

**RESEARCH ON RUPTURE AND DRAWING BEHAVIOR OF SEPS/HDPE
COMPOSITES**

by

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University of Pittsburgh, 2018

We examine the stretching behavior of rubber–plastic composites composed of a layer of styrene-ethylene/propylene–styrene (SEPS) rubber, bonded to a layer of linear high density polyethylene (HDPE) plastic. In tension, the SEPS layer showed homogeneous deformation, whereas the pure HDPE layer usually break in the tensile process. We tested bilayer and trilayer structure composites separately. For bilayer, the experiment result proved that by bonding the SEPS layer, HDPE layer can be stretched to a higher degree without failure, and composite laminates showed behavior intermediate between the plastic and the rubber.

For trilayer, dog-bone shaped samples of rubber, plastic, and SEPS/HDPE trilayer with rubber: plastic thickness ratio in the range of 0.417-3.625 were subjected to uniaxial tension tests. The local stretch and the maximum local stretch was quantified by force-extension measurements and digital image correlation analysis of video recordings of these tests. The specimen result proved that the brittleness of plastic layer in trilayer structure was also reduced. Under the same strain rate, the degree of necking and drawing of trilayer reducing as the rubber: plastic ratio increased.

TABLE OF CONTENTS

ACKNOWLEDGEMENTS	XI
1.0 INTRODUCTION.....	1
2.0 EXPERIMENT METHOD	8
2.1 FABRICATION.....	8
2.2 TENSILE TESTS.....	9
2.3 ANALYSIS BY DIC	9
2.4 TRILAYER ACTUAL THICKNESS MEASUREMENT.....	10
3.0 BEHAVIOR OF HDPE/SEPS BILAYERS	13
3.1 BEHAVIOR OF PURE HDPE.....	13
3.2 BEHAVIOR OF PURE SEPS.....	17
3.3 BEHAVIOR OF SEPS/HDPE BILAYERS.....	19
4.0 MECHANICAL BEHAVIOR OF HDPE/SEPS/HDPE TRILAYER.....	30
4.1 MECHANICAL PROPERTIES.....	33
4.2 DIC FOR TRILAYERS	41
4.2.1 Qualitative behavior of tensile deformation.....	42
5.0 SUMMARY	46

BIBLIOGRAPHY..... 48

LIST OF TABLES

Table 1. Actual Thickness of Extruded Trilayer Sample.....	12
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LIST OF FIGURES

Figure 1 The nominal stress-strain response for rubber, LLDPE, LLDPE-Rubber bilayers of rubber:plastic thickness ratio 1.2 and 4.0 stretched at a rate of 120 mm per minute.....	4
Figure 2 (a) The extension-load curves (b) true stress-true strain curves of HDPE at different cross head speeds.....	5
Figure 3 (a) Cracked HDPE-Rubber composites (b) Drawing HDPE-Rubber composites (c)Delaminated HDPE-Rubber composites.....	6
Figure 4 Images of samples during tensile deformation of dogbone-shaped samples of HDPE plastic. (a) HDPE plastic at 40 mm/min (brittle behavior). (b) HDPE plastic at 40 mm/min (drawing behavior). (c) HDPE plastic at 900 mm/min (brittle behavior)	14
Figure 5 Strain-stress curves of compression molding HDPE under five different strain rates.	16
Figure 6 Images of rubber being stretched at 40 mm/min. Appearance at 900 mm/min is very similar and not shown.....	17
Figure 7 Stress-strain data for SEPS rubber at 40 mm/min and 900 mm/min.....	18
Figure 8 Appearance of bilayer of thickness (a) 78 μ m HDPE, 162 μ m rubber measured at 40 mm/min, (b) 62 μ m HDPE, 131 μ m rubber measured at 900 mm/min.....	20

Figure 9 Strain-stress curve of HDPE and its composites, dot lines mean the single layer at 40 mm/min. B. Force vs elongation for the same samples. C-D. Bilayer Force-Extension of experimental data versus calculated data based on equation 3.....	22
Figure 10 (A) Extension-Force curve with two strain rates (900mm/min), the rubber: plastic thickness ratio is approximately equal to 2, 2.5, 3 and 3.5. (B) corresponding Strain-stress curve.	24
Figure 11 The draw stress of different plastic:rubber ratio thickness at two speeds. The blue hollow dots represent the high speed and the green dots represent the low speed.	26
Figure 12 Snapshots at 900 mm/min showing differences in drawing. The rubber:plastic ratio of upper is roughly 1:1 and roughly 4:1 of the bottom one.	26
Figure 13 The mechanical work needed when bilayers come to 90 mm elongation in different rubber:plastic thickness ratio. The work calculation for HDPE work is based on its highest elongation before failure.	27
Figure 14 (a) Extruded HDPE at 40mm/min (b) Extruded HDPE at 900mm/min.....	31
Figure 15 (a-e) Extension-load curve of trilayer measured at 40 mm/min.....	34
Figure 16 The images of multiple necking happened.	35
Figure 17 The $\sigma_{nom,p}$ of different actual thickness ratio under the 100mm extension.	37
Figure 18 (a-e) Strain-stress curve of trilayer measured at 40 mm/min.	38
Figure 19 Draw stress of trilayer in different rubber:plastic thickness ratio	39
Figure 20 The mechanical work need when composites come to 90 mm elongation in different rubber:plastic thickness ratio.	40

Figure 21 (a-b). a is nominal ratio 0.25 sample, b is nominal ratio 4 sample.....	41
Figure 22 (a-d) The local stretches vs. time of different nominal ratio.	42
Figure 23 natural draw ratio of different nominal ratio.	44
Figure 24 (a) The short gauge length sample. (b) The drawing image of the extruded HDPE specimen. (c)The local stretch vs. frame change of extruded HDPE.	45

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1.0 INTRODUCTION

The response of a solid to external forces can vary greatly due to the magnitude of the forces and the characteristic of material. For some materials, if the stress is too high, the material may fracture. Other materials tend to deform permanently. However, if the stresses are low enough, the material can deform elastically. For instance, a ruler made of plastic may break easily, a metal ruler may bend permanently, whereas a rubber ruler will bend, but recover once the force is removed. Furthermore, if a rod or a bar with uniform cross section is pulled, the performance of the bar can vary greatly from different types of material. An elastomeric material stretches uniformly with a correspondingly uniform decrease in thickness. In contrast, a bar made from other kinds of polymer such as polyethylene or polycarbonate, tends to develop a neck in tension[1], Necking is a typical yielding behavior, often accompanied by plasticity. Basically, once the material is deformed beyond the yielding point, the deformation become permanent and cannot be recovered anymore. The necking process is as below: for example, if there is a long bar or rod under tension, first, the material shows a localized thinning somewhere (usually a defect location). This thinner part bears a higher local stress than the other parts of the material which then accentuates the neck. This necking raises the local stress further, causing further thinning in turn[2]. However, there are some exceptions like semicrystalline polymers for which, after the neck appear, there is no further thinning but instead the neck spreads by recruiting

surrounding material into the neck. Such neck propagation is sometimes called “cold drawing” a term originally introduced by Carothers and Hill[3]. Hutchinson and Neale have explored the elastic solid and an inelastic flow theory solid with both rate-dependent and rate-independent behaviors of neck propagation [4]. This drawing becomes a permanent deformation will not recover even though the stress is removed.

In practical applications, it is possible to realize the desired properties (mechanical, thermal, transport) by combining multiple materials into one. One simple way of combining two materials is by simply laminating together two different materials into a bilayer. Different combinations of layers are expected to have distinct properties. Generally, a bilayer is expected to have a combination mechanical properties of its original components. As the example we mentioned last paragraph, if is a ruler is made from a layer of plastic bonded to a layer of rubber, but also avoid the rupture due to the existence of rubber. For example, Xiang et al [5] studied the composites that made by bonding a copper film to a polymer substrate, according to their conclusion, the Cu film can sustain strains up to 10% without appreciate cracks by adhering a Cu film on a polymer substrate. Other researchers have similarly found that metal structure can be strengthened by bonding to plastics[6]. While there has been much research on polymer multilayers[7], almost all these papers are on small deformation. So, our research is on large deformation behavior in multilayers considering its potential value for toughening and improving the crack tolerance.

An example of the interesting effects possible from bonding rubbers to plastics is given by Li and Suo who explored the deformation of a yielding layer bonded to an elastomeric layer by theory and simulation[8]. For single layer in the stretching process, the yielding layer developed a single neck at first, and then failed at the neck location. In contrast, constitutive

equation for the elastomer was such that it showed uniform thinning and stretching. Bilayers of the yielding layer and the rubber layer showed three types of behavior: First, when the elastomer layer is thin, the multilayer developed a single neck. For thicker elastomer layers, multiple necks appeared simultaneously. Finally, when the elastomer thickness is large enough, the composites thinned homogeneously to large strain. These simulations suggest that by bonding elastomeric layer, the plastic (the yielding layer) can be stretched to a higher degree without failure. Those simulations were done assuming the layers were metal, and hence allow for the possibility of making metals stretchable by bonding them to rubbers.

Polymeric plastic however can also show stable drawing. Previous experiments from our lab have examined this[9]. Bilayers were made by bonding a rubber layer to a low-density polyethylene (LLDPE) plastic layer. Fig. 1 shows the nominal stress strain response of pure LLDPE plastic, SEPS rubber and laminate composites with two different rubber:plastic thickness ratios. The nominal strain is defined as the ratio of the crosshead displacement to the gauge length (20 mm). The curve for the SEPS rubber increases monotonically while the LLDPE plastic shows a sharp rise in stress at small strain, followed by a necking behavior. The LLDPE-Rubber composites behavior is similar to the LLDPE: Firstly, a homogeneous deformation appeared, followed by necking and then drawing. But the maximum stretch developed in the necked region saturated at a much lower value than the LLDPE, another finding is that bilayers deformation became more homogeneous than the single polyethylene and the degree of necking has decreased with increasing rubber thickness (Fig. 1).

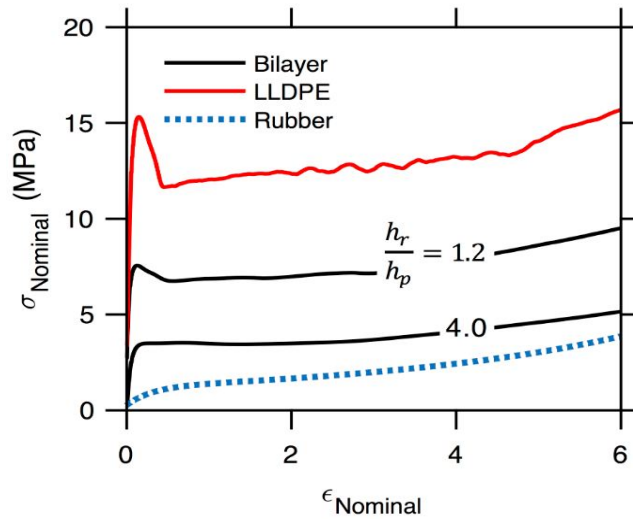


Figure 1 The nominal stress-strain response for rubber, LLDPE, LLDPE-Rubber bilayers of rubber:plastic thickness ratio 1.2 and 4.0 stretched at a rate of 120 mm per minute.

This conclusion is on LLDPE-Rubber multilayer and clearly prove the rubber component can change the mechanical behavior of plastic material greatly.

In this thesis, we seek to conduct similar experiments with high density polyethylene (HDPE) as the plastic layer. High-density polyethylene (HDPE) is a polyethylene plastic made from petroleum, which is known for its large strength to density ratio. HDPE has been widely applied in packaging industry because its low cost, light weight and good chemical resistance.

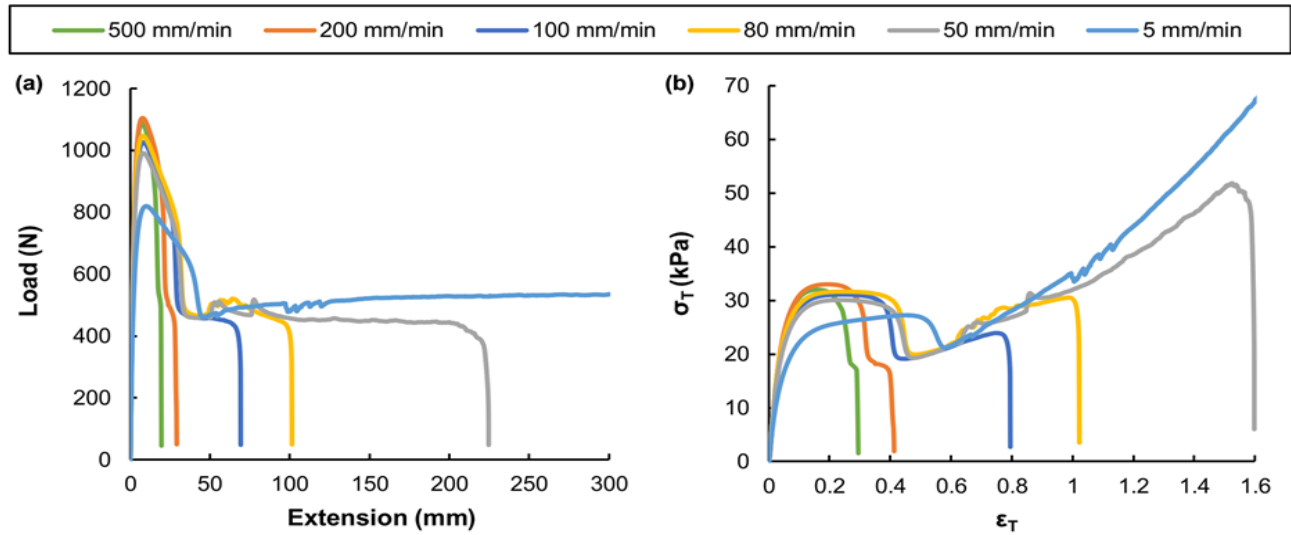


Figure 2 (a) The extension-load curves (b) true stress-true strain curves of HDPE at different cross head speeds.

