Essential work of fracture of glass bead filled low density polyethylene

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Recently, there has been an increasing interest in introducing high modulus inorganic rigid particles into polymer matrices because of the cost-effective character and the ability to reinforce the matrix, different from reduction in tensile strength and modulus caused by the compliant rubber particles. There are also examples of the toughening effect of these particles such as glass bead, $CaCO_3$ etc. filled into polymers such as high density polyethylene (HDPE) [1, 2], polypropylene (PP) [3] and so on in the literature. In these cases, the toughness of the composites was generally evaluated by means of impact tests.

When studying the failure of ductile materials, where a large plastic zone at the crack tip develops, and the energy dissipation is no longer confined to a small local zone near the crack tip, the J-integral approach has been used traditionally. Recently, the essential work of fracture (EWF) method, first developed by Broberg [4], has gained popularity due to the experimental simplicity and the intensive works of Karger-Kocsis [5–10], Mai [11–14] and other researchers [15, 16], and has been used extensively to study the fracture behavior of a wide range of polymeric materials.

The EWF concept of Broberg proposed that the non-elastic region at the tip of the crack may be divided into an inner fracture process zone (IFPZ) and an outer plastic deformation zone (OPDZ), as shown in Fig. 1. The total work of fracture, W_f, is then partitioned into two terms: the essential work of fracture W_e expended in the IFPZ to form a neck and subsequent tearing, and the nonessential work of fracture W_p dissipated in the OPDZ where various types of deformation such as shear yielding and microvoiding may be functioning. So,

$$W_f = W_e + W_p \tag{1}$$

and physically, W_e and W_p are expressed as:

$$W_e = w_e BL \tag{2}$$

and

$$W_P = w_P \beta BL^2 \tag{3}$$

Here, L is the ligament length, B is the specimen thickness, w_e is termed the specific essential work of fracture. It was claimed by Karger-Kocsis [17], that the specific essential work of fracture does represent a true material parameter when the EWF method is applied for suitable polymers. w_p is termed the specific non-essential work of fracture. β is a dimensionless factor associated with the shape of the plastic zone.

Introducing Equations 2 and 3 into Equation 1 gives:

$$w_f = \frac{W_f}{LB} = w_e + \beta w_p L \tag{4}$$

where w_f is the specific total work of fracture. For a given thickness, w_e is regarded as a material constant and provided the term βw_p remains independent of the ligament length, a linear relationship is expected between w_f and L. The positive intercept of this line with w_f-axis gives w_e and its slope gives βw_p.

The EWF analysis is based on some important assumptions and restrictions on sample dimensions. These assumptions and restrictions were analyzed in many literatures as mentioned above and will not be presented here.

It is has also been shown in several studies [15, 16, 18–20] that the parameter w_e may be estimated reasonably well via crack opening displacement (COD) of the advancing crack tip using a simple relationship of the form:

$$w_e = M\sigma_y \mathrm{COD} \tag{5}$$

where M is plastic constraint factor and σ_y is the yield strength of the material. To obtain COD, the extension to break values, e_f, were plotted against ligament length L. A linear dependence exists between the two parameters, i.e.,

$$e_f = e_o + e_p L \tag{6}$$

The intercept value e_o has been identified as the COD of the advancing crack tip, and e_p is the slope of the fitted line.

Although the main objective of the EWF method is to determine w_e, it is nevertheless useful and a worthwhile exercise to consider partitioning of w_e. Literatures [9, 21] have shown that when full-yielding of the ligament region coincides with the maximum load on the load–displacement curve, W_f may be partitioned into two components. But this was not considered in the present paper.

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0022-2461
DOI: 10.1007/s10853-005-4398-x

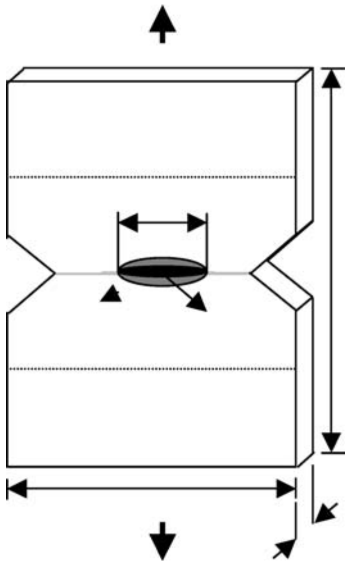

Figure 1 DENT sample used in EWF test.

Mouzakis and Karger-Kocsis [10] have studied the fracture behavior of hybrid composites consisting of isotactic polypropylene (PP), thermoplastic styrenic elastomer and glass beads by means of EWF theory, but the effect of bead size, and glass bead content on the fracture behavior of glass bead filled polymers by means of EWF test was rarely found in the literature. In the present work, glass beads of different particle size were filled into low density polyethylene, and the fracture behavior of the composites was evaluated by the EWF approach. And in this work, when recording the load-displacement curves, the elongation to break was recorded at the same time for calculating the COD.

The LDPE used was a granular material 18D produced by DaQing Petro., with the Melt Flow Rate (190, 2.16 kg) as 1.28 g/10 min. The glass beads were hollow beads produced by TianXingJian LTD, Yibing (PRC), with the trade mark Cenospheres. The average diameters of the untreated glass beads used were 0.71, 1.23, and 2.15 μm respectively.

After simple mixing, the LDPE and the glass beads of different bead size with predetermined proportion (mass percentage) were put into a TSSL-25 co-rotating twin extruder of L/D = 23/1 and D=25 mm made by Chengguang Chemical Institute, Chengdu, PRC, to blend in the molten state of LDPE, with a temperature profile in the range of 175 to 220 °C, and then the extrudate was pelletized. After drying to remove the attached moisture during extrusion and pelletizing, the pellets were injection-molded into