling very thin films, which are difficult to manipulate by hand. Often assembly can be facilitated through the surface tension of a fluid, in a guided assembly process known as capillary origami. The basic mechanics of the method are easy to understand: 1.) a two dimensional shape is cut in a thin film. 2.) the surface tension of the fluid/air interface of a fluid drop placed on the thin film will cause the film to bend, ultimately wrapping the drop. 3.) the fluid is allowed to evaporate leaving an assembled shape. When working with the

DISTRIBUTION A: Distribution approved for public release.

technique we noticed two broad unanswered questions. What exactly happens at the contact line, and what if the films 'stick' to their substrates?

Our subsequent studies of the physical details governing the contact line of a fluid on a slender structure resulted in a publication (submitted to *Soft Matter*). More importantly, the work is a timely addition to an ongoing debate of how soft substrates are deformed by fluids (elastocapillarity). We are currently working on another publication related to the influence of adhesion. The basic idea is to explore how this additional force is accommodated by the thin films, and use the resulting knowledge to design new origami structures only made possible by adhesive interactions in a thin film.

A final product of our exploration of thin bent films is a set of experiments which investigated the influence of roughness on thin film bending. Here a film was patterned into regions of high and low modulus, and then mechanically deformed. The work was published in *Soft Matter*, and made the cover of the journal.

The next goal of this proposal was to study the internal dynamics of thin (<100 nm) polymer films using a fluorescent molecule as a proxy for local dynamics. The experiment involves a 'knife edge' indentation of a soft elastomer which supports a thin polymer film. The film has an incorporated fluorescent molecule, which we photobleach with a polarized light source. We then stress the film (with the indenter) and observe the photobleached segment of film in two orthogonal polarization states. The recovery of intensity in the photobleached segment of polymer film is then related to the internal mobility of the polymer as a function of its stress and temperature. We have completed the mechanical part of this experiment with the help of several undergraduate students, but due in part to a graduate student leaving the group the experiment is incomplete. The essential problem is that we are not properly accounting for the inherent non-linearity of the photomultiplier tubes we use as detectors (I don't believe we are reliably able to compare intensities between the two channels). We will continue to work on this experiment until we have the details perfected.

The final goal of our YIP proposal was to examine crumpled polymer films from a 'materials' perspective. The basic idea is that crumpled thin films represent a unique and possibly useful lightweight material system, but can only be used if there are reliable material/property relations available. In this area we have made considerable progress towards understanding this basic physics. We began by testing force-displacement curves of crumpled PC and PDMS films confined by two parallel walls. We noted that both the elastic (PDMS) and the plastic (PC) materials showed power law behaviour (observed previously) and both materials showed considerable hysteresis between indentation and retraction of the confining walls. This is expected of the plastic material (energy is dissipated in the sharp folds) but is not expected of an elastic material such as PDMS. We have since proven that the hysteresis in the PDMS crumples is due to adhesion between the sheets by coating the rubber films with micron scale solid particles and through theoretical analysis.

Two competing theories are currently available for analysis of the crumpled films. The first, based on stretching ridges (two d-cones connected by a section of film) predicts that force follows a particular fixed power of displacement ($F \sim x^a$, where F is force, x is displacement and a is a fixed constant). The second model is based on energy storage in sharp folded regions of film and also predicts a power law relationship between compressive force and displacement but shows that the power law exponent, a, varies depending on details of the structure. Sparing the details of our analysis, we have found both

DISTRIBUTION A: Distribution approved for public release.

models are insufficient to describe the compression data we measure. We propose an empirical relation, very similar to the second model, which ***quantitatively*** captures the behaviour of both materials. We also show how relaxation in the crumpled structure is directly related to the material relaxation (as measured for single bends above) and has nothing to do with the particular structure present. This is important; it means crumple dynamics are predictable from material properties alone and have little to do with the actual structure.

Beyond our basic goal to supply structure-property relations, our experiment and model allow us to quantify the importance of adhesion in the crumpled structures. Adhesion has been completely ignored in complex folded structures, being assumed to be only a weak contribution. To the contrary, we find presence of adhesion to change the magnitude of the compressive force by an order of magnitude. This work has been submitted to Physical Review Letters, and we are hopeful reviewers will agree our belief of the importance of our work. Finally, the work allows us to scale down to nanometer thickness films. We believe that dynamics will change upon reaching sub 100nm thicknesses which we should be able to measure reliably (we are in the process of doing so).

Finally, we have been exploring the interaction of two d-cones in thin films (a stretching ridge, one of the four possible building blocks for any origami structure). The work is motivated in part by theoretical models of crumpling, and in part by the basic idea that the number of d-cones present in a sheet should be a quantity minimized during collapse. In other words, why have two d-cones if only one is necessary? We again use confocal microscopy and force measurement to monitor structure during the compression of a single 'ridge' in a sheet of PDMS or PC. Behaviour is complex, and doesn't seem to follow current models. For example, we see evidence of a 'snap-through' instability in the elastic materials (two d-cones 'jump' together during compression), which has not been noted before. We also note two failure modes for the plastic PC films. Here the d-cones are 'pinned' by plastic deformation in their cores and don't move during compression. The PC films collapse either through a 'cascade' of crumpling from the original d-cones towards the center of the structure, or through a 'pure' buckling in the center of the ridge. We are currently writing a paper aimed at a top journal describing the effects.

We have been active in demonstrating our work to the public at every possible opportunity. The relation of our work to origami makes it very accessible; in the past we have talked with boy scouts troop and kindergarten classes for example. While the children may not deeply understand the science and engineering, they are very excited by the 'fun' that you can have when bending paper. We hope that this inspiration, provided by the Air Force Office of Scientific Research, will lead them to consider careers in science, engineering and the defence of the nation down the road.

DISTRIBUTION A: Distribution approved for public release.