Available online at www.sciencedirect.com
ScienceDirect
Procedia Engineering 10 (2011) 704–709
Engineering Procedia

ICM11

Impact response of polyethylene nanocomposites

I. Mohagheghian[a], G. J. McShane[a], W. J. Stronge[a]*

[a]Department of Engineering, University of Cambridge, Cambridge, CB2 1PZ, UK

Abstract

The quasi-static and dynamic behaviour of Linear Low Density Polyethylene (LLDPE) and two LLDPE nanocomposites were studied. Nanocomposites consisting of LLDPE filled with 1% carbon black and 0.5% nanoclay fillers, by weight, were considered. Under quasi-static tensile loading, an improvement in the energy absorbing capability was achieved by adding 1% carbon black fillers. However, during quasi-static puncture and dynamic impact loading, the advantage provided by the fillers was lost. Thermal softening due to adiabatic heating under high strain rate deformation and differences in the state of stress are considered as reasons for this reduction.

Selection and peer-review under responsibility of ICM11

Keywords: Nanocomposites; impact; energy absorption

1. Introduction

Recent studies have shown that the combination of a polymer with nanoscale fillers can increase the stiffness and strength compared to unfilled material, as well as leading to improvements in toughness, and impact energy absorption [1]. For example, adding 4.2-wt% nanoclay increases the tensile strength of nylon 6 from 69 MPa to 107 MPa, and simultaneously doubles the tensile modulus [2]. Song et al. [3] noted that the tensile strength of PU can be improved by 120% by adding nanoclay fillers. Introducing only 1 wt% multi-walled carbon nanotubes (MWNTs), the tensile modulus and the tensile strength are greatly improved by 115% and 120%, respectively compared to unfilled PA6 [4]. The small size of these nanofillers (typically 0.1 to 100 nm) is comparable to the size of polymer molecular chains, and results in a considerably higher interaction surface area between the filler and the matrix than conventional micro-sized fillers. Improvements in the mechanical properties of the polymers are achieved with little increase in the material weight.

The goal of the present study is to investigate the potential for polymer nanocomposites in light armour applications, where impact energy absorption at low weight is important. To date, there have been few reported studies on the application of polymer nanocomposites in ballistic protection. Hsieh et al. [5] investigate the ballistic impact strength of polycarbonate-layered silicate nanocomposites. Abdelkader et al. [6] consider the ballistic

* Corresponding author. Tel.: +44-1223-332652; fax: +44-1223-332662.
E-mail address: wjs@eng.cam.ac.uk.

Selection and peer-review under responsibility of ICM11 doi:10.1016/j.proeng.2011.04.117

performance of polycarbonate filled with multi-walled carbon nanotubes (MWNTs) in order to assess their potential in lightweight personnel armour applications. Their results show that the addition of carbon nanotubes improves the energy absorbing capability of the polycarbonate in a ballistic impact. Laminated hybrid composites, consisting of conventional composite plies and intercalated layers of nanocomposites, have also been explored for body armor applications [7].

In this paper, Linear Low Density Polyethylene (LLDPE) and LLDPE nanocomposites filled with nano-scale carbon black (CB) and nanoclay (NC) have been studied both quasi-statically and under impact loading by 12.5mm diameter hemispherically tipped projectiles. Little data has been published to date on the dynamic response of LLDPE-based nanocomposites. LLDPE is a low cost polymer, with comparatively low strength and high ductility compared to more traditional impact-resistant polymers such as polycarbonate. We aim to investigate in this study the degree to which nanofillers can enhance its performance under impact loading.

2. Material characterisation

2.1. Material preparation

Polyethylene powder was first blended with the required weight fraction of CB or NC fillers in a twin screw extruder. The filler powders were pre-treated in order to achieve better dispersion within the polymer matrix. Square plates of side length 130 mm and thickness 2.6 mm were then compression moulded from the blended mixture. Moulding pressure and temperature was controlled to ensure good dimensional accuracy of the final plates.

2.2. Quasi-static uniaxial tensile tests

Dogbone shaped specimens were machined from plates of the unfilled LLDPE and its two nanocomposites plates based on the ASTM D638-03 type V specification, and tested to failure in an Instron screw driven tensile testing machine. Three repeat measurements were taken for each specimen type, to assess repeatability. In order to select a suitable filler weight fraction for the study, tensile tests were performed on nanocomposites with 0.5%, 1% and 2% of fillers by weight. Considering the total energy absorption up to the point of tensile failure, 1% for CB fillers and 0.5% for NC were selected as the optimum compositions. In both cases, the strain at break, and consequently the energy absorbed, reduced if the filler content was increased further.