However, comparing with other polyethylenes, HDPE is relatively brittle, especially at high rates of deformation. Fig. 2(a) shows the force vs elongation curves for HDPE film at six different crosshead speeds taken from a previous publication[10]. At low strain rate, the samples showed stable drawing (5mm/min). Their dogbone shape sample is approximately 170 mm long, 19 mm wide, the gauge length is 57 mm. The HDPE film cracked at small strain if the crosshead speeds were higher than 50 mm/min and moreover the rupture occurred at smaller deformations with increasing strain rate. Later in this thesis, we will show that the HDPE selected in our research shows similarly brittle behavior at high rate. The central question for this research is whether bonding rubber to the HDPE can reduce the brittle characteristic of HDPE. Essentially, the new composite material may be able to sustain a higher strain without fracture than pure HDPE.

The quantitative details and conclusions will be discussed later in this thesis, the essential idea can be illustrated by showing three possible response of HDPE-Rubber composites in tensile test observed experimentally. Fig 3(a) shows that sometimes the HDPE/rubber composites break in a brittle manner similar to pure HDPE. Typically, we have found that this phenomenon happens because of poor sample quality. In some occasions, as Fig 3(c) shown, if there a composite was poorly bonded in some regions, the HDPE film was delaminated from the elastomer film. In such cases, the HDPE layer fractures and delaminates as the rubber keeps stretching. Fig 3(b) is the “best case” scenario where, the composite stretches without failure or delamination. This case shows that the brittle characteristic of HDPE can be reduced by rubber bonding.



Figure 3 (a) Cracked HDPE-Rubber composites (b) Drawing HDPE-Rubber composites (c)Delaminated HDPE-Rubber composites

Therefore, the goal of this thesis is qualitatively describe how elastomer layer modified the brittle characteristic of HDPE and also quantitatively analyze what factors have an influence on the toughening effect.

This thesis is organized as below: In section 2 we introduce the procedures of all experiments and a briefly introduce on Digital Image Correlation (DIC) video analysis method. Section 3 describe the results the HDPE/Rubber bilayer. These bilayers were made in our lab “by hand” using compression molding. In section 4, the results of HDPE/Rubber/HDPE trilayer structure composites will be discussed. These trilayers were made by co-extrusion in collaboration with the PolymerPlus company. In section 5, an overall summary will be presented.

2.0 EXPERIMENT METHOD

2.1 FABRICATION

Bilayers were prepared by bonding HDPE (DMDA 8007) films to SEPS rubber(styrene-ethylene/propylene-styrene) films using compression molding. The SEPE and HDPE making process are as below: Firstly, according to the desirable sample thickness, we chose different thick metal spacer, the spacer prevents the sample from further compressed when the sample film has been compressed to the same thickness as the metal spacer. We weighed material based on its density and area of the metal spacer. Then placed the material and the metal spacer on a metal plate and all material were placed inside the metal spacer. Another metal plate was placed on its top. In order to easily separate the film from the metal plate, we inserted mylar film between the material and the plate. 150°C was taken as the temperature used during compression molding. Then we used methanol to eliminate air bubble between two single layers, waited overnight until there is no air exist between two layers and due to methanol has fully evaporated, we repeated the same compression molded process above to make two single layers bonded.

Trilayers films were made by PolymerPlus company using co-extrusion.

Dog-bone shaped samples (6 mm width and a nominal gauge length of 20 mm) were cut from the bilayer composite sheet. Considering about the thickness deviation of composites sheets, each dog-bone film has been measured again by micrometer caliper to assure the accuracy.

For the video analysis, small black glitter particles were then stuck onto the rubber side surface (for pure plastic sample, silicone oil was used to keep the surface sticky) to serve as markers for Digital Image Correlation (DIC) analysis.

2.2 TENSILE TESTS

For tensile test, the Instron tensile testing machine (model 31) was used in the range of 0 - 250N. The speed rate of this machine is from 0-1000 mm/min. The experiment temperature is room temperature. The tensile test was conducted at different crosshead speeds and recorded by camcorder. Similar experiments were conducted on the different thickness of the SEPS and the HDPE layers individually.

2.3 ANALYSIS BY DIC

Since the deformation of the material was not uniform along its length, the stretch profile on the sample surface needed to be computed for characterizing deformation. Finite element based interpolation technique was used to estimate the evolution of the stretch distribution on the

sample surface from the position of the finite number of marker points. Marker positions, in terms of pixel counts, were tracked at each frame of the recorded video of the specimen deformation by using Blender™ (Stichting Blender Foundation, Amsterdam, Netherlands) software suite. The marker positions from the first video frame was used to construct the reference configuration, which was a 2D finite element mesh of three-noded triangles with nodes located at the marker locations. The markers locations were triangulated, and then the stretch map was generated by evaluating the stretch in the axial direction of each triangular element. The process was repeated at all frames of the video recording to generate the stretch evolution with time, on the sample surface.

2.4 TRILAYER ACTUAL THICKNESS MEASUREMENT

In the extrusion process, the relative thickness of the various layers is controlled by specifying the flow rate of the extruders. The nominal thickness ratio of the films, and the nominal overall thickness is shown in the first two columns of Table 1. The corresponding nominal thicknesses may not be sufficiently accurate for this research. In particular, the extruded films had visible gradient in properties perpendicular to the machine direction and hence the nominal values cannot be treated as accurate. Therefore, it is necessary to measure the actual layer thicknesses before we test the trilayer. In this case, we used a mass-based measurement to estimate thickness. The procedure is as below: firstly, after a dogbone shape sample was cut from the extruded sheet, another rectangular piece was cut along the same flow line. The length and width of the rectangular sample was measured, its area calculated, and the film weighed.

Then this was placed into toluene for ten minutes. After ten minutes, most of SEPS rubber had dissolved and the two plastic layers could be separated easily. The small amount of SEPS still sticking onto the plastic films was removed by washing the plastic layers back into toluene for five minutes. The two plastic films were removed from toluene, wiped, and weighed. The HDPE density is 0.965 g/cm^3 , the SEPS density is 0.95 g/cm^3 . Both the original thickness of the trilayer and the thickness of the plastic was then estimated using the density of 0.96 g/cm^3 for both the rubber and the plastic.

As a crosscheck, thickness was also measured by using micrometer. We choose three points in each film, measured its thickness and used the average value for its thickness, which are also listed in table 1. From table.1 we can find there is a significant difference between nominal ratio and actual ratio, especially for ratio a rubber:plastic ratio of 2. There are also modest differences between the ratios measured by weighing and by micrometer. We believe that these are attributable the roughly 2-3 microns error in micrometer measurements, and hence prefer to use the mass-based measurement in calculations. We will continue to use the nominal ratio for designating samples in this thesis, but we will reiterate the difference between nominal and actual values.

Table 1. Actual Thickness of Extruded Trilayer Sample

mass-based measurement (micron)						
nominal ratio	nominal thickness	h_t	h_p		h_r	actual ratio
0.25	30	34	13	11	10	0.417
0.667	35	40	14	11	15	0.600
1	60	51	13	13	25	0.962
2	70	65	8	7	50	3.333
4	70	74	9	7	58	3.625
micrometer measurement (micron)						
nominal ratio	nominal thickness	h_t	h_p		h_r	actual ratio
0.25	30	30	12	10	8	0.364
0.667	35	37	12	10	15	0.682
1	60	47	13	9	25	1.136
2	70	61	9	7	45	2.813
4	70	73	8	7	58	3.867

3.0 BEHAVIOR OF HDPE/SEPS BILAYERS

We will first discuss force data measured during tensile testing experiments. After measuring the data in tensile test, for the data only in force-extension form, the formulas below were used to calculate the nominal stress and nominal strain:

$$\varepsilon = \frac{\Delta L}{L_0} \quad (1)$$

$$\sigma = \frac{F}{A_0} \quad (2)$$

Where ΔL is the sample elongation, L_0 represents the gauge length of the sample, A_0 is the undeformed cross-section area.

3.1 BEHAVIOR OF PURE HDPE

Before discussing the bilayer composites, we will examine two single layers first. As Fig.2 shown in the previous chapter, pure HDPE is brittle and breaks at small strain when strain rate is 50 mm/min or higher. We find similar results in our experiment. At the strain rate of 10 mm/min, one sample tested failed while another sample showed drawing behavior, but this

drawing stopped and sample failed at 20mm elongation. At the stretching speed of 40 mm/min, HDPE was somewhat brittle: three of five specimens tested failed (Fig. 4a), whereas two showed stable drawing (Fig. 4b). At higher speeds, all HDPE samples were brittle and failed at a small nominal strain (Fig. 4c). In brief, if the speed below 40mm/min, the compression molding HDPE sample may show two behaviors (drawing/ductile behavior or brittle behavior), once the strain rate higher than 40mm/min, failure of the specimen could be foreseen.

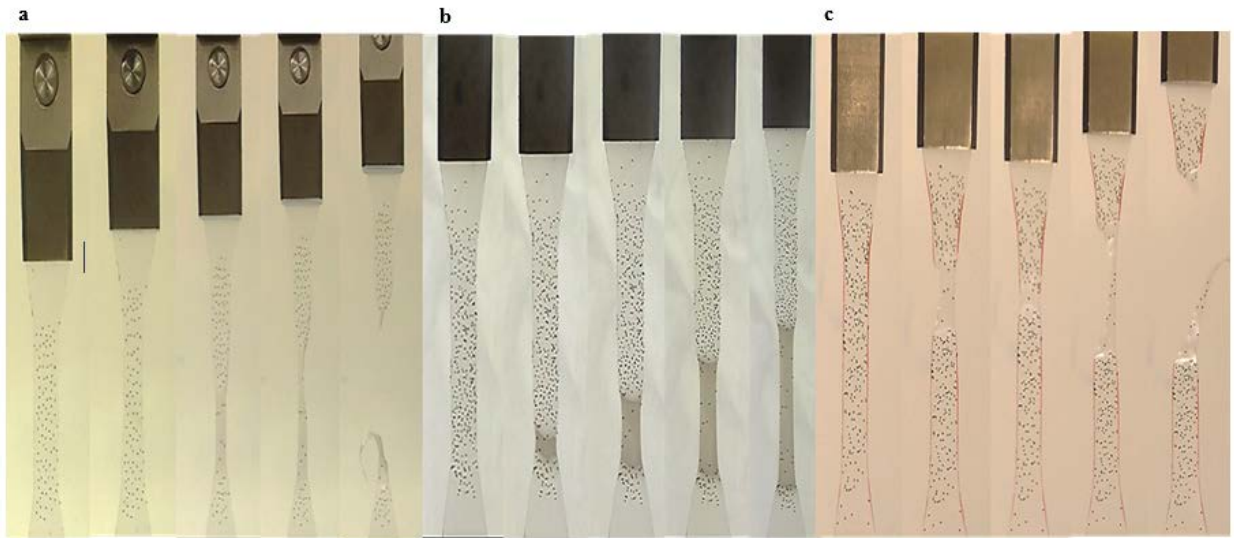


Figure 4 Images of samples during tensile deformation of dogbone-shaped samples of HDPE plastic. (a) HDPE plastic at 40 mm/min (brittle behavior). (b) HDPE plastic at 40 mm/min (drawing behavior). (c) HDPE plastic at 900 mm/min (brittle behavior)

Representative mechanical data (stress-strain curves) for such behaviors are shown in Fig. 5. Stable drawing is indicated by the plateau in the nominal stress-strain curve. Such a plateau is evident in two samples: one at 10 mm/min and one at 40 mm/min. All other samples

fail at small strain. All samples show a peak in force due to necking, since the neck has a smaller cross-sectional area than the original sample, the total force reduces upon neck initiation, and so does the nominal stress. Since the decrease in nominal stress is primarily attributable to a decrease in cross sectional area at the neck, it is sometimes called geometric softening[1]. According to our video record, the force curve began to drop at the same time when the sample started necking. The force is the product of the true stress and the true cross-sectional area. Obviously the cross sectional area must reduce as the sample is stretched. Consistent with the Considere criterion[1] when the force expected in homogeneous deformation decreases, necking must start. Our observation that necking starts when the force reduces is consistent with this.

