Acta Materialia 127 (2017) 220–229

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Size dependent morphologies of brittle silicon nitride thin films with combined buckling and cracking

Sen-Jiang Yu [a], Shi-Chen Li [b], Yong Ni [b, *], Hong Zhou [a]

[a] Department of Physics, China Jiliang University, Hangzhou 310018, PR China
[b] Department of Modern Mechanics, CAS Key Laboratory of Mechanical Behavior and Design of Materials, University of Science and Technology of China, Hefei, Anhui 230026, PR China

ARTICLE INFO

Article history:
Received 6 December 2016
Received in revised form
20 January 2017
Accepted 20 January 2017
Available online 24 January 2017

Keywords:
Thin film
Buckling
Delamination
Cracking
Residual stress

ABSTRACT

We report on size-dependent morphological characteristics of buckle-driven delaminations in large-scale brittle silicon nitride films in accompany with ridge cracking. The buckling morphologies fall into four distinct categories in a phase diagram. They start as straight-sided blisters with or without ridge cracks from the film edge and then spread as telephone cord blisters with or without ridge cracks into the central region of the sample gradually. The buckle-delamination size is found to decrease with the increase of residual stress, different from the previous reports. Theoretic analysis based on Föppl-von Kármán plate theory with strong mixed-mode interfacial adhesion elucidates this abnormal behavior and explains why the ridge-cracked straight-sided blister always appears with much smaller delamination size in comparison with the coexisting uncracked telephone cord blister with large delamination size. The ridge-cracked buckle-delaminations destabilized into bubble-like blisters are also recovered by dynamic simulations based on phase field modeling, in good agreement with the experimental observations.

1. Introduction

Thin films deposited over substrates are widely used in various technological applications, such as protective coatings [1], interconnect structures [2] and flexible electronic devices [3,4]. Their reliabilities are critically influenced by typical failure modes driven by residual stresses [5,6]. If the residual stress is compressive it develops surface wrinkles [7] or buckle-delaminations [6,8], while if the residual stress is tensile it develops channel cracks [6]. Interestingly, when the film is brittle, i.e., hard transparent indium-tin oxide (ITO) coating on optical polymers [3], its failure modes under stress are strongly coupled, leading to rich and intriguing morphologies. For example, when the film is subject to a uniaxial stretch, parallel longitudinal cracks (perpendicular to the stretch direction) form in accompany with the transverse buckling of the film stripes by the compression due to the effect of Poisson's ratio [9–11]. If the film undergoes a uniaxial compression, the film develops longitudinal buckling perpendicular to the compression direction and transverse cracking perpendicular to the buckles [12]. It is further reported that when the film on soft elastic substrates is under cyclic uniaxial compression/tension, transverse/longitudinal cracks and longitudinal/transverse buckles form, resulting in complex coexisting buckling and cracking patterns [13]. Besides the Poisson effect, strong coupling between buckles and cracks can also be caused by a rapid rise of a local bending induced tensile strain, where ridge cracks or bottom cracks can form during the formation of buckle-delamination blisters [14–23]. To improve the mechanical reliability of these film structures, it is highly desirable but still challenging to reveal how the interplay between buckles and cracks results in these morphological changes that indicate complex failure paths.

Buckling the film into the third dimension would redistribute the elastic energy between the stretching and bending modes of deformation and produce indirect nonlinear elastic interaction between two inclusions [24]. In the cracked graphene, molecular simulations and theoretic analysis show that the out-of-plane distortion further releases in-plane elastic deformation and leads to Griffith strength reduction [25]. Buckling mode of deformation not only influences the Griffith strength that characterizing the onset of crack motion but also modifies the subsequent crack propagation path [26–32]. The spontaneous ordering interlocking crack pattern observed in colliding floating sheets is possibly

---

* Corresponding author.
E-mail address: yni@ustc.edu.cn (Y. Ni).

http://dx.doi.org/10.1016/j.actamat.2017.01.038
1359-6454/

attributed to the delicate interplay between cracks and buckles [26,27]. More recently, prescribed out-of-plane distortion has been used to precisely regulate the paths of cracks [32]. In comparison with the advance to understand the role of buckles on cracks, it remains unclear how the cracks impact on the post-buckling profile although mechanics of ridge-cracked straight-sided buckle-delamination has been established [14–23]. Recent experiments report that the formed ridge-cracked buckles are sometimes unstable and evolve into bubbles in accompany with ridge cracking [21]. In addition, film-thickness dependent cracking and buckling behaviors have been observed in several film-substrate systems [33,34]. But the size-dependent morphologies of thin films with combined buckling and cracking are still unclear, especially when the complex coexisting modes appear in the same film sample.