Figure 1 shows typical uniaxial tensile results for LLDPE and its nanocomposites. For all three material types, the specimen displays an initial elastic response, followed by a load drop after yield, accompanying neck formation. There follows a plateau in nominal stress corresponding to propagation of the neck. Subsequently the nominal stress increases due to strain hardening, followed by final rupture. Very large strains are observed at rupture, with the gauge section elongating by more than 20 times its original dimensions. Comparing the three materials included in Figure 1, it can be seen that adding 1% CB has a positive effect on the total energy absorption, by postponing the final rupture. In contrast, the NC decreases the strain to failure of the LLDPE. For both filler types, there is a small increase in the initial yield strength, but no observable difference in the subsequent strain hardening. The average results of repeated tests on LLDPE and its nanocomposites are summarised in Table 1.

Table 1. Summary of quasi-static tensile test results for LLDPE and its nanocomposites. Averages of multiple tests are shown.

Material	Yield strength (MPa)	Nominal strain at break
LLDPE	16.23 ± 0.25	22.50 ± 0.72
LLDPE+1% CB	17.28 ± 0.32	25.22 ± 0.39
LLDPE+0.5% NC	16.83 ± 0.27	20.67 ± 0.61

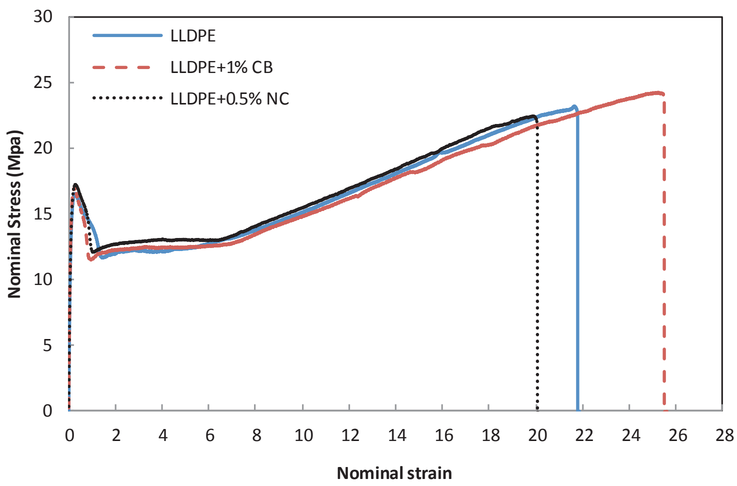
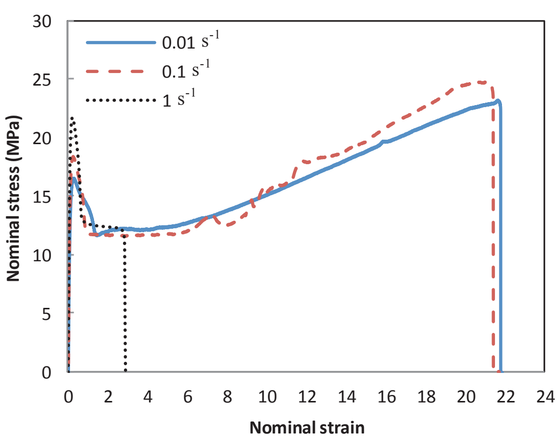


Figure 1. Typical nominal stress-nominal strain curves for LLDPE and its nanocomposites.

2.3. The effect of strain rate on the uniaxial tensile response

The uniaxial tensile tests were repeated at higher strain rates (0.1 s^{-1} and 1 s^{-1}) using the same specimen geometry and Instron screw driven tensile testing machine. The influence of strain rate on the tensile response is shown in Figure 2a for pure LLDPE. Increasing the strain rate to 0.1 s^{-1} increases the initial yield strength and strain hardening, but reduces slightly the strain at failure. However, at a strain rate of 1 s^{-1} , the material fails soon after neck formation, and the neck no longer propagates along the whole gauge length. As a result the ability of the material to absorb energy significantly decreases. A similar behaviour was observed for the nanocomposite specimens. Figure 2b shows the nominal strain at break for LLDPE and its nanocomposites at different strain rates. LLDPE with 1% CB shows a greater reduction in ductility due to the increase in strain rate than the unfilled polymer. In contrast, the LLDPE with 0.5% NC shows a smaller reduction. Consequently, the influence of the nanofillers almost entirely vanishes at 1 s^{-1}, the three materials achieving comparable strain at break.

