Measurement of debonding in cracked nanocomposite films by ultrasonic force microscopy

A. P. McGuigan,[a] B. D. Huey, G. A. D. Briggs, and O. V. Kolosov
Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom

Y. Tsukahara and M. Yanaka
Toppan Research Institute, Toppan Printing Company Ltd., Sugito-machi, Kitakatsushika-gun, Saitama 345-8508, Japan

(Received 27 August 2001; accepted for publication 6 December 2001)

This letter reports the application of ultrasonic force microscopy (UFM) to investigate subsurface fracture mechanisms during tensile loading of nanocomposite films consisting of a brittle glass on a ductile polyethylene terephthalate (PET) substrate. Such materials are used in packaging applications where a gas barrier is required to maintain the product quality. Cracking or debonding of the surface glass layer results in destruction of the gas barrier properties of the film. Accurate evaluation of the continuity or discontinuity at the crack edge within the layered material is crucial for the correct characterization of both adhesive failure and crack propagation. Here simultaneous atomic force microscopy and UFM images are compared for a range of tensile strains to identify debonded regions of the glass film from the PET. Debonding occurred for strains greater than 6%. In some films, this debonding increased with applied strain. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1450058]

Nanocomposite films consisting of a brittle glass deposited on a polymer substrate are used as packaging materials due to their transparency, microwave compatibility, and light weight.[1,2] During manufacture, storage, and use, such materials are subjected to deformation that can result in both cracking and debonding of the glass layer. Since the function of this layer is to provide a barrier to both gas and water vapor, cracking or debonding of the glass layer will provide pathways for gas permeation resulting in a poor quality packaging material.[3] To first order, gas permeation through cracked films varies linearly with crack density but only logarithmically with mean crack width (including debonded length).[4] Moreover, debonding due to inadequate adhesion between the glass layer and the substrate can lead to a stress concentration in the glass layer that produces further cracking. Characterization of these failure mechanisms is important to achieve improved durability in nanocomposite films.

Surface cracking in such materials has been extensively studied using a variety of techniques such as scanning electron microscopy (SEM),[5–7] optical microscopy,[8] and atomic force microscopy (AFM).[3] Characterization of glass debonding, however, has previously been experimentally inaccessible over the strain range of interest due to its interfacial nature and the nanoscale thicknesses involved. Optical observations do not offer sufficient resolution, AFM measurements characterize topographic changes only, and SEM generally requires a conducting coating on the film to eliminate charging of the exposed polymer substrate in the region of the cracks. Certain AFM-related techniques however, do allow characterization of this debonding region, for example ultrasonic force microscopy (UFM) or phase contrast AFM. UFM allows buried elastic features to be detected because it is sensitive to local changes in mechanical compliance.[9] In this letter we report on the application of UFM for the characterization of debonding behavior associated with *in situ* tensile straining up to 20% of three proprietary nanocomposite materials consisting of glass layers on poly(ethylene terephthalate) (PET).

Films were mounted and strained *ex situ* at a rate of $0.01\ \mathrm{min}^{-1}$ using a dc, 6000 rpm, Mavilor motor. Displacement was monitored throughout the straining process using a Testatronic TTD20 linear variable differential transformer output reader calibrated using a micrometer gauge. Samples were imaged at a variety of strains by AFM and UFM using a specially designed frame holder that allowed the films to be removed from the *ex situ* straining while remaining under tension. For UFM, a contact AFM measurement is performed while the sample is simultaneously vibrated ultrasonically using a piezoelectric transducer (in this case at approximately 2.4 MHz with amplitude modulation between 0 and <10 Å at 2.71 kHz). The subnanometer surface oscillation results in a high frequency tip–sample indentation, which is additionally modulated at 2.71 kHz. The cantilever responds at the modulation frequency only, with a deflection that depends on the nonlinear tip–sample interaction and hence on the local mechanical properties of the sample. The resulting UFM signal is detected via a lock-in amplifier.[10]

All of the films studied exhibited cracking upon the application of a tensile strain.[11] It was therefore important to initially establish the effect of purely topographic features (cracks) on UFM contrast before investigating debonding. Both AFM and UFM were calibrated using a silicon standard consisting of 1.5 μm wide rectangular plateaus on a 3 μm pitch with a height of 22 nm (Silicon-MDT, TGZ-01). The UFM signal varied only at the plateau edges due to a change in the local tip–sample contact area, but the plateau separation measured by AFM and by UFM was the same within

[a]Author to whom correspondence should be addressed; electronic mail: alison.mcguigan@utoronto.ca