In this paper, we report on the size-dependent morphologies and underlying mechanisms of buckle-driven delaminations with or without ridge cracking in brittle silicon nitride films. Due to the high hardness, corrosion resistance, oxidation resistance, high temperature resistance, chemical inertness, insulativity and transparency, the silicon nitride films are now widely used in many high-tech fields such as microelectronics, micromachining, optoelectronics, solar cells, surface modifications, hard coatings etc. For example, the silicon nitride films are frequently used as dielectric layers and capping layers in low emissivity coated glass industry. The coated glass usually needs to be tempered at a high temperature (e.g., 700 °C) before application. The tempering process can introduce a high residual compressive stress into the film, resulting in the formation of buckle-driven delaminations. The buckles always propagate from the film edge to the central region of the sample in accompany with gradual stress relief. Furthermore, the silicon nitride film is brittle and thus the buckles are susceptible to forming ridge cracks. Therefore, the annealed silicone nitride film system can provide a platform to investigate the size effect of buckle-driven delaminations with or without ridge cracking and understand the complex coexisting bucking and cracking modes.

We fabricate many samples with large scale buckling and cracking morphologies. Using optical microscopy and atomic force microscopy (AFM) measurement, we have found that the observed morphologies fall into several distinct categories, understood by a phase diagram. The behavior of buckle-delamination size versus residual stress is different from the previous prediction wherein the buckle-delamination size increases with increasing the residual compression [35,36]. Theoretic analysis based on Föppl-von Kármán plate theory with strong mixed-mode interfacial adhesion is provided to elucidate such different behavior and explain why the ridge-cracked straight-sided blister always appears with much smaller delamination size in comparison with the coexisting uncracked telephone cord blister with large delamination size. Dynamic simulations based on phase field modeling are further performed to get a closer understanding of the development after a secondary instability of metastable ridge-cracked buckle-delaminations.

2. Experimental procedure

The samples were prepared by using an off-line coating production line (Apollon G 3210/7-H, Leybold). The target was $Si_{90}Al_{10}$ rotating hollow cylinder with 99.9% purity, 3191 mm length and 152 mm diameter. The substrate was rectangular float glass with 900 mm length, 600 mm width and 6 mm thickness. Before deposition, the base pressure of the chamber was below 1.2×10^{-4} Pa. The working gas was mixture of 600 sccm N_2 and 750 sccm Ar, and the pressure was fixed at 0.8 Pa during deposition. The sputtering power was 70 kW, the moving speed of the sample was 0.5 m/min and the film thickness was about 1100 nm. After deposition, the sample was cut into small pieces with about 50×50 mm^2 in size. They were put into a 700 °C muffle furnace for 4 min and then removed from the furnace and naturally cooled in air. The overviews of the samples were taken by a common camera (Iphone 5S). The buckling morphologies were investigated by an optical microscope (Leica DMLM) equipped with a charge coupled device camera (Leica DC 300). The 3-D structures of the buckles were determined by an atomic force microscope (Dimension 3100, Veeco) operated in tapping mode.

3. Experimental results

3.1. Size-dependent buckling morphologies

Our experiment shows that the as-prepared silicon nitride film is flat and no buckling pattern can be observed. After annealing, a high compressive stress is introduced into the film due to thermal mismatch between the film and the substrate. As a result, the film is partially delaminated from the glass substrate to form various buckling patterns. An overview of the annealed sample is shown in Fig. 1. The buckled region appears frosted, because of light scattering from the uneven sample surface. The buckling patterns generally nucleate at the film edge and then spread into the central region gradually. The spreading speed is strongly dependent on the annealing time: larger annealing time corresponds to larger spreading speed. Furthermore, the spreading speed in each sample is not uniform. Generally, the spreading speed at the early stage (near the film edge) is much large, and then it decreases gradually [21]. The buckling patterns tend to align parallel to the film edge and thus they look like a series of concentric rings (see Fig. 1). The oriented growth of the buckle-delamination may be resulted from the slight compression anisotropy [8]. As we have numerically simulated, the small deviation of the isotropic compression will lead to the alignment of the buckle-delamination perpendicular to the larger compression direction. When the deviation of the isotropic compression is large, i.e. in the case of uniaxial or quasi-uniaxial compression, the straight-sided blisters become energetically favorable instead of the telephone-cord buckles. We believe that formation of the buckle delamination with concentric rings like pattern is the result of the radial compression greater than the circumferential compression. However the distribution of the residual stress is significantly dependent on the annealing history and

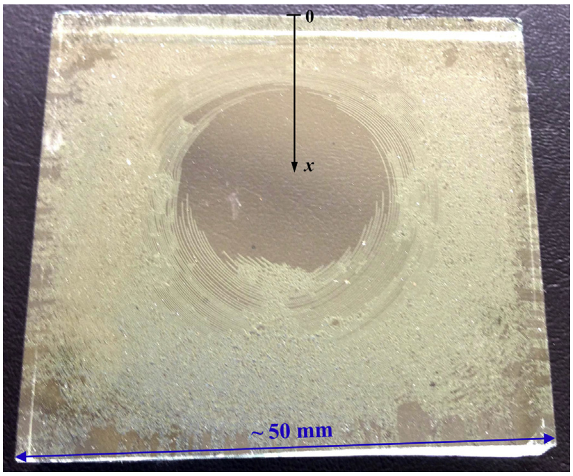

Fig. 1. Overview of an annealed silicon nitride film sample. The distance from the film edge is defined as *x*. The sample size is about 50×50 mm^2.