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Intrinsic vulnerabilities to mechanical failure in nanoscale films

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Abstract

We use molecular simulations to explore how sample dimensions and interfacial properties impact some generic aspects of the mechanical and structural behavior of nanoconfined materials. Specifically, we calculate the strain-dependent properties of minimum-energy thin-film particle configurations (i.e., inherent structures) confined between attractive, parallel substrates. We examine how the relationship between the transverse strain and the stress tensor (the equation of state of the energy landscape) depends on the properties of the film and substrate. We find that both film thickness and film–substrate attractions influence not only the mechanical properties of thin films, but also the shape and location of the "weak spots" where voids preferentially form in a film as it is strained beyond its point of maximum tensile stress. The sensitivity of weak spots to film properties suggests that nanoscale materials may be intrinsically vulnerable to specific mechanisms of mechanical failure.

Keywords: Nanoscale films; Mechanical failure; Potential energy landscape; Molecular simulations

1. Introduction

Materials confined to very small spatial dimensions behave differently than in the bulk. In addition to showing quantum confinement effects, they also display thermodynamic, kinetic, and mechanical limits of stability that depend on sample size, shape, and the characteristics of their interfaces. Specific examples of property modifications include surface-induced phase transitions, shifts of the bulk glass transition, and interface-mediated modes of mechanical failure (Gelb et al., 1999; Forrest and Dalnoki-Veress, 2001; Hutchinson and Suo, 1992). Unfortunately, because molecular-scale processes in highly inhomogeneous environments are difficult to resolve experimentally, a mechanistic picture for precisely how nanoconfinement impacts stability has been slow to develop. This presents a practical barrier to the design of technological applications, in particular those relying on solid-state nanostructures to exhibit mechanical integrity over a broad range of conditions.

In this Article, we study an elementary model system that sheds new light on how sample dimensions and interfacial properties can influence the mechanical behavior of nanoconfined materials.

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Specifically, we use molecular simulations to calculate the strain-dependent properties of mechanically-stable films of particles confined between attractive, parallel substrates. Although analogous studies have been carried out for models of isotropic materials, this is, to our knowledge, the first systematic investigation of the relationship between the transverse strain and the pressure tensor of the inherent structures (minimum potential energy configurations) of highly inhomogeneous films. Our main finding is that both sample dimensions and substrate attractions substantially influence not only the mechanical properties of thin films, but also the morphology and location of "weak spots" where voids preferentially form in a film as it is strained beyond its point of maximum tensile stress. Although the precise role that these weak spots play in dynamic deformation processes is presently unknown, they appear intimately linked to material failure by quasistatic tensile deformation. Moreover, the sensitivity of weak spots to film properties suggests that nanoscale materials may be intrinsically vulnerable to specific mechanisms of mechanical failure.

Since plastic deformation and failure are inherently dynamic events, and since the molecular-scale rearrangements that accompany them in amorphous materials are still poorly understood, molecular dynamics (MD) simulations would appear to represent an ideal theoretical tool for their investigation. In fact, MD simulation studies over the past decade have been instrumental in gaining insights into deformation processes in polymeric and small-molecule materials (Falk and Langer, 1998; Gersappe and Robbins, 1999; Rottler and Robbins, 2001, 2003; Stevens, 2001; Gersappe, 2002; Capaldi et al., 2002; Varnik et al., 2004; Van Workum and de Pablo, 2003; Yoshimoto et al., 2004). These insights have facilitated the interpretation of experiments and have aided in the introduction of simple theories for viscoplastic flow (Falk and Langer, 1998). On the other hand, despite recent advancements in algorithms for long-time dynamics, MD simulations are still limited to accessing relatively short time and length scales. Thus, it is currently computationally prohibitive to use MD to exhaustively explore the effects that sample dimensions and interfacial conditions have on the mechanisms of mechanical failure, even for simple model systems. The development of alternative methods for probing the molecular-scale origins of failure in glasses is still essential.

One complimentary approach is to calculate how the properties of a material's mechanically-stable inherent structures depend on the state of strain. This strategy has typically been implemented in one of two ways to investigate deformation, tensile strength, and failure of amorphous solids. The first method (Mott et al., 1993; Hutnik et al., 1993; Malandro and Lacks, 1997, 1998, 1999; Lund and Schuh, 2003a,b; Maloney and Lemaître, 2004a,b) calls for subjecting an ensemble of inherent structures, created at a prescribed state of strain or stress, to an athermal quasistatic deformation process consisting of alternating steps of small affine deformation followed by potential energy minimization. Malandro and Lacks (1997, 1998, 1999) have used this protocol to investigate the connection between strain-induced plastic rearrangements in amorphous materials and the annihilation of minima on the material's potential energy landscape. A similar implementation has also been used by Maloney and Lemaître (2004a,b) to study the energy fluctuations associated with amorphous plasticity and the behavior of elastic constants near the onset of material failure.

Alternatively, one can generate collections of inherent structures at each macroscopic strain state of interest by mapping equilibrium particle configurations from high-temperature simulations to their local potential energy minima (Stillinger and Weber, 1982). This procedure, which we adopt here, has been primarily employed by Debenedetti, Stillinger, and coworkers (Sastry et al., 1997; Roberts et al., 1999; Utz et al., 2001; Shen et al., 2002) to determine how the inherent structure pressure of 'bulk' glass-formers depends on density, a relationship that has been termed the equation of state of an energy landscape (EOSEL) (Debenedetti et al., 1999). Several trends have emerged from simulated EOSELs that give insights into the mechanical properties of amorphous solids. For example, inherent structures formed with densities higher than the material's Sastry density ρ_S (shown in Fig. 1a) are structurally homogeneous, whereas "weak spots" (Sastry et al., 1997) contained in lower-density equilibrium configurations develop into fissures or voids upon energy minimization. Thus, ρ_S is a material property that represents the minimum density for which mechanically-stable solid structures can remain structurally homogeneous and void free. Moreover, the corresponding isotropic tension $-p_{IS}(\rho_S)$ is the maximum amount that an inherent structure of that material can sustain prior to