

Impact Deformation and Fracture Morphology of a Multi-layer HDPE Material

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ABSTRACT

This paper investigates the impact deformation and fracture of a co-extruded multi-layer polymeric material. The material consists of a layer of ethylene vinyl alcohol, EvOH, two layers of linear low density polyethylene (LLDPE) based adhesive and three layers of high density polyethylene (HDPE). The multi-layer material was driven dart impact tested with velocities of 0.4 m/sec and 4.0 m/sec at 23°C and -40°C temperature conditions. The fracture morphology of the material was studied using scanning electron microscopic (SEM). An increase in energy absorption up to the maximum load was observed at -40°C; whereas, the total energy absorption decreased when compared to those tested at ambient temperature conditions. SEM observation revealed that the EvOH layer was the major contributor to the reduced ductility of the multi-layer system.

INTRODUCTION

Multi-layer polymeric materials are preferred in many industrial applications because of their superior overall performance than the monolayer polymers. Typically, one polymer may exhibit high ductility and the other may possess better impermeable properties to prevent solvent diffusion. This combination therefore yields a high mechanical performance and better chemical resistance polymer system [1]. Some of the key applications of multi-layer materials can be found in pipeline systems for gas and fuel transportation, and automotive fuel tanks.

The present work investigates the deformation and fracture behavior of a co-extruded multi-layer polymeric fuel tank material. In this multi-layer material system, the high density polyethylene (HDPE), a semi-crystalline thermoplastic polymer with excellent mechanical performance, is interleaved with ethylene-vinyl alcohol (EvOH), a polymer with impermeable properties necessary to prevent solvent diffusion. These two intrinsically incompatible polymers, from an adhesion perspective, are bonded with a linear low density

polyethylene (LDPE) based adhesive to yield better mechanical and chemical properties than a monolayer fuel tank material [1].

Like most common semi-crystalline thermoplastics, the HDPE exhibits exceptionally high ductility at an ambient temperature and slow loading conditions; however, its mechanical properties vary significantly with loading rates and temperatures [2-7]. This nature of the semi-crystalline polymers can be explained by Eyring's equation [6] which describes the yield stress dependence on the strain rate and temperature.

Considering the vast application and importance of HDPE, significant scientific research efforts have been devoted to study the mechanical, thermal and fracture characteristics of the HDPE at quasi-static and dynamic loading conditions [2-5, 8-13].

Dasari and Misra [2] studied the strain rate sensitivity of HDPE and PP (polypropylene), and investigated its concurrent micro-structural failure evolution. The HDPE was more strain rate sensitive than polypropylene (PP). Scanning electron microscopic (SEM) observations of the fractured HDPE surface showed extensive fibrillation at low strain rates, and a combination of fibrillated fracture and craze-tearing at higher strain rates. Martins et al. [4] studied critical energy release rates as a function of impact velocity, and analyzed the micro and macro failure mechanism of linear medium density polyethylene (LMDPE). The results of their investigation were compared with Clutton and Channell's [14] work for linear low density polyethylene (LLDPE) and linear high density polyethylene (LHDPE) material. Investigation by Martins et al. showed that the intrinsic toughness of polyethylene was not significantly different amongst the three grades of polyethylene.

Abu-Isa et al [8] studied the behavior of the HDPE multi-layer fuel tank material and its individual constituents at ambient temperature and -40°C. The mechanical, thermal and rheological properties showed that the HDPE and the regrind HDPE remained ductile at -40°C.

where as the EvOH demonstrated a brittle behavior. Compared to its constituents, the multi-layer fuel tank material was relatively brittle and showed lower impact strength at -40°C, a behavior similar to the EvOH. Mallick and Zhou [9] investigated the tensile and creep properties of the multi-layer HDPE material at 23°C, 50°C and 75°C. A strong temperature dependent tensile behavior was observed for the multi-layer system.

Quiu and Shanahan [1] studied the adhesion between grafted PE and EvOH in a co-extruded and blow molded multi-layer HDPE polymer system. Peel tests at various peel angles, temperatures (0°C to 70°C) and peel speeds were conducted for adhesion studies. Merah et al [5] investigated the temperature effect (-10°C to 70°C) on the tensile properties of HDPE pipe materials. Krishnaswamy [10] investigated a wide range of HDPE pipes subjected to extensive hydrostatic pressure at multiple hoop stress levels and temperature conditions (23°C, 60°C and 80°C). The HDPE pipes failed in ductile mode for all tested conditions. The ductile failure was mainly driven by the material density or crystallinity and was independent of the molecular weight, molecular weight distribution, and the branching content and distribution. An increase in polymer performance at higher temperatures, observed in HDPE pipes was mainly ascribed to the relaxation of residual stresses in the polymers.