(a)



(b)

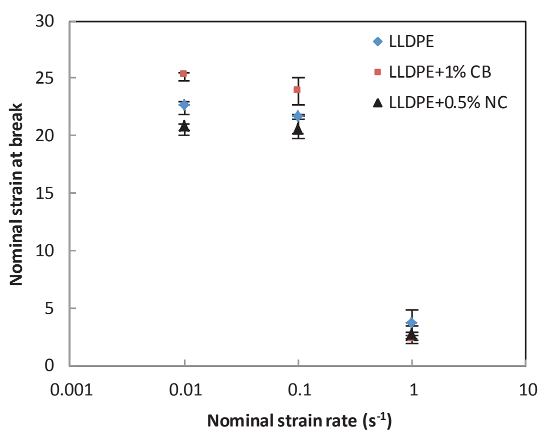


Figure 2.. (a) The influence of strain rate on the nominal stress-strain curve of unfilled LLDPE; (b) The change in nominal strain at break with strain rate for unfilled LLDPE and its nanocomposites.

2.4. Quasi static puncture

In order to assess the influence of the loading conditions on the performance of the materials, quasi-static puncture tests were performed, using identical plate and indenter tip geometry to the impact experiments (described subsequently). Circular plates of diameter 100 mm and clamped around the periphery were loaded centrally using a hemispherically-tipped indenter of diameter 12.5 mm. The rate of indentation was maintained at 1mm min^{-1}. The indenter force and displacement were measured, and are shown in Figure 3 for the unfilled LLDPE and its nanocomposites. Repeat measurements (not shown) indicate good repeatability of the results. In each case, the load increases to a maximum, at which point a tensile neck develops around the indenter. The neck then propagates, accompanied by a load drop, before final rupture. The results show that subject to quasi-static indentation loading, both the CB and NC filled polymers absorb less energy at rupture than the unfilled polymer.

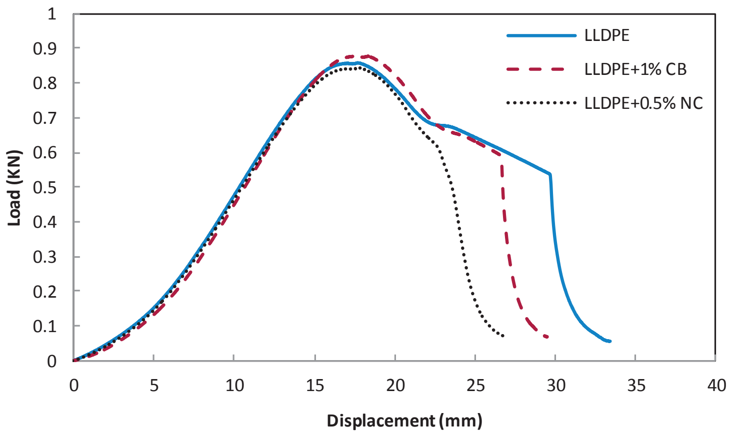


Figure 3. Force-displacement curves for the quasistatic puncture of LLDPE and its nanocomposites at a rate of 1 mm min^{-1}.

3. Impact experiments

A series of projectile impact experiments were conducted on 2.6 mm thick polymer plates. Circular plates of diameter 100 mm clamped around the periphery were considered. The steel projectiles (mass 20.2 g) were circular cylinders of diameter 12.5 mm with the tip machined into a hemisphere of the same diameter. The projectile was fired using a gas gun. The position and velocity of the projectile was measured before, during and after impact using high speed photography.

Figure 4a illustrates the deformation of an unfilled LLDPE plate after impact, for an initial projectile velocity of 45.5 ms^{-1}. Note that at this impact speed, the projectile has penetrated the plate, yet has been arrested due to the resistance posed by elastic cavity expansion. Deformation has localised around the circumference of the projectile, with neck formation followed by void nucleation, coalescence and finally rupture. A cap of material is separated at the tip of the projectile. For comparison, the failure observed during a quasi-static puncture experiment (Section 2.4) is shown in Figure 4b for the same material. Quasi-statically, the neck which develops around the circumference of the indenter does not remain localised. Instead, the neck propagates, forming a cap which thins uniformly, achieving a large strain before rupture (note the uniformly thin, translucent membrane in figure 4b). Under impact loading, the localisation of the deformation means that the tensile strains in the neck are comparatively small. A nearly identical failure mode is observed for the two types of nanocomposite under impact loading.

