Buckling-driven self-formation of microchannels
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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2011

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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1.1 Nano and microfabrication technology

Miniaturization is the continuous downsizing of components and devices for applications in micro-electronics, optoelectronics and micro/nano-electro mechanical systems (NEMS, MEMS). The beginning of the twentieth century was very important in modern human history with the advent of electronics and computers. Exceptional developments in micro-electronics enabled computers to be generally accessible and thus laying the foundation for modern technology. The tendency towards building small-scale components and products in all fields of science (mechanical, optical, electronic, chemical or even biological) has become increasingly desirous, allowing products and devices to be easier to carry, easier to store, and much more convenient and efficient to use. As a result of the quest to build smaller structures, the field of nano- and microfabrication technology emerged. Specialized fabrication strategies have been developed in the last three decades to realize micro- and nanosystems, such as photo-lithography [Okazaki, 1991], focused ion beams [Moon et al., 2007] and soft lithography [Xia and Whitesides, 1998]. The continuous downsizing of consumer electronics (e.g. smart phones) is the outcome of the considerable progress made in this field.

Much attention has been focused on generating ordered surface patterns, wrinkles and channels at the nano- and micron scale with controllable dimensions. Recent developments in this area has lead to a wide range of fascinating applications such as tunable optical gratings [Edmondson et al., 2006; Edmondson and Huck, 2004; Xia et al., 1996], flexible electronics [Baca et al., 2008; Hsu et al., 2002a; Jiang et al., 2007b; Kim et al., 2008; Lacour et al., 2003; Sun et al., 2006], micro-reactors [Watts and Wiles, 2007], micro- and nanofluidic channels [Malachias et al., 2008; Mei et al., 2007], actuation/sensing devices [Comrie and Huck, 2008], particle separators [Efimenko et al., 2005], templates for microstructure fabrication [Huck et al., 2000; Peng et al., 2004; Schäffer et al., 2000], scaffolds for tissue engineering [Langer and Vacanti, 1993] and surfaces for marine anti-fouling [Efimenko et al., 2009].
1. Introduction

1.2 Surface patterning by buckling

Almost all methods employed in generating patterns/order at small length scales rely on the three step process of conventional lithography, thin film deposition and etching. In the search for alternative approaches, a new method based on the buckling of pre-stressed thin films has emerged with promising features compared to conventional lithography based techniques. Buckling, wrinkling and other mechanical instabilities have long been considered as besetting phenomena to be avoided for better performance of structures. However, buckling can also be exploited for the well-controlled generation of regular surface patterns in the microand nanometer range [Bowden et al., 1998]. The spontaneous formation of ordered structures through buckling eliminates the necessity of pre-patterning in contrast to lithography-based methods, thus evolving as a promising cost and time-efficient alternative approach. It is worth noting that the self-assembly or self-organization of order at length scales much smaller than the induced feature sizes is an appealing feature of this technique. Many different ways of buckling-driven pattern and channel formation have been explored in the literature. Blisters of various morphologies (straight, circular and telephone-cord) can be formed through buckling-driven delamination of pre-stressed thin films from a rigid substrate [Gille and Rau, 1984; Moon et al., 2004; Ortiz and Gioia, 1994]. Wrinkle formation can also be achieved when the film does not delaminate “from”, but buckle “into” the substrate. In this case, the substrate is compliant and pre-strained by heating or stretching before the stiff film is attached [Bowden et al., 1998; Huck et al., 2000; Jiang et al., 2007a], thus inducing compressive stresses in the film. The generation of anisotropy in the pre- or postbuckling stress state allows for a preferred direction of buckling, causing ordered ridging patterns. A wealth of studies has been devoted to understand the mechanism of wrinkle/pattern formation in the above two methods [see e.g., Hutchinson and Suo, 1992; Ortiz and Gioia, 1994; Song et al., 2008; Suo and Hutchinson, 1990]. Recently, a new method of buckle-driven channel formation is reported, which is based on the partial release of pre-stressed thin films from a rigid substrate by chemical etching [Malachias et al., 2008; Mei et al., 2007] or electrolysis [Edmondson et al., 2006; Edmondson and Huck, 2004], causing the films to buckle-up, followed by bond back of the film onto the substrate. In this method a thin film is grown on a substrate during which eigenstrains are induced, causing compressive film stresses to develop. Next, the film-substrate system is exposed to an external stimulus (chemical etching or an electric pulse) which reduces the interface strength between the film and the substrate, allowing the film to release its eigenstrains through buckling from the substrate. Finally, the system is left for drying, during which the partially buckled-up film bonds back to the substrate due to cohesive forces of attraction between the film and the substrate, freezing in the final geometry of the channel. Despite its success in generating well-controlled ordered channels of (sub)micrometer/nanometer dimensions, the underlying mechanics of pattern formation is not yet well understood. This will be the focus of this thesis.
1.3 Objective and outline of the thesis