The literature study reveals the multifaceted behavior of the multi-layer semi-crystalline polymer (HDPE/EVOH/LDPE), depending on morphology, thermal, and mechanical loading conditions. This behavior reinforces the need for further characterization of HDPE fuel tank material subjected to impact loads.

This paper investigates the deformation and fracture of a co-extruded multi-layer HDPE material under lateral impact loading. The multi-layer material was driven dart impact tested with dart velocities of 0.4 m/sec and 4.0 m/sec at ambient and -40°C temperature conditions. The fracture morphology of the material was studied using scanning electron microscopy (SEM).

EXPERIMENTAL

MATERIAL - The material investigated in this work is a co-extruded high density polyethylene (HDPE) multi-layer fuel tank material [15]. Figure 1 presents a schematic representation of the multi-layer structure. Starting from the inner wall, the layered materials and their volume fractions are: virgin HDPE (40%), LDPE based adhesive (2.5%), EvOH (3%), LDPE based adhesive (2.5%), regrind HDPE (40%) and virgin HDPE loaded with carbon black (12%). Since the volume fraction of the HDPE layer is over 90% in this multi-layer system, it is referred as the multi-layer HDPE in the text thereafter.

The multi-layer HDPE fuel tanks are manufactured by extrusion process.

Inergy Automotive Systems, a fuel tank supplier, provided the multi-layer HDPE fuel tanks for the investigation.

DRIVEN DART IMPACT EXPERIMENT - Instron VHS 80/20 servo-hydraulic testing machine and an environmental chamber shown in Figure 2 were used for the driven dart impact experiments. The testing machine operates with a close-loop control at velocities lower than 0.1 m/sec, and with an open-loop control for velocities above 0.1 m/sec. The Instron environmental chamber has a temperature control system with accuracy of $\pm 2^\circ\text{C}$. A thermocouple attached to the specimen was used to monitor the specimen temperature. The load, displacement, and time data were recorded by the data acquisition system of the testing machine. The velocity was calculated by the data acquisition software Fast Track 8800® (High Rate Program).

The specimens were square plates (4X4 in) machined from multi-layer HDPE fuel tanks. The specimen was clamped between a pair of 76mm diameter annular anvils (driven dart impact fixture) and impacted by a semi-spherical end metal dart of 20mm in diameter. A schematic representation of the specimen clamped in a driven dart fixture is shown in Figure 3. All the specimens were impacted from the outer side; that is, the virgin HDPE with carbon black side. The tests were conducted for 4 m/sec and 0.4 m/sec dart velocities at ambient temperature and -40°C conditions. Liquid nitrogen was used for cooling for the tests conducted at sub-ambient temperature. The fixture along with the clamped specimen was cooled down in the environmental chamber. The specimen reached -40°C in about 30 min from the room temperature. In order to achieve an equilibrium temperature state the specimens were soaked for another 30 minutes before the impact tests.

POST-MORTEM CHARACTERIZATION - The fractured surface of the impact tested multi-layer HDPE fuel tank material was analyzed using a cold field emission SEM (Hitachi S-4800).

The samples used for SEM observation were cut from impact tested specimens along the fractured edge, as indicated in Figure 4. The samples were coated with gold palladium for about 10 sec and the surface morphology was inspected with an acceleration voltage of 3 kV in the SEM.

RESULTS AND DISCUSSIONS

DRIVEN DART IMPACT RESULTS - Load, displacement and time data was recorded using Fast Track 8800 for the driven dart impact experiments. Figure 5 shows the typical velocity and load history plots for a nominal velocity of 4 m/sec. Figure 6 shows the typical load-displacement curve and an energy plot for 4 m/sec dart velocity at -40°C. The energy absorption was measured as the area under the load-displacement curves; it was obtained by integrating the load-displacement curves. Figure 7 and Figure 8 show the comparison between

ambient temperature and -40°C load-displacement curves for 0.4m/sec and 4m/sec dart velocities, respectively. The load-displacement curves show similar behavior: an initial linear deformation region, a region of nonlinear deformation and the maximum load for the two temperature conditions. However, beyond this maximum load, the load deflection behavior is different for the -40°C condition. After the maximum load, the load drops much faster in the -40°C curves compared to the room temperature curves. This is then followed by a rapid load decrease in both cases.