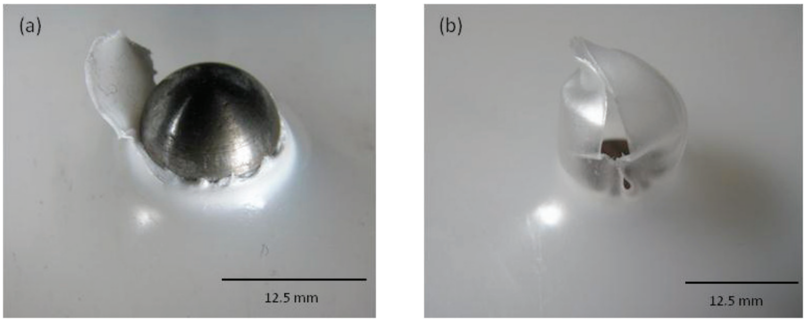


Figure 4. (a) Unfilled LLDPE plate after impact by a projectile with initial velocity 45.5 ms^{-1}; (b) Unfilled LLDPE plate perforated by quasi-static indentation at a rate of 1mm min^{-1}.

The impact performance of pure LLDPE and its nanocomposites is summarised in Figure 5. Figure 5a shows the variation in the residual velocity with impact velocity, a positive residual velocity indicating motion in the impact direction. At low velocities, the plate is not perforated and the projectile rebounds. There are a range of impact velocities over which the projectile has zero residual velocity. In these cases the plate is penetrated, and the projectile arrested (as in Figure 4a). At higher velocities, the plate is fully perforated and the projectile passes through, recording a positive residual velocity. Figure 5b shows the energy absorbed during the impact, measured from the change in the projectile's kinetic energy. The unfilled and nanocomposite plates show very similar performance over the full range of impact velocities. No benefit in ballistic limit is observed for either NC or CB fillers.

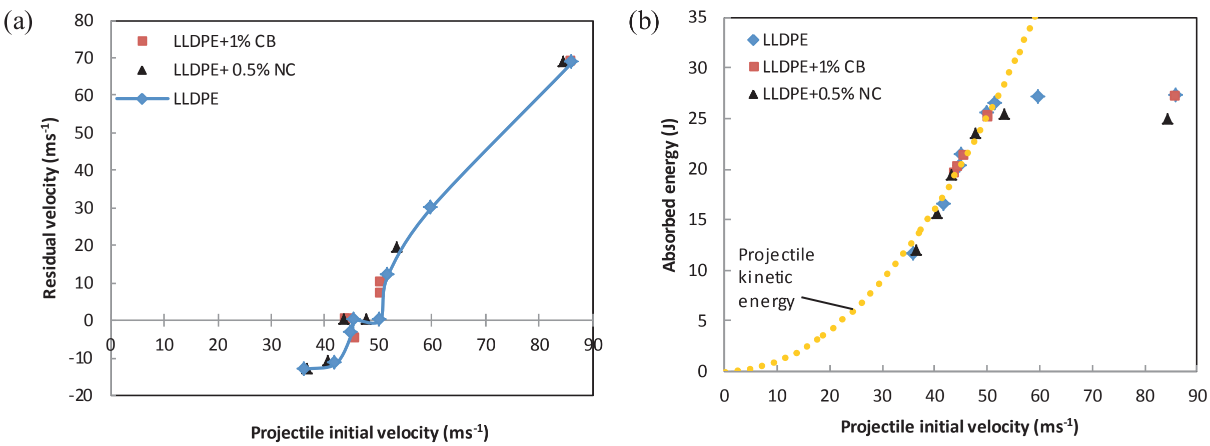


Figure 5. (a) Residual velocity versus initial (impact) velocity of the projectile and (b) energy absorbed during the impact, for pure LLDPE and its nanocomposites.

4. Discussion and conclusions

The energy absorbing capability of pure LLDPE and LLDPE filled with 1% CB and 0.5% NC were investigated under both quasi-static and dynamic conditions. An improvement was observed in the energy absorbing capability of LLDPE under quasi-static tensile loading when nanofillers were added. However, the improvement was lost under both dynamic tensile loading and quasi-static puncture testing. In the latter case, the performance was measurably reduced. Under impact loading, filled and unfilled LLDPE performed similarly. Identifying the causes of the observed behaviour will require further investigation. However, possible reasons are as follows.