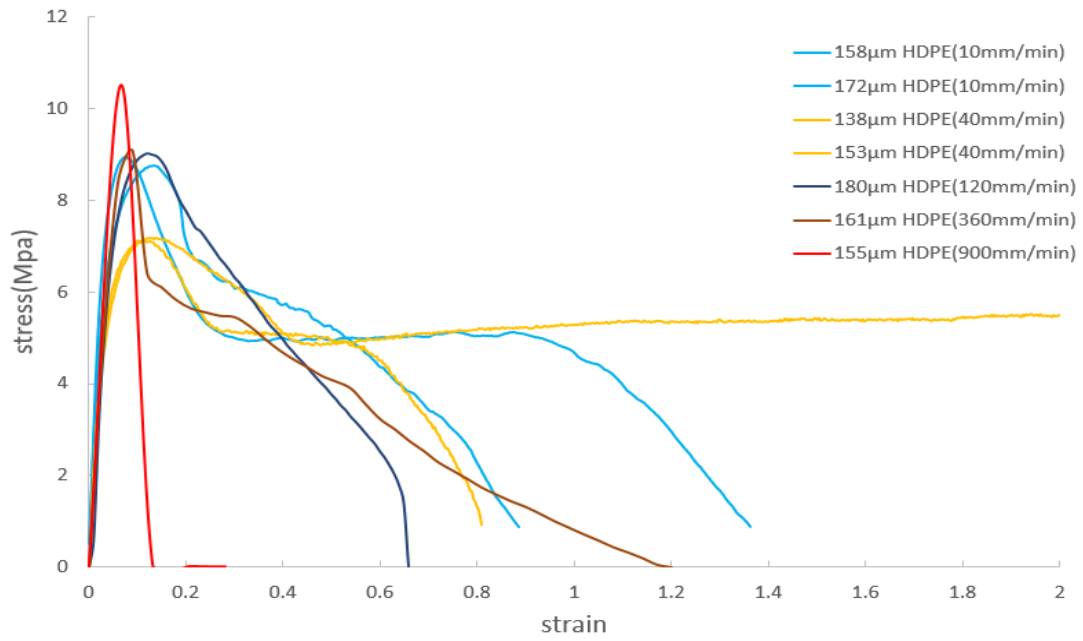


Figure 5 Strain-stress curves of compression molding HDPE under five different strain rates.

3.2 BEHAVIOR OF PURE SEPS

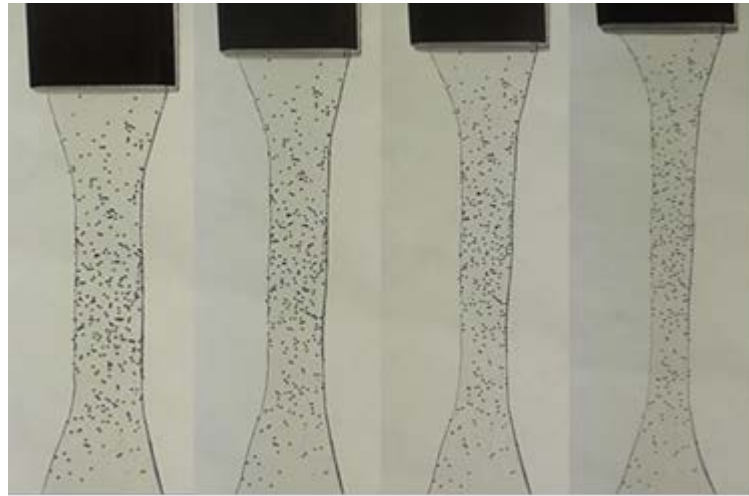


Figure 6 Images of rubber being stretched at 40 mm/min. Appearance at 900 mm/min is very similar and not shown

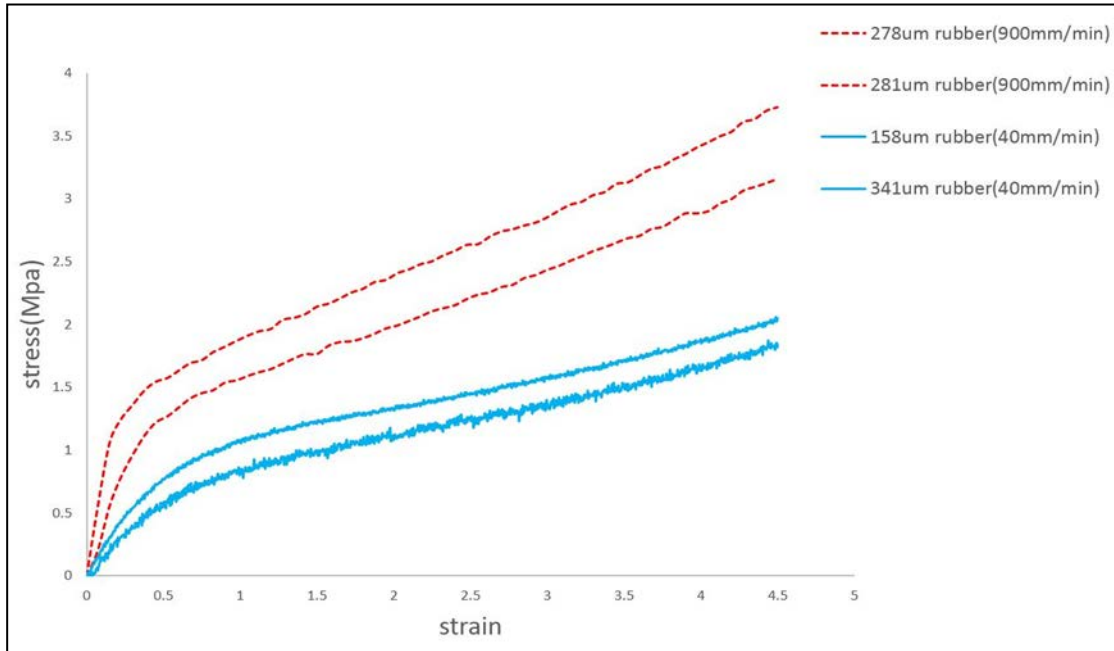


Figure 7 Stress-strain data for SEPS rubber at 40 mm/min and 900 mm/min

Different from HDPE, the rubber's deformation process is homogeneous without localized stretching (Fig.6). Fig.7 shows, that unlike HDPE, two SEPS show a similar trend in stress vs strain curve, and at either speed, the SEPS can be stretched significantly without failure. The stress increases somewhat with increase in strain rate, although the shape of the curve remains unchanged.

3.3 BEHAVIOR OF SEPS/HDPE BILAYERS

Fig.8 is the image of the SEPS/HDPE bilayers in the tensile test at 40mm/min and 900mm/min. The bilayers were drawing in both strain rate, there is no failure of the specimen, even at the speed of 900mm/min, nearly the highest speed of the Instron machine, the samples show over 90 mm elongation during stable drawing without failure. The extension-force curves under two strain rates were plotted in Fig.9(A) and Fig.10(A), the corresponding strain-stress curves in Fig.9(B) and Fig.10(B). For comparison, the two sets of HDPE response (one which draws, and the other which fails) from the previous Fig.5 and Fig.7 are repeated, as well as the data for the pure rubber. Typically, all samples have intermediate mechanical behavior between the plastic and the rubber. One characteristic is for all bilayers in the same speed show a similar deformation process, firstly composites show a sharp rise in stress at small strain, followed by a peak, eventually show a plateau. The most important result from Fig.10 is that at 900 mm/min, even the minimum rubber thickness ratio (95 micron SEPS) is able to prevent the HDPE layer from rupture. Surprisingly, the qualitative behavior is weakly dependent on rubber:plastic ratio. Almost all samples show a peak in the force-elongation data suggesting that in almost all cases, a neck develops. There is some trend at high speed that the peak is weaker as rubber thickness increases. Moreover, the draw stress in 900mm/min is obviously higher than in 40 mm/min, the peak stress value is between 3 to 4Mpa in low strain rate while between 6 to 7Mpa in high strain rate condition. The composites behavior is more depending on strain rate rather than the ratio of two single layers.

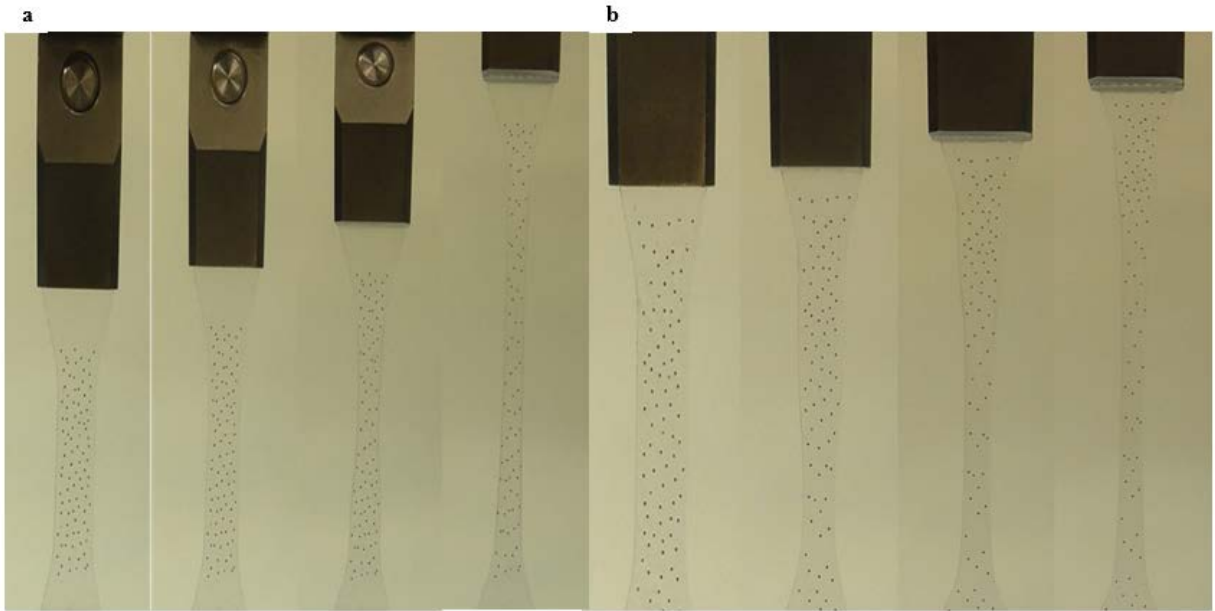
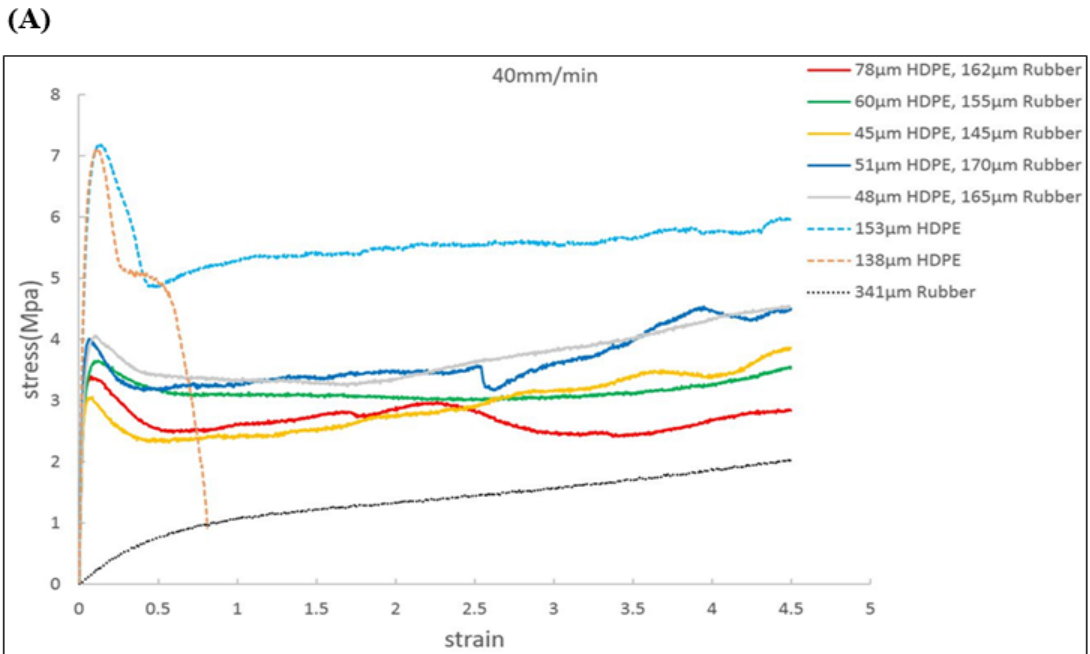
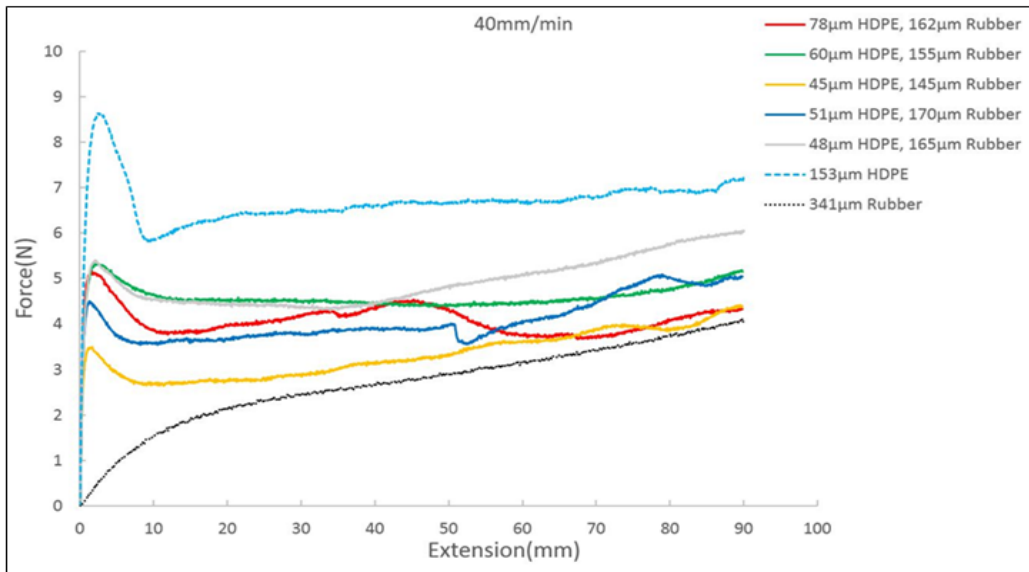