The objective of this thesis is to develop a fundamental understanding of the mechanism of channel formation based on the release and bond back of pre-stressed thin films on rigid substrates. We investigate how various system parameters such as the film geometry (film size, thickness and shape), film properties (elastic modulus $E$, Poisson’s ratio $\nu$ and eigenstrain $\varepsilon^*$), interface energy $\Gamma$ and the method of interface strength reduction (uniform electrolysis versus directional etching) control the final channel geometry (channel morphology, height and width). We will carry out a dimensional analysis to identify the key dimensionless parameters that fully capture the channel formation process in order to provide insight for design. The thesis is organized as follows.

Chapters 2 and 3 deal with the formation of line buckles in polymer films by the electrolysis process. In this chapter a two-dimensional finite element model is developed to describe the mechanics of buckling-driven delamination of the film from the substrate and the process of draping back. A cohesive zone model is implemented to mimic the interface cohesion between the film and the substrate during delamination and bond-back. Buckling-up will be studied through a ‘2-step’ process in which first the eigenstrain is applied and subsequently the interface strength reduced. The results will be compared to the well-known process of buckling-driven delamination (a ‘1-step’ process) in which the interface delaminates instantaneously when eigenstrains are applied. Finally, the bond-back process is analyzed and the governing dimensionless parameters are identified.

In chapter 4 we describe the self-organization of linear nanochannel formation by releasing the eigenstrains through directional under-etching of a sacrificial layer between the film and substrate. Analytical calculations are carried out to obtain the critical buckling strain and wavelength at the onset of buckling. We use three-dimensional finite element simulations to study the evolution of the buckling wavelength and height as a function of eigenstrain for different etch depths. Finally, bond-back simulations are performed to obtain the final channel morphologies in terms of channel spacing, channel width and height. The results of the model are compared with the experimental observations, showing good agreement.

In chapter 5 we study the mechanics of pattern formation in pre-stressed polygonal films. Three-dimensional simulations are carried out to study the evolution of the different deflection patterns as a function of eigenstrain during buckling-up and as a function of interface strength during bond-back. For rectangular films the results are compared to analytical solutions based on minimization of energy. Films of different shape (square, rectangular, pentagonal and hexagonal) are analyzed and the results are captured in scaling relations.

Chapter 6 describes the mechanics of nanochannel network formation for the situation in which the etchant solution enters the system through periodically-spaced pre-defined etch pits. First, we study the evolution of deflection patterns around a single etch-pit. The evolution of wavelength is compared with the master curve of linear channel simulations studied in chapter 4, showing close correspondence. A semi-analytical method has been developed to describe the transition of channel morphology from edge channels to corner channels as a function of film geometry, thickness and eigenstrain. Next, we study the channel evolution in a $3 \times 3$ etch-pit array. The networks are formed in InGaAs films, which have a very strong
interaction with the substrate during the etching process unlike the Si-Ge systems as studied in chapter 4. We show that the presence of film-substrate interaction is essential to account for the complex nanochannel networks reported in the experiments.

Finally, in chapter 7 we summarize the results obtained in this thesis, draw conclusions and present an outlook for future developments.