The effect of adiabatic heating on the failure of polymers has been reported in a number of studies, for example [8] and [9]. The authors argue that local heating can destabilise the necking process and cause rupture soon after neck formation. The temperature increase due to adiabatic heating for highly crystalline polyethylene can be of the order 30-80 °C [9]. In the current study, under both impact loading and uniaxial tension at higher strain rates, a lack of neck propagation was observed. It is possible that this localisation of deformation is due to thermal softening as a result of adiabatic heating. The lack of neck propagation may also explain the similar performance of unfilled and nanofilled LLDPE. The quasi-static tensile results indicate that the nanofillers provide a benefit only in the later stages of tensile deformation, by delaying rupture during the drawing out of the neck. In the absence of neck drawing, their influence may be reduced. In addition to localising deformation, it is also possible that strain rate dependent heating effects may degrade toughening mechanisms in the filled polymers. Considering Figure 2b, it appears that a strain rate of 0.1 s^{-1} is sufficiently low to avoid failure immediately at the onset of necking. However, the effectiveness of the carbon black fillers in delaying rupture of the polymer is nonetheless reduced. It is possible that toughening mechanisms such as crazing and fibrillation which are promoted by the presence of nanofillers become less effective as the temperature is increased due to the increase in strain rate.

The state of stress during impact deformation of the filled polymers may also be a significant factor in their relative underperformance. Quasi-statically, it was observed that indentation loading resulted in premature failure of the filled polymers compared to the unfilled LLDPE, in contrast to results under uniaxial tensile stress. It therefore appears that the development of damage in the filled polymers is sensitive to the state of stress. The influence of multiaxial stress states on damage development in these nanocomposites is currently poorly understood, particularly at high strain rates.

In conclusion, it appears that minimising heating effects during impact loading is important if the potential benefits provided by nanofillers are to be achieved. One strategy to achieve this would be to increase the strain hardening of the polymer matrix. This would help to stabilise neck development, encouraging neck propagation, minimizing adiabatic heating effects, and activating the large tensile strain regime in which the fillers appear to provide a benefit in enhanced ductility. However, the damage mechanisms occurring at the filler scale at these strain rates and under impact loading conditions also need to be better understood.

Acknowledgment

The authors are grateful to Prof M. Song and Dr D. Cai of the Chemical Engineering Department at Loughborough University, UK for assistance with the fabrication of polymer specimens. The EPSRC Engineering Instrument Pool is acknowledged for the loan of the high speed camera.

References

[1] Sun L, Gibson RF, Gordaninejad F, Suhr J. Energy absorption capability of nanocomposites: A review. *Compos Sci Technol*; 2009, **69(14)**: 2392-2409.

[2] Okada A, Usuki A. The chemistry of polymer–clay hybrids. *Mater Sci Eng*; 1995, **3(2)**:109–115.

[3] Song M, Hourston DJ, Yao KJ, Tay JKH, Ansarifar MA. High performance nanocomposites of polyurethane elastomer and organically modified layered silicate. *J Appl Polym Sci;* 2003, **90(12)**: 3239-3243.

[4] Liu T, Phang IY, Shen L, Chow SY, Zhang WD. Morphology and Mechanical Properties of Multiwalled Carbon Nanotubes Reinforced Nylon-6 Composites. *Macromolecules*; 2004, **37(19)**: 7214-7222

[5] Hsieh AJ, Song JW, Neboand J, Singh A. Ballistic Impact Measurements of Polycarbonate Layered-Silicate Nanocomposites. NTEC 2001 *Conference Proceedings, Society of Plastic Engineers, CT*; 2001, 2185–2190.

[6] Abdelkader M, Withers JC, Loutfy RO, Moravsky A, Sennett M. The Investigation of Carbon Nanotube for Lightweight Armor Material. *In 23rd Army Science Conference;* 2002, Orlando, FL.

[7] Grujicic M, Pandurangan B, Angstadt DC, Koudela KL, Cheeseman BA. Ballistic-Performance Optimization of a Hybrid Carbon-Nanotube/E-Glass Reinforced Poly-Vinyl-Ester-Epoxy-Matrix Composite Armor. *J Mater Sci*; 2007, **42(14)**: 5347–5359.

[8] Hillmansen S, Haward RN. Adiabatic failure in polyethylene. *Polymer*; 2001, **42(22):** 9301-9312.

[9] Fager LO, Bassani JL. Neck propagation in polymers with adiabatic heat generation. *Mech Mater;* 1990, **9(3)**: 183-194.