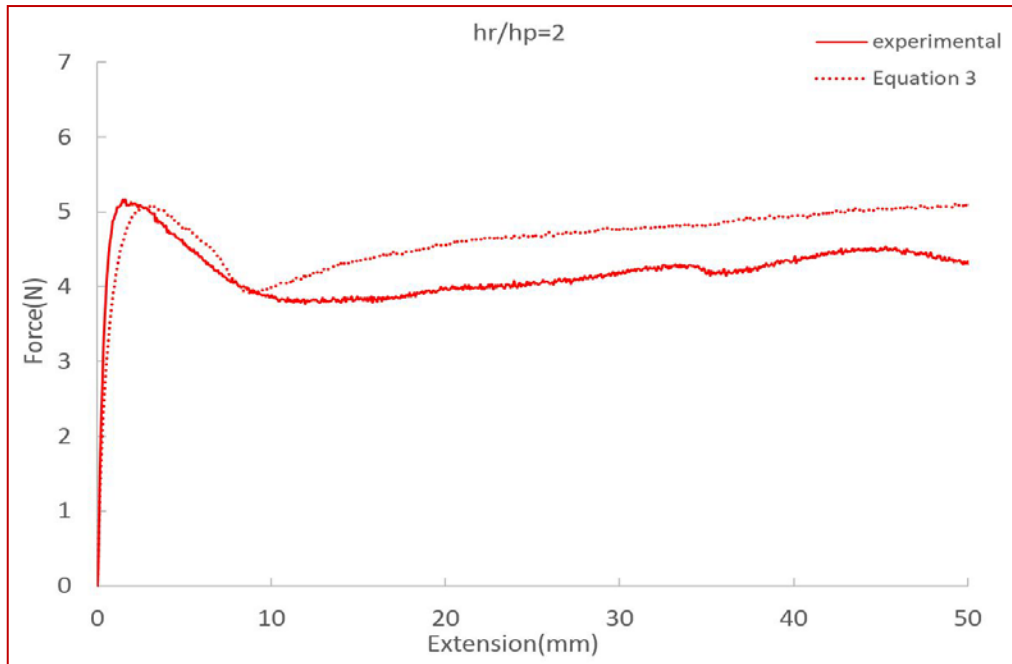
Figure 8 Appearance of bilayer of thickness (a) 78µm HDPE, 162µm rubber measured at 40 mm/min,
 (b) 62µm HDPE, 131µm rubber measured at 900 mm/min.



(B)



(C)



(D)

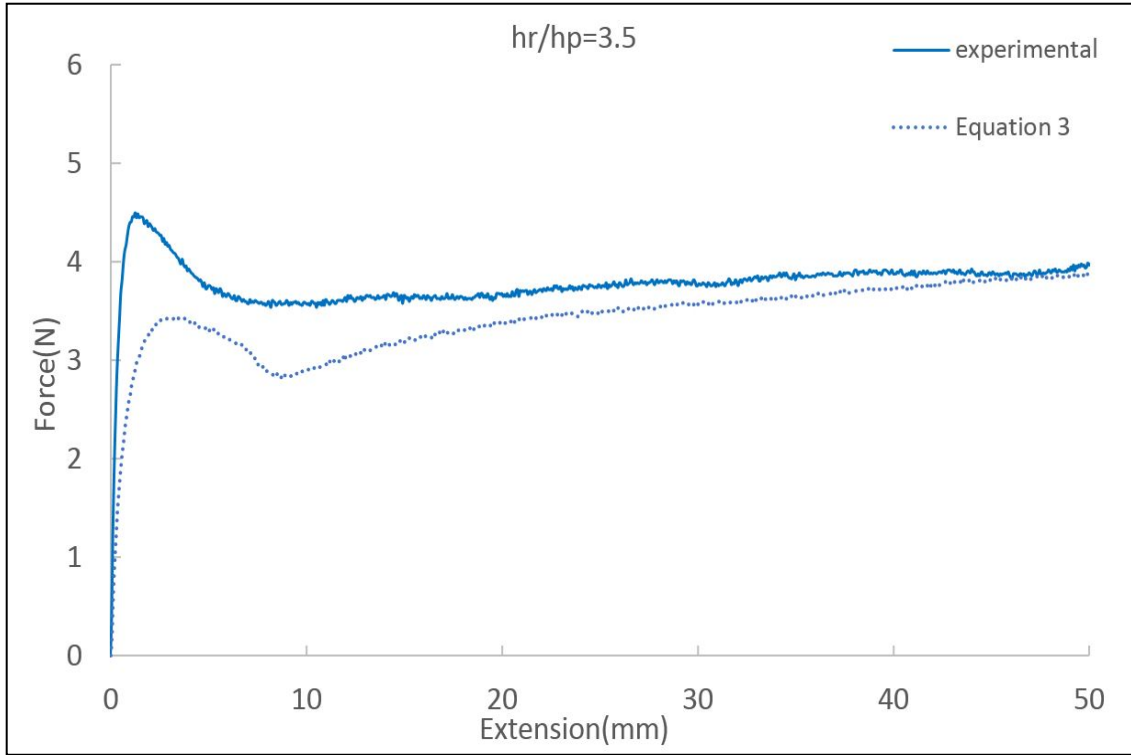


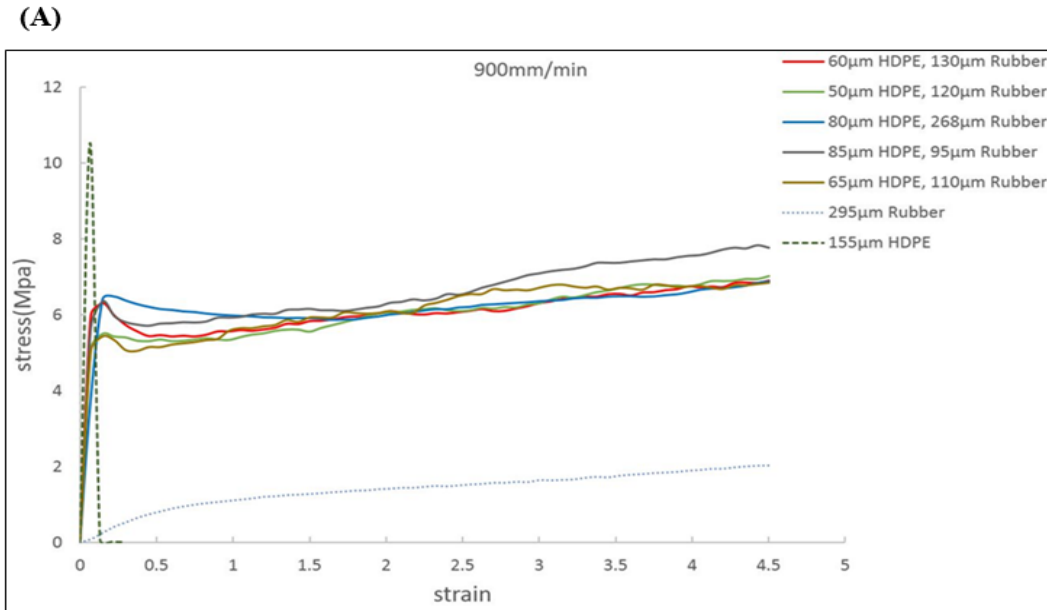
Figure 9 Strain-stress curve of HDPE and its composites, dot lines mean the single layer at 40 mm/min. B. Force vs elongation for the same samples. C-D. Bilayer Force-Extension of experimental data versus calculated data based on equation 3.

It is interesting to compare the measurements with a simple model of a layered composite, which is to treat the total force as a sum of the force in each layer.

$$F_{\text{bilayer}} = F_r + F_p = w[h_r\sigma_{\text{nom},r} + h_p\sigma_{\text{nom},p}] \quad (3)$$

where h_r and h_p are the rubber and plastic layer thickness respectively, w is the sample width, and $\sigma_{\text{nom},r}$ and $\sigma_{\text{nom},p}$ are the nominal stresses for the rubber and plastic measured independently at the same nominal strain. The predictions of equation (3) are shown in Fig. 9(C)

and (D) as dashed lines. By comparing the prediction curves with experimental curves, we found that those curves were developed in a similar trend, which means the properties of bilayer are approximately equal to the combination of two single layers properties.



(B)

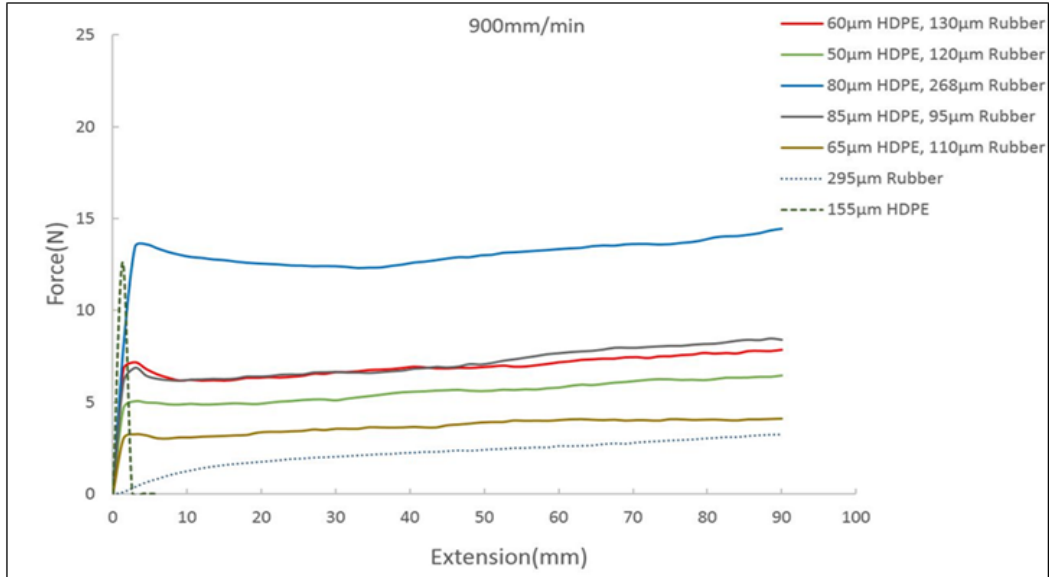


Figure 10 (A) Extension-Force curve with two strain rates (900mm/min), the rubber: plastic thickness ratio is approximately equal to 2, 2.5, 3 and 3.5. (B) corresponding Strain-stress curve.

To further quantify the data, Fig. 11 plots the draw stress and we see that it is nearly independent of rubber/plastic ratio. The draw stress is the minimum stress value after the peak nominal stress and HDPE draw stress only in drawing case. Note that draw stress at 900 mm/min could not be measured due to the film always ruptured without stable drawing after the peak in the force.

