Study of the Ductile Fracture of PA11/EVOH Multilayer Films with the Essential Work of Fracture Method for Hydrogen Storage Applications

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Abstract

The new requirements for gaseous hydrogen storage have leaded the way for research into materials with high mechanical and barrier performance. The multilayer coextrusion process enables the combination of properties from multiple polymers by freely increasing the number of layers through the successive folding technique known as the " layer-multiplying " or " baker's " technique (1). The polymer composite films created in this way can contain layers on the scale of hundreds of nanometers, with interfacial properties that strongly influence the material's microstructure and mechanical response. These films are prone to transitions in fracture behaviour from ductile to brittle, which can potentially lead to structural failure (2). These transitions can be influenced by many parameters, such as polymer's sensitivity to environmental conditions (humidity, temperature), long-term mechanical loads (creep, fatigue) and short-term loads (dynamic stress, impacts). Multi-physical coupling can also initiate such transitions in fracture behaviour. In this application, the interaction between gaseous hydrogen and high pressure can lead to the formation of small voids, known as cavitation (3). These diffuse cavities within the material volume can significantly increase the likelihood of brittle fracture in the polymer. Thus, the fracture properties of the multilayers polymer films in ambient and embrittling conditions is to be determined, in relation with the application context. Multilayer films of PA11-EVOH models have been produced using the coextrusion technique, with the number of layers varying from 3 to 4097 layers. Preliminary physicochemical characterizations have been carried out on these produced films. Initial results allow for an assessment of the ductile fracture properties of these PA11/EVOH multilayer films, with a focus on the influence of film structuring at the layer-fold scale on their crack resistance. To this end, quasi-static fracture tests in mode I were conducted at ambient conditions. These tests rely on a formalism specific to the Elastic-Plastic Fracture Mechanics for testing elastoviscoplastic materials under plane stress conditions. This methodology, developed by Cotterel and Reddel in 1977, is known as the Essential Work of Fracture (4). Based on a separation of the energies involved in macroscopic fracture, this methodology

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enhances the description of ductile fracture mechanisms. It allows, in particular, the comparison of fracture resistance across different multilayer films (5). Post-mortem microscopic observations (optical and electron) of the fracture surfaces allowed for a discussion of the link between the local mechanisms involved at the micro and nanoscale, and the macroscopic fracture behaviour of these complex composite materials. This multi-scale material is subject to complex fracture mechanisms, involving a principle fracture in the plane perpendicular to the tensile direction, as well as multiple fractures in the tensile plane, aligned with the orientation of the polymer layers. Initial observations suggest weak adhesion between the PA11 and EVOH layers, leading to significant delamination between the layers. Moreover, diffuse damage due to cavitation within the EVOH layer were observed. The intensity of these mechanisms and their distribution are strongly related to the number of layers in the films, as well as to the stresses experienced during tensile testing. These mechanisms could explain the general ductile macroscopic behaviour of the films, as well as the differences in macroscopic fracture resistance observed, which are related to the number of layers. In parallel, the EVOH-PA11 films were subjected to rapid decompression cycles under gaseous hydrogen in severe conditions to develop cavities. Their fracture behaviour will be compared to that of the intact films under the same ambient conditions.

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