Table 1 and Table 2 provide the maximum loads and the displacements at maximum load for room temperature and -40°C conditions at 0.4m/sec and 4m/sec dart velocities, respectively. The deflection for maximum load at -40°C was lower than the ambient temperature deflection. Both the peak load and the slope of the load displacement curves increased at -40°C. The effect of temperature on the maximum load and the deformation of the multi-layer HDPE material were similar to the behavior observed by Evans [16] for HDPE bumpers.

Stiffness and flexural modulus - The slope of a load-displacement curve provides the stiffness for a material. The slopes of the load-displacement curves were determined in the range of 0mm -10mm displacement.

The flexural modulus of the material can be obtained from the slope of the load-displacement curves. Dear et al [17] estimated the in-plane modulus of sheet molding compound (SMC) and glass mat thermoplastic (GMT) materials from a drop weight impact test using a simply supported plate bending formula. The driven dart test configuration in the current work is close to the case of a flat circular plate with fixed edge under uniform load over a very small central circular area. In this case, the relationship between the center deflection w and the total load P is given approximately as [18]

$$E = \frac{3a^2(1-\nu^2)}{4\pi h^3} \left(\frac{\Delta P}{\Delta w} \right) \quad (1)$$

where a is the radius of the circular portion of the specimen (38mm), ν is the Poisson's ratio, E is the flexural modulus, h is the thickness of the plate and $\left(\frac{\Delta P}{\Delta w} \right)$ is the slope of the load displacement curve.

Equation 1 was used to estimate the flexural modulus in the present study. Table 3 provides the flexural modulus obtained from driven dart impact at the two temperatures and the dart impact velocity conditions. An increase of about 135% to 145% was observed in the flexural modulus of the HDPE fuel tank material for the two dart impact velocities. A decrease in the modulus and the stiffness with an increase in temperature of the fuel tank material follows the behavior of a neat HDPE, which is almost 90% of the fuel tank material [5].

Energy absorption - The area under the load-displacement curve gives the measure of the energy absorption. In this study the energy absorption up to the maximum load and the total energy absorption are considered for comparison, which is in line with the work of other investigators [19, 20]. Table 4 provides the average energy at the maximum load and the average total energy for different temperature and velocity conditions. It shows that the energy absorption up to the maximum load increased by about 10% while the total energy absorption decreased by about 5% for materials tested at -40°C. The energy absorption up to the maximum load is attributed to the higher modulus and strength of the fuel tank material at lower temperatures. However, this did not offset the effect of reduced ductility and hence there was a net reduction of 5% in the total energy absorption.

DEFORMATION AND FRACTURE - In general, the deformation characteristics of thermoplastics are governed by stretching and separation of tangled-up thread-like macromolecules. This stretching process is time driven and, therefore, the occurrence of deformation loading to fracture is governed by the rate of loading. Additionally, elevated temperatures facilitate such rearrangements. Molecular chains can adapt more easily to the applied stresses at slow loading rates and elevated temperatures than at higher loading rates and low temperature [21].

Deformation behavior - In general, the specimen deforms globally before localized penetration. This behavior was consistent for both the specimens tested at ambient temperature and -40°C. Figure 9 shows a typical deformation process in a driven dart impact test.

Post-mortem analysis -

Visually inspection - To understand the failure mechanism, driven dart impact tested specimens were examined first by visual inspection. Figure 10 and Figure 11 show the comparison of the fuel tank material fractured at 0.4m/sec and 4m/sec dart velocities, respectively, for the two temperature conditions. The specimen tested at ambient temperature with 0.4m/sec dart velocity (Figure 10a) exhibited a deep drawn pocket and the final fracture occurred along the circumference of the pocket wall. At -40°C, the deep drawn pocket behavior almost disappeared (Figure 10b). The specimen tested at ambient temperature with 4m/sec dart velocity (Figure 11a) also exhibited a drawn pocket but its depth reduced as compared with that tested at 0.4m/sec. The fracture morphology of the specimen tested at -40°C with 4m/sec dart velocity (Figure 11b) was similar to that tested at -40°C with 0.4m/sec dart velocity and displayed even less stretching. There was no deep drawn pocket and the fracture moved from the circumference toward the center of the impacting site. This is an indication that the -40°C temperature condition had a more significant effect on ductility than impact velocities. The sudden drop in load after the peak load in the load-displacement curves supports this claim.