Furthermore, there are also differences in the degree of homogeneity of the stretching. This is shown in Fig 12 which shows snapshots from the videos recorded at 900 mm/min at rubber:plastic ratio of roughly 1:1 and roughly 4:1. We have seen the similar trend in LLDPE/Rubber bilayers: the non-homogeneity of deformation reduces as rubber thickness

increases. This trend is quantitatively described by using DIC in the chapter four. Here the figures are shown only for illustrating that the degree of homogeneity changes with rubber thickness ratio.

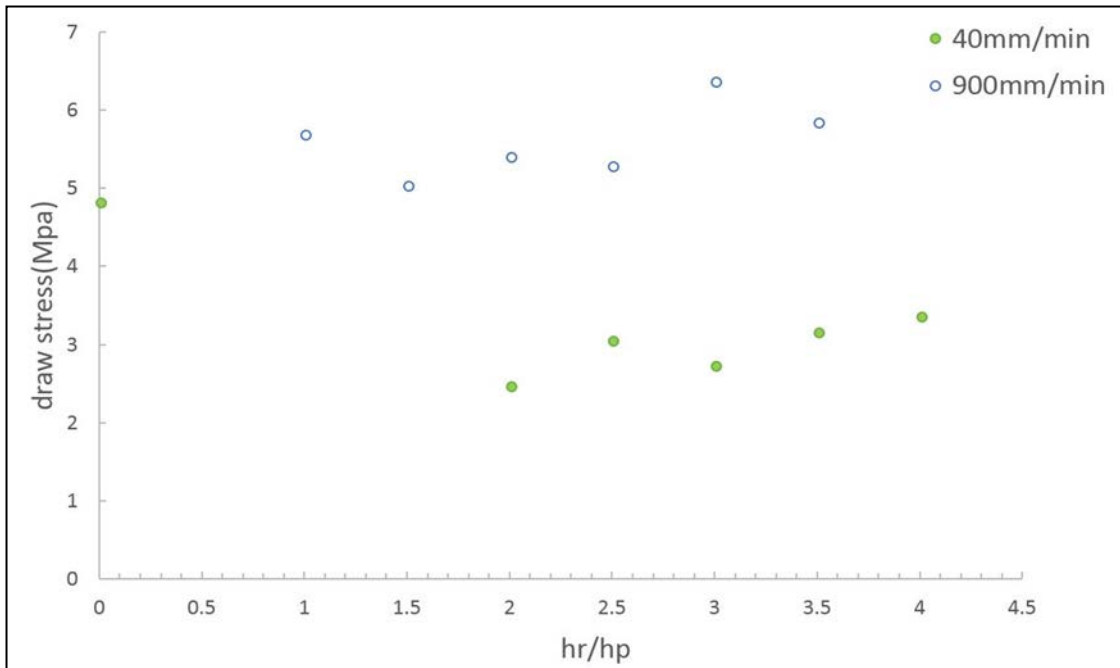


Figure 11 The draw stress of different plastic:rubber ratio thickness at two speeds. The blue hollow dots represent the high speed and the green dots represent the low speed.



Figure 12 Snapshots at 900 mm/min showing differences in drawing. The rubber:plastic ratio of upper is roughly 1:1 and roughly 4:1 of the bottom one.

Finally, the most direct measure of toughening is to examine the mechanical work done in the deformation process, i.e. the area under the force-elongation curve. To compare across all samples, this work should be normalized by the sample volume, which corresponds to energy dissipation or the work done per unit volume.

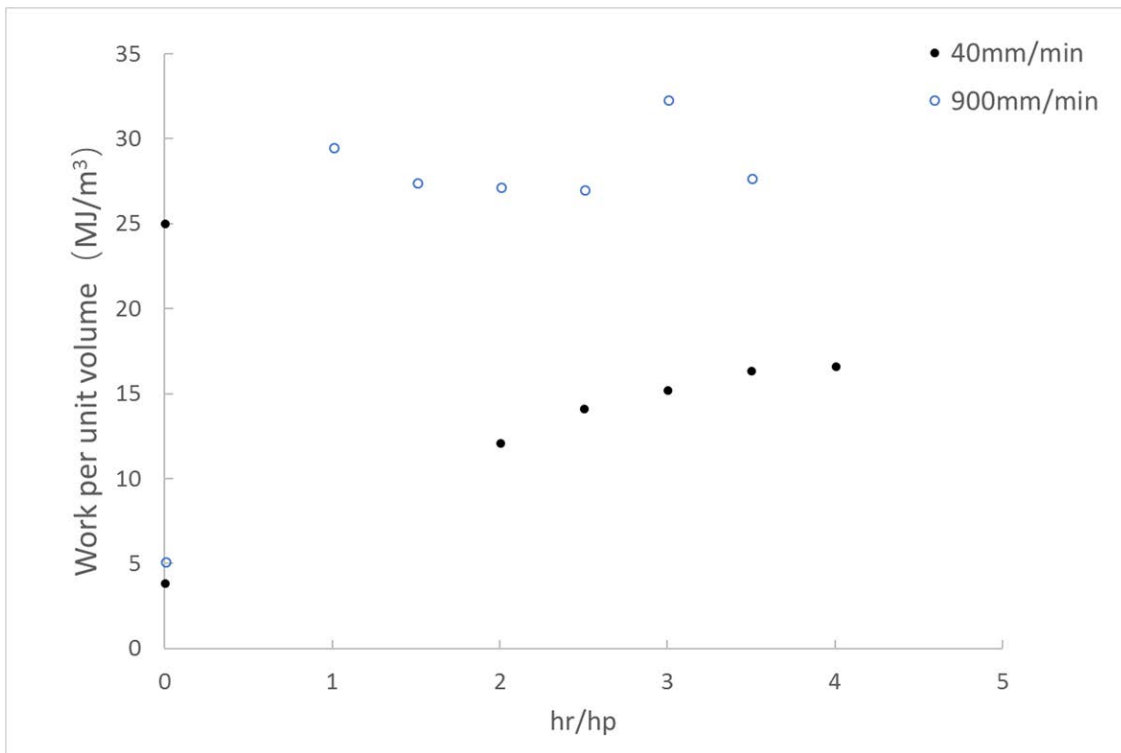


Figure 13 The mechanical work needed when bilayers come to 90 mm elongation in different rubber:plastic thickness ratio. The work calculation for HDPE work is based on its highest elongation before failure.

The work per volume was calculated by integrating under the nominal stress vs nominal strain curve numerically using the below formula:

$$w = \frac{1}{2} \sum_{i=1}^n (\sigma_{i+1} + \sigma_i) \cdot (\varepsilon_{i+1} - \varepsilon_i) \quad (4)$$

Where we choose 150% strain as the integration limit (which corresponds to 90 mm elongation) and n is equal to 1353 here, which corresponds to the number of data points recorded.

The work per volume is shown in Fig. 13. The green dots represent the low speed and blue hollow dots represent the high speed. Obviously for samples that are brittle (pure HDPE at 900 mm/min or sometimes also pure HDPE at 40 mm/min), the work calculation gives a very low number. Apart from the difference between pure HDPE and the bilayers, there is no significant trend. If the HDPE behaves in a ductile fashion, then the bilayers have reduced work per unit volume as compared to HDPE, chiefly because the rubber develops low stress. In all other cases when HDPE is brittle, the bilayers have much higher work per unit volume because their failure is delayed to high strain.

However, during the experiment process, we noticed that there is a great number of disadvantages in bilayers which limitations are definitely constrict the further study. Firstly, the quality of bilayers is difficult to control, and hence the sample-to-sample variability is relatively big. Secondly, bilayers are asymmetric, i.e. the neutral axis from a mechanical viewpoint is not the same as the geometric center axis. Furthermore, it is difficult to reduce the layer thickness below about 25 microns by compression molding. Furthermore, sample preparation is very tedious and not scalable.

For all these reasons, we now switch to trilayer composites made by co-extrusion. These were kindly made by PolymerPlus company. The extruded trilayers consist of SEPS and DMDA-8007 HDPE, which are the same materials as used in bilayer. A sheet of HDPE was also extruded under the same conditions. In next chapter, we will discuss the mechanical behavior of trilayers in detail and further analyze the local stress when the films deforming by DIC.

4.0 MECHANICAL BEHAVIOR OF HDPE/SEPS/HDPE TRILAYER

In the previous chapter on rubber-plastic bilayer, we quantitatively described the mechanical properties of bilayers and showed that the elastomeric layer can effectively reduce the brittle characteristic of HDPE. The calculation of the area under the load deformation curve also suggests that the energy absorbed of composites is much more than both the rubber and the plastic. Therefore, the composites have a considerably potential value for application of improving toughness.

The focus of this chapter is still on modification of brittle behavior of the plastic due to rubber. However, before discuss the mechanical properties of trilayer, it is necessary to present the characteristic of the trilayer sheets. The samples are available as rolls of roughly 25cm width. The samples thickness is not uniform along the width while they are relatively uniform along the machine direction, so all samples were cut at the same width location. Furthermore, although the HDPE is the same as used in the previous chapter, its mechanical behavior is different from the HDPE that was compression-molded we made in our lab. The extruded HDPE invariably shows brittle behavior in tensile test, no matter whether the strain rate is 40mm/min or 900mm/min (in Fig.14 (a) and (b)). This is in contrast to the compression-molded HDPE which could draw at low rates but is brittle at high rates. We suspect that the reason for this is orientation developed in the film during extrusion. Since the polymer in the extruded films is already oriented, it is harder for the sample to draw further in a tensile test. To demonstrate this, a simple test on

extruded HDPE was conducted. A metal plate was heated to 150 Celsius, and coated with a thin layer of polyethylene glycol liquid for preventing the sample from sticking to the plate. A 4 cm long extruded HDPE strip was placed on the top of the plate. After having been heated 1 minute, we found that there is a 0.3 cm shrinkage of the HDPE sample along the machine direction, consistent with orientation of extruded HDPE.

From Fig. 14 (c) and (d), both the extruded HDPE at different strain rate ruptured at the small strain. Further support comes from the observation that the yield stress of the extruded HDPE is more than 15 MPa, which is higher than the approximately 8Pa of the molded material of the previous chapter.

For the same extruded HDPE, the yield stress of the 900mm/min is higher than 40mm/min, similar trend as the compression-molded HDPE. Since the plastic is already brittle at 40 mm/min rate, it is not necessary to study both high and low strain rates. Therefore, in this chapter, the strain rate was not varied; all experiments were conducted at 40 mm/min.

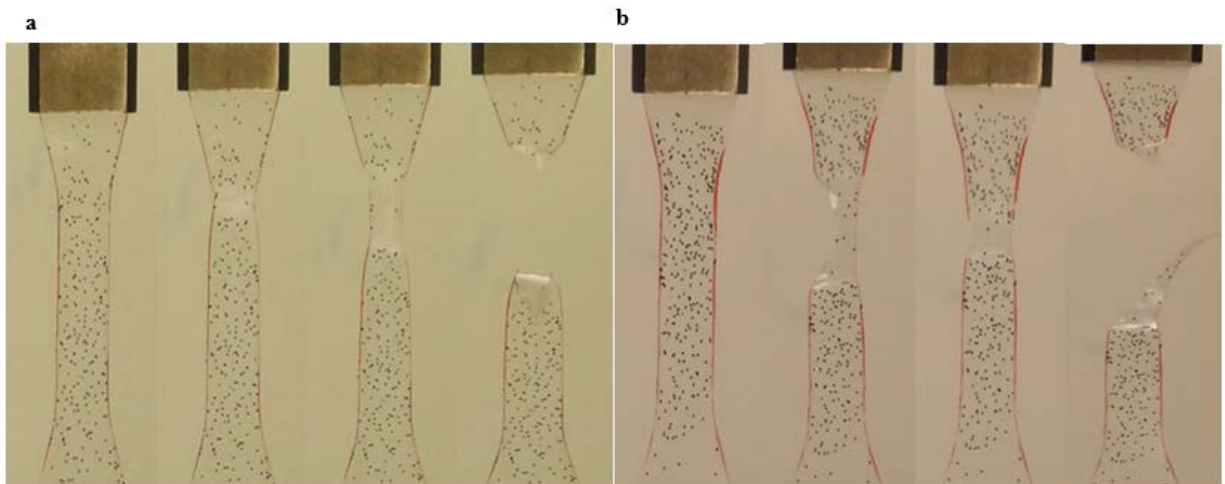


Figure 14 (a) Extruded HDPE at 40mm/min (b) Extruded HDPE at 900mm/min

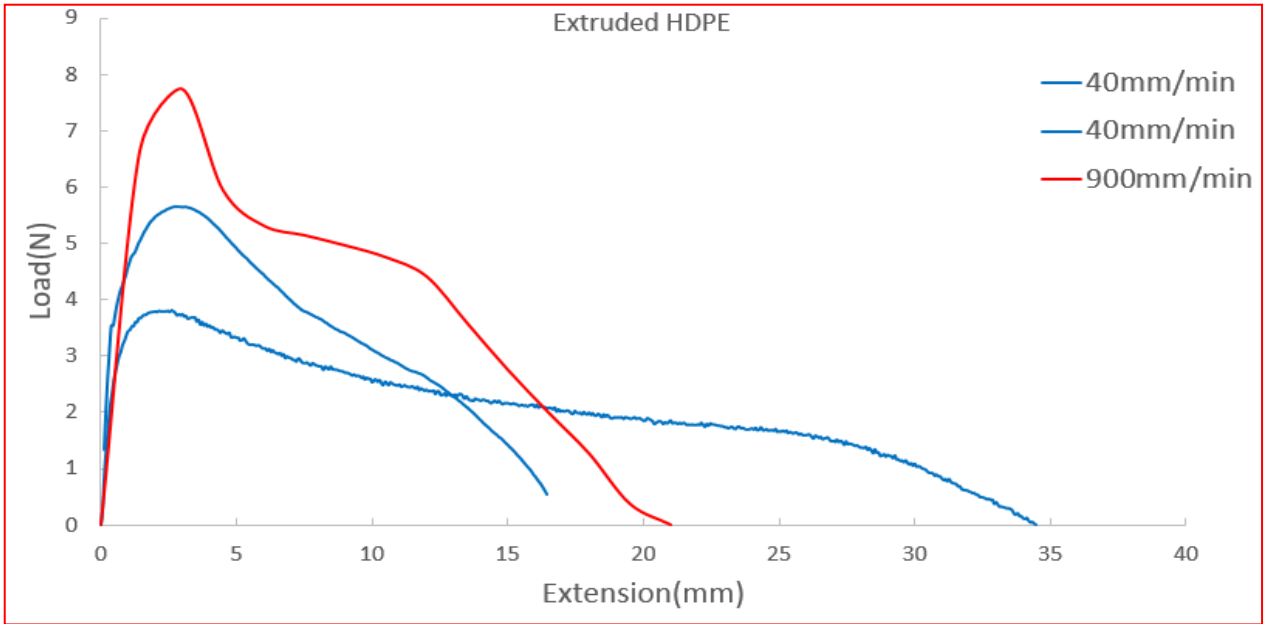


Figure 14. (c) Extension-load curve of extruded HDPE at two strain rates.

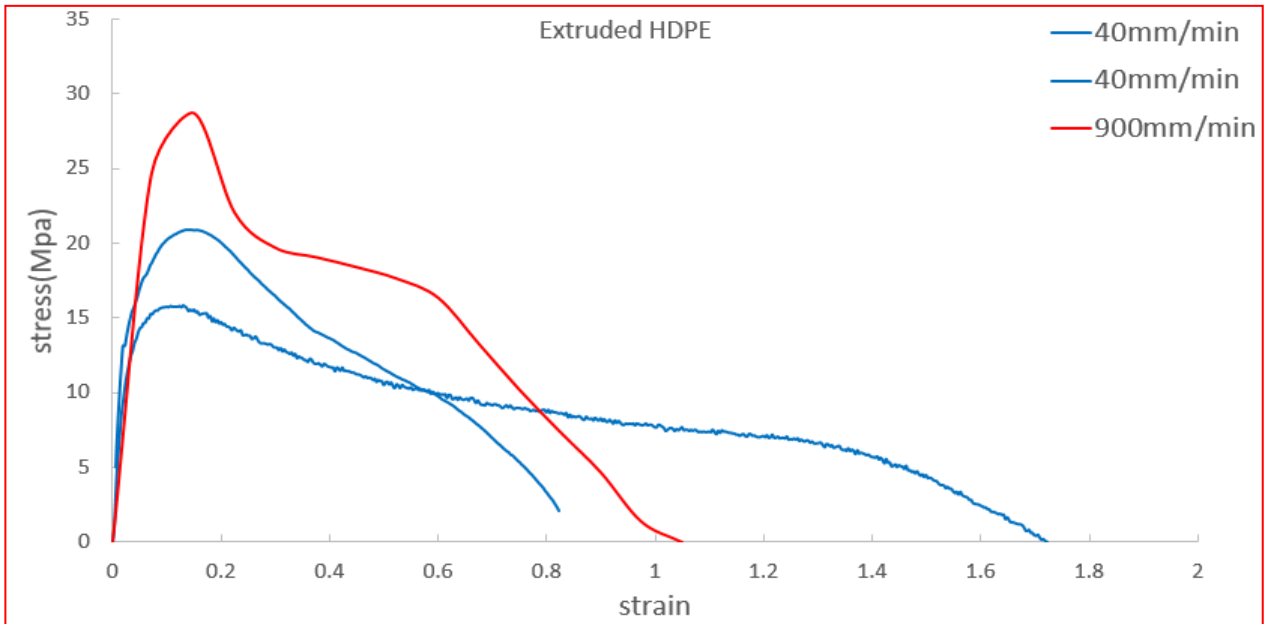


Figure 14. (d) Strain-stress curve of extruded HDPE at two strain rates.

4.1 MECHANICAL PROPERTIES

Similar to the previous chapter, this part begins with a discussion of the strain-stress curve of the trilayer composites. As in the previous chapter, measurements were conducted on dogbone-shaped samples. Due to the expected sample orientation, we must be consistent about directions and hence all samples were cut such that the tension was applied along the machine direction. We have five different ratios of trilayer as listed in Table 1, whose load-extension curves are shown in the figure 15. Each sub-figure in Fig. 15 shows two measurements for each sample.

Figure 15 confirms the result of the previous chapter: the trilayer composites also can be stretched to a higher degree without failure as compared to the pure HDPE extruded sample that was shown in Fig.14. Of all the twelve samples tested, only one (Fig. 15 a) failed at small elongation. These results suggest that even in the case of the smallest rubber:plastic ratio, the rubber can reduce the brittleness of HDPE greatly. It should be noticed that some curve showed abrupt drops during the drawing process, this phenomenon was presented in Fig. 15 a and 15 c. The videos suggest that such drops are due to the multiple necking happen on the sample (Fig.16). The formation of a second neck causes the nominal stress to reduce, and then increase again back to the stable drawing value. The multiple necking is not the main concern of this thesis, we only explain why the curve drop here.

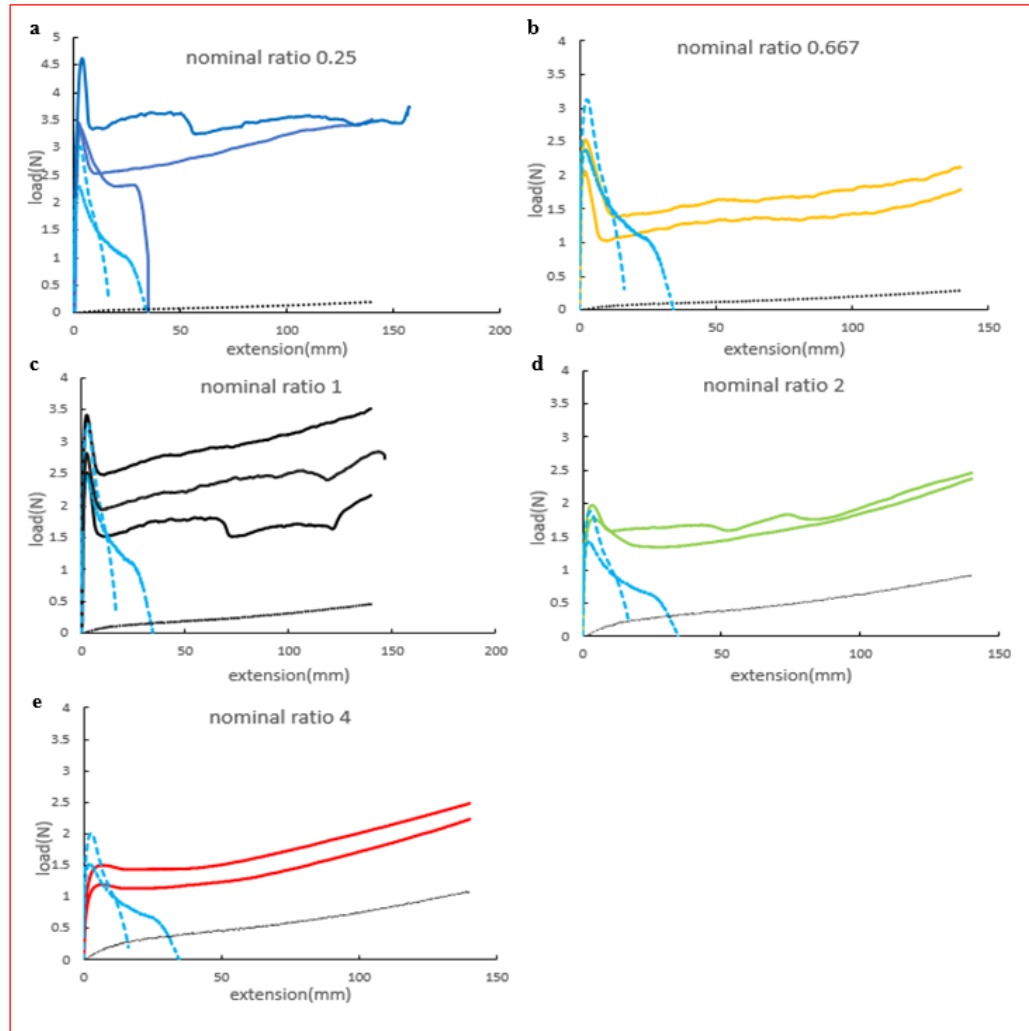


Figure 15 (a-e) Extension-load curve of trilayer measured at 40 mm/min.



Figure 16 The images of multiple necking happened.

In addition, for comparison each sub-figure also shows the force expected from a free-standing plastic layer of the same total thickness as the two plastic layers in the trilayer, and free-standing rubber layer of the same thickness as the rubber layer in the trilayer. Until the 30mm extension at which the pure HDPE went ruptured, we noticed that the F_{total} is always slightly higher or almost equal to F_p , where the subscript “p” indicates plastic. At the small extension condition, most of the force needed to stretch the trilayer comes from the plastic, there is slightly dependent on rubber layer, so the trilayer and HDPE has similar forces and peak loads. However, at the high extension condition, it is difficult to compare them directly because of the single HDPE cannot be stretched to that degree. Yet, at least at the lowest rubber:plastic ratios, there is no doubt that the force in the trilayer far exceeds the force developed in the rubber layer. This suggests that most of the force needed to stretch the trilayer comes from the plastic; the main role of the rubber is to prevent the failure of the plastic.

In brief, from Fig.15, we believe that the trilayer performance is similar to pure HDPE at small strain where the HDPE does not show brittle behavior, but at the high strain where the HDPE always fails, the trilayer performance is better than simply bonding two layers together. We plotted the nominal plastic stress of trilayer at 100mm elongation condition in Fig.17, the HDPE can't be stretched to 100mm, but the trilayer and the rubber data at 100mm is available, therefore, we assumed $F_{total} = F_p + F_r$ and calculated the nominal plastic stress in 100 mm condition. With nominal rubber:plastic ratio increasing, the plastic stress decreases accordingly.

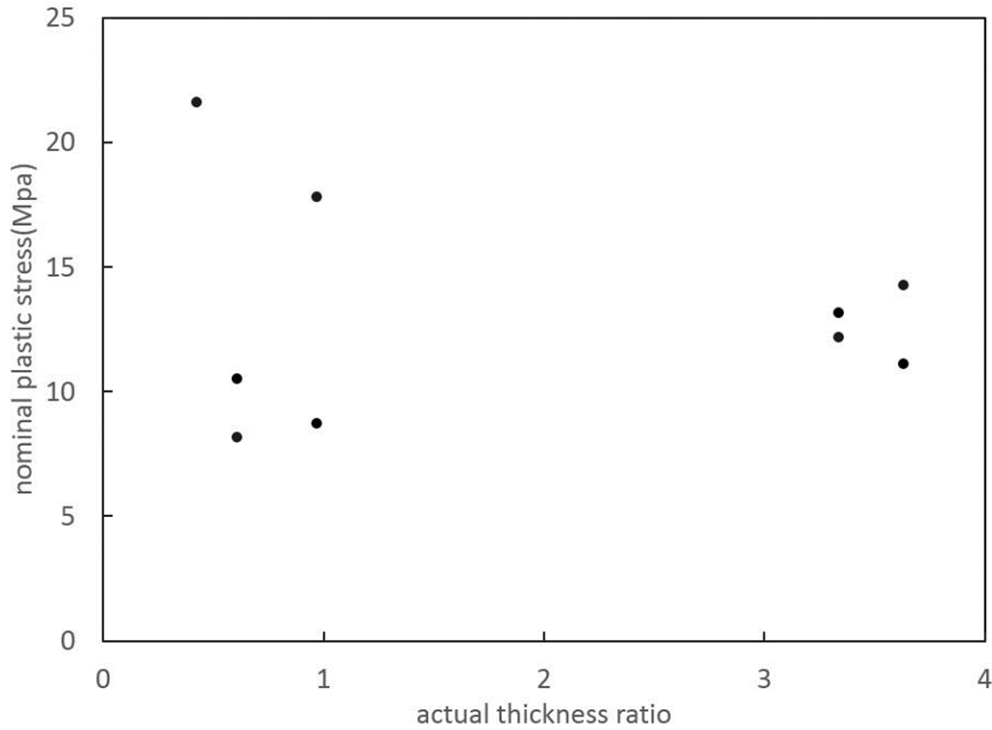


Figure 17 The $\sigma_{nom,p}$ of different actual thickness ratio under the 100mm extension.

The total thickness of the samples of Fig. 15 are not equal, making it difficult to compare them to each other. Therefore, we turn to the nominal stress-strain curves calculated from the load-elongation curves. But by converting extension-load data into nominal strain- nominal stress data, in fig.18, some trends become obvious. Firstly, the degree of necking decreases with increasing rubber thickness ratio. For example, for $h_r/h_p = 0.25$, there is a sharp decrease of stress after the peak, as expected for a plastic material undergoing necking. For $h_r/h_p = 4$, these curves are almost completely flat, indicating a decrease in necking behavior. Secondly, the

stress near the peak as well as the stress during drawing reduces as plastic layer thickness increases.

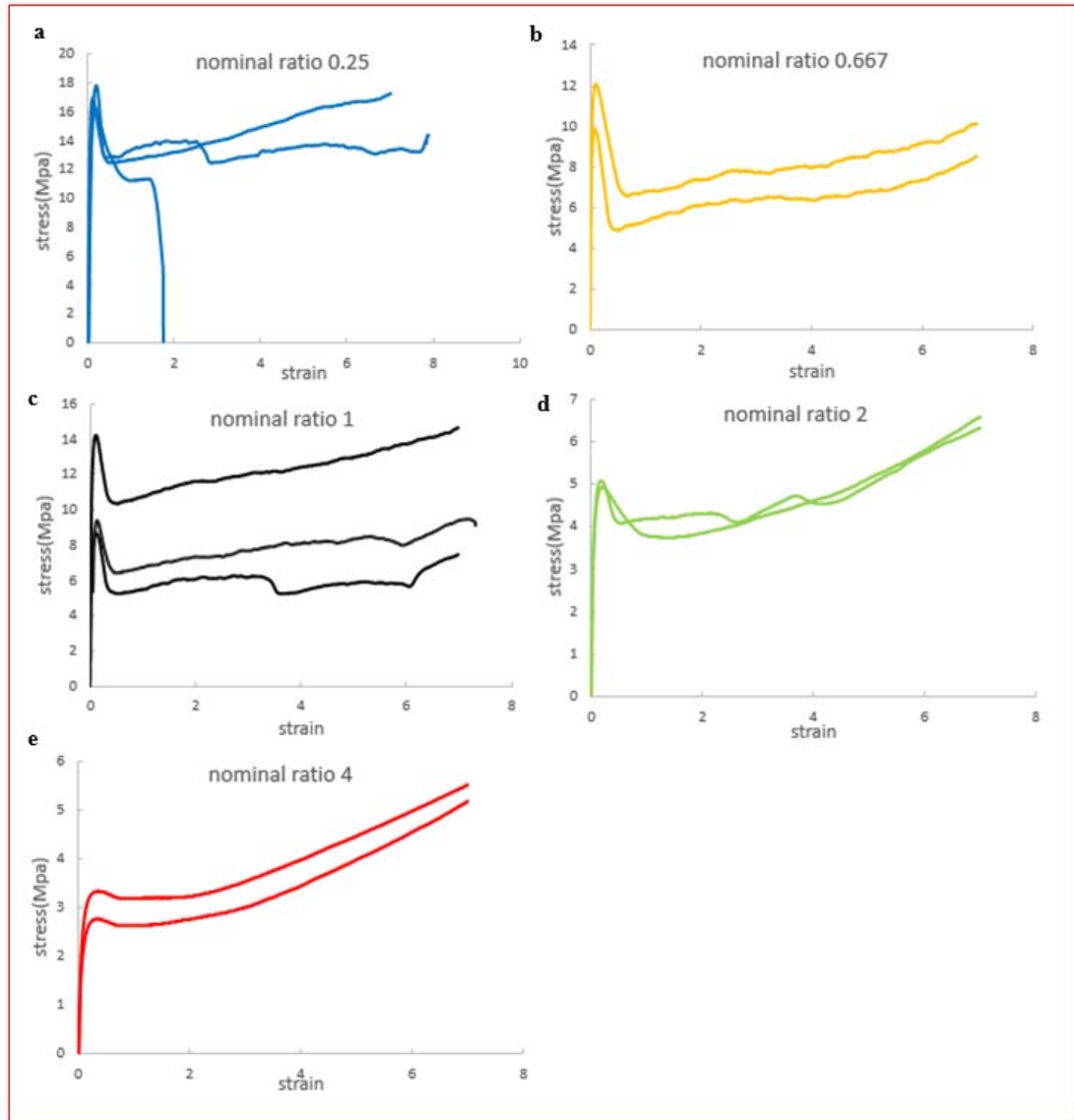


Figure 18 (a-e) Strain-stress curve of trilayer measured at 40 mm/min.

To illustrate the latter point, we estimated the nominal draw stress for trilayers and plotted them in Fig.19. It should be noted that no point is indicated for $x=0$, because pure HDPE samples did not show stable drawing, and hence their draw stress could not be estimated.

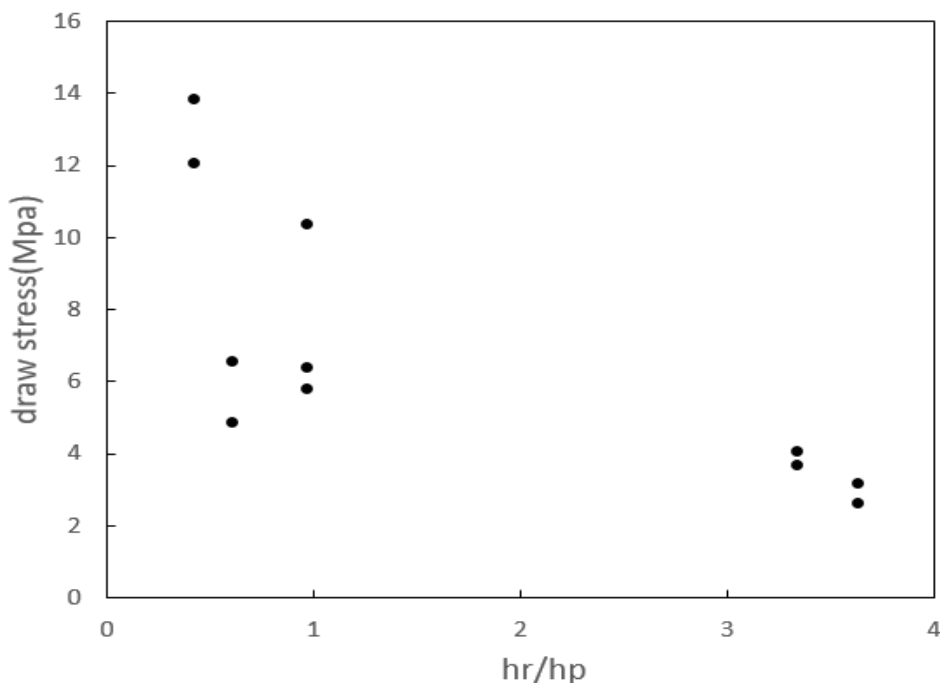


Figure 19 Draw stress of trilayer in different rubber:plastic thickness ratio

Similar to the previous chapter, to quantify toughness, we calculate energy dissipated per unit volume (i.e. work done) in Fig.20 as before integral of area under the stress-strain curve. The result of mechanical work absorption of trilayers will be discussed here. The first conclusion is whether one layer or two layers HDPE, the rubber layer always helps sustain a higher strain without failure, therefore, both bilayer and trilayer composites can absorb more mechanical energy in the tensile process than pure HDPE. Second, due to the small amount of rubber can

reduce the brittleness of HDPE effectively (nominal ratio 0.25), and as we mentioned below that with increasing rubber thickness will reduce the draw stress, the energy absorption also has an inverse correlation with rubber thickness. However, there are no points between $h_r/h_p = 1.5$ to $h_r/h_p = 3$, more experiments in this range are necessary in order to making the conclusion solid and comprehensive.

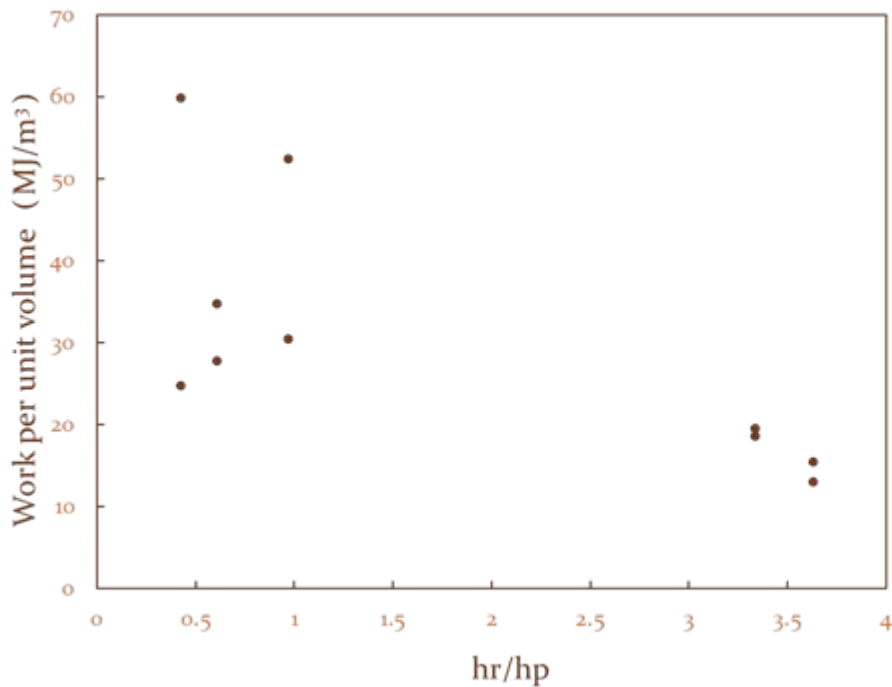


Figure 20 The mechanical work need when composites come to 90 mm elongation in different rubber:plastic thickness ratio.

4.2 DIC FOR TRILAYERS

We have discussed the nominal stress of trilayer. Since the deformation of the specimen was not uniform along its length, the corresponding stretch distribution was not uniform. Furthermore, as shown in Fig. 12, for the bilayer, the homogeneity of deformation changes with different rubber thickness. We also find this trend in trilayer sample. Fig.21 (a-b) show two extreme examples of $h_r/h_p = 0.25$ and $h_r/h_p = 4$ during stretching. By comparing these two ratios, we find that the sample with the nominal ratio of 0.25 (Fig. 21a) shows unambiguous necking. In contrast, we find that at the nominal ratio of 4 (Fig. 21b), deformation is roughly homogenous and the necking is as not as obvious as the left image.

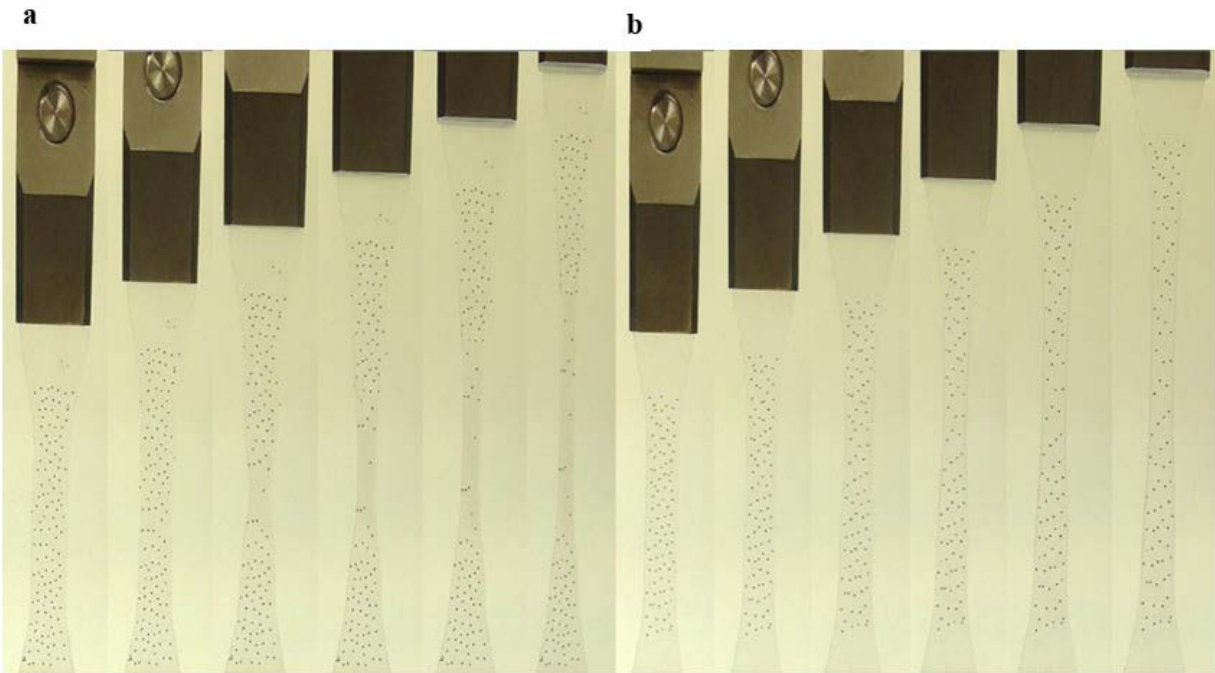


Figure 21 (a-b). a is nominal ratio 0.25 sample, b is nominal ratio 4 sample.

4.2.1 Qualitative behavior of tensile deformation

Generally, the homogeneity of deformation increases as rubber thickness ratio increases. Therefore, to the local strain of composites during the tension process, we applied the DIC method to track the local strain and plotted the result in Fig.22(a-d).

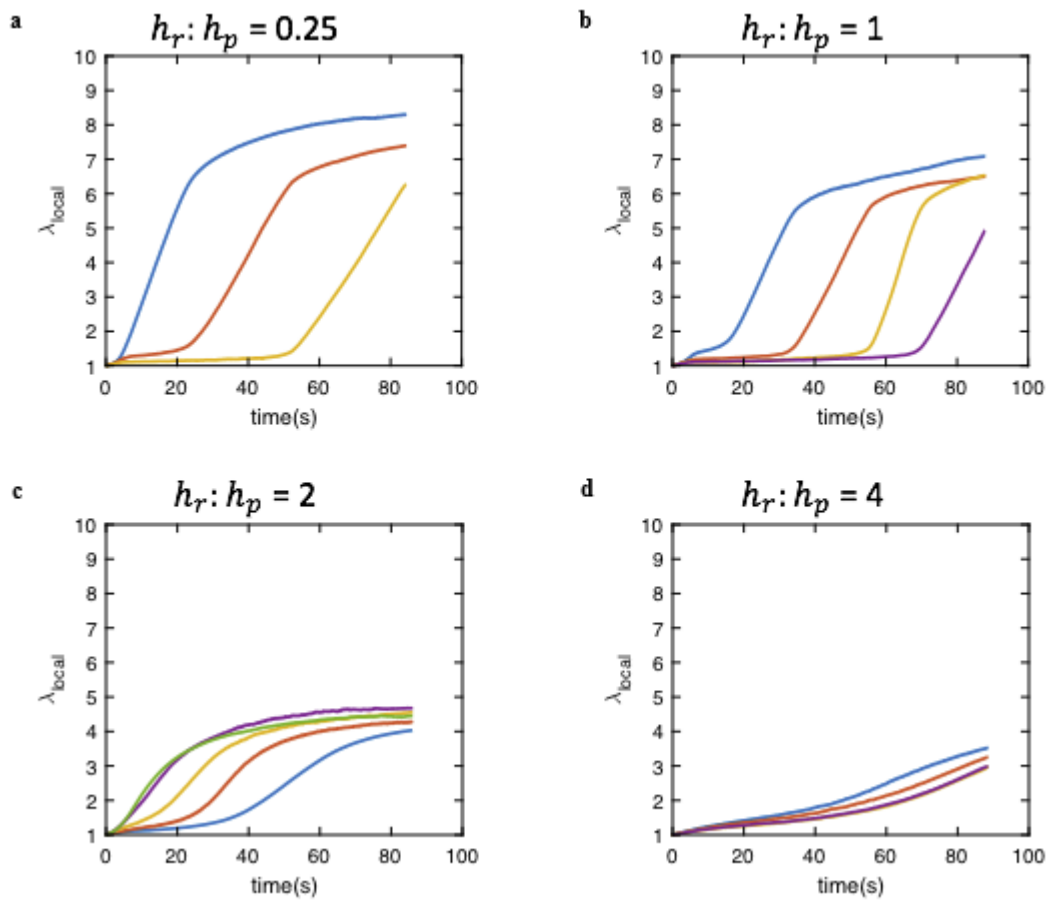


Figure 22 (a-d) The local stretches vs. time of different nominal ratio.

We choose three or four markers on the sample which showed well-developed necking. Based on each neighboring pair of markers, the local stretch between them could be estimated as a function of time. Each curve in Fig.22(a-d) corresponds to a pair of markers. Firstly, by observing the different curves of the same nominal ratio, we find that all curves tend to plateau to a well-defined stretch value after 80 s, this phenomenon represents the drawing had turned into a stable drawing. Correspondingly, there is a value of this plateau, we call the average value of those lines as plateau stretch. This value can be regarded as the natural draw ratio of this specimen, defined as the steady state stretch at which the neck stabilizes for a cold drawing plastic[11]. In our case, however, not every pair of markers shows a plateau, simply because that pair of markers escaped from the field of view before the material between them was fully drawn. This problem is not severe in nominal ratio 2 and 4 sample due to its necking propagation is faster than nominal ratio 0.25 or 1 specimen. Therefore, we only use the lines where its plateau clearly appeared. We plotted those values in Fig.23. There is a clear trend that the natural draw ratio decreases as nominal ratio increase.

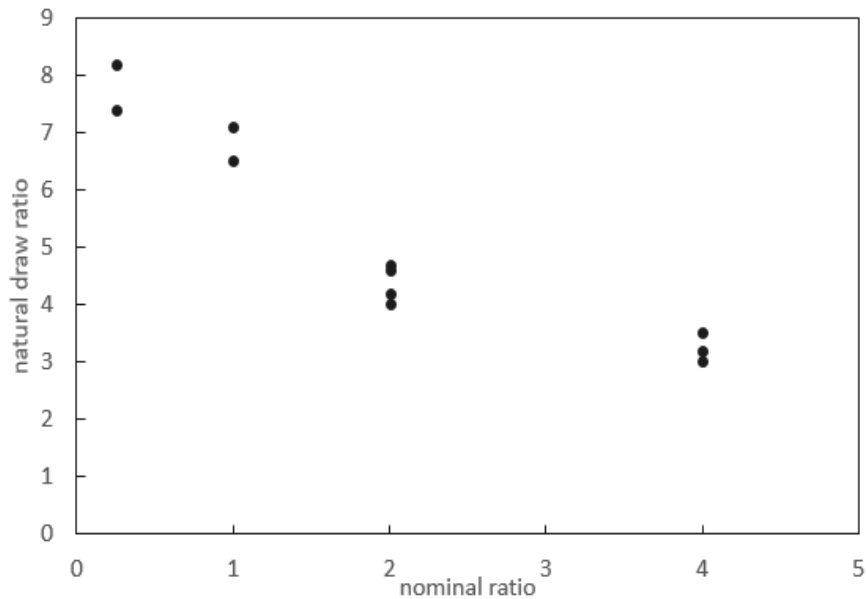


Figure 23 natural draw ratio of different nominal ratio.

From the result of maximum stretch of trilayer, we believe that the non-homogeneity of deformation reduces as rubber thickness increases. SEPS rubber has the best homogeneity during the deformation. For extreme case, $h_r/h_p=0$, pure extruded HDPE, due to its brittleness, it is difficult to find the maximum local stretch. However, Fig.24c plots one curve of extruded HDPE, which was obtained by using a different shaped HDPE. This sample has a shorter gauge length (10mm) than the others (Fig.24a). With this shape, we were able to get stable drawing behavior in our test(Fig.24b). So this result might not be entirely reliable, but from this figure we observed that the HDPE has the highest natural draw ratio (roughly higher than 12), this result is in agreement with the trend in Fig.23. So we believe that this figure supports our conclusion of deformation process at some degree.

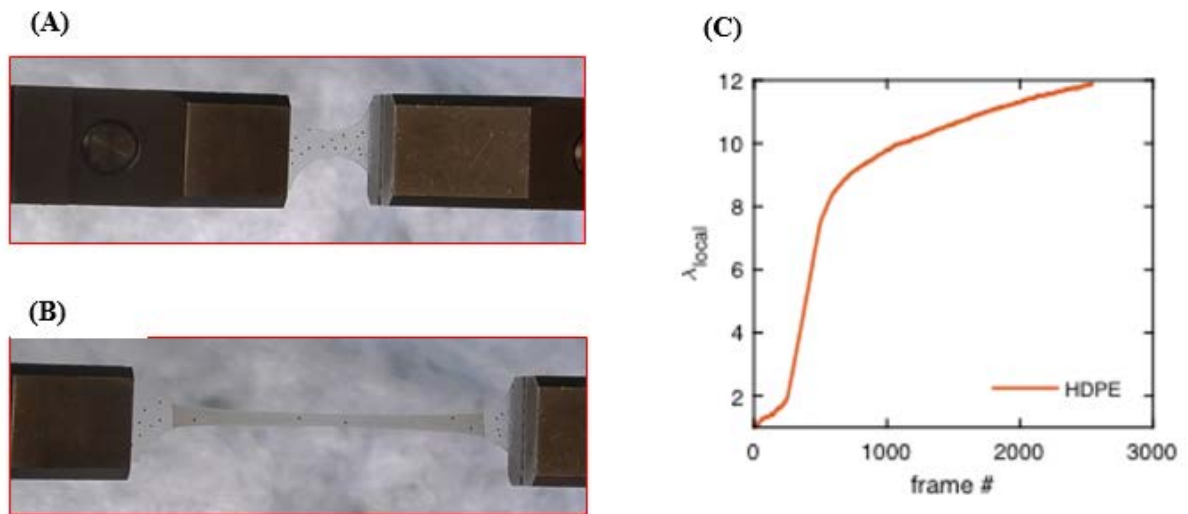


Figure 24 (a) The short gauge length sample. (b) The drawing image of the extruded HDPE specimen. (c)The local stretch vs. frame change of extruded HDPE.

5.0 SUMMARY

This thesis is centered on the tensile behavior of HDPE/SEPS bilayer laminate films and HDPE/SEPS trilayer laminate films. Before that, we examined the brittleness of pure HDPE and showed the dogbone shaped HDPE samples were somewhat brittle at low strain rate (less than 40mm/min), whereas the sample invariably were brittle and failed at a small nominal strain at high speed (greater than 40mm/min), at the same time, the yield stress of specimen increases with increasing strain rate.

For bilayer part, our experiment proved that by bonding a rubber layer on the plastic layer, the brittleness of HDPE was greatly reduced, and the plastic layer can be stretched to a higher deformation degree without failure. Even in the 900mm/min, the fastest strain rate of our machine, the composite specimens still showed a stable drawing. The bilayer has an intermediate mechanical behavior between the HDPE and the SEPS. Besides that, according to our experiment result, the bilayer behavior was weakly dependent on plastic: rubber ratio while the composites were more depending on strain rate, the draw stress and the peak stress value in high strain rate condition is always higher than in low speed condition. Correspondingly, the energy dissipation of composites has the similar trend. There is a higher value of energy absorption in the high strain rate condition during the tensile process.

For trilayer part, we find a similar conclusion that the rubber layer can reduce the brittleness of HDPE and prevent the plastic layer from breaking at low strain in the tensile

process, even in the smallest rubber:plastic ratio (nominal ratio 0.25). Moreover, in the same strain rate condition, the experiment result suggests that the draw stress and the peak stress decrease with increasing rubber thickness ratio and most of the force needed to stretch the trilayer comes from the plastic; the main role of the rubber is to prevent the failure of the plastic. The data generated from DIC suggests that the maximum stretch decreases as rubber thickness ratio increases, correspondingly, the pure plastic has the greatest maximum stretch whereas the SEPS rubber does not show drawing. The result also proved that by adding rubber thickness into the composites, the non-homogeneity of deformation reduces as rubber thickness increases. This trend is similar to the LLDPE/SEPS composites which result have been published from our lab.

There are still few works need to be done in the further study. Firstly, although our different nominal ratio samples show a clear trend in deformation of behavior, we can see there is no data of in the range of rubber:plastic ratio from 1 to 3, by examining more experiment and inserting more points into the figures, the trend from the figures will become more unambiguous and our conclusion will be more solid. Second is the issue of sample-to-sample thickness variability. This was relatively poor in our compression-molded samples. But even for the extruded films, even when we only cut sample along the machine direction, there are still some the differences of sample thickness. There is no doubt that our results will be more precise if the variability can be eliminated. Furthermore, in the study process, we noticed that the sample performance is greatly dependent on the sample quality, usually a small damage or notch on the sample could cause the specimen failure directly. Therefore, it is necessary to study the notched sample and figure out the notch's depth and shape play what roles in the specimen deformation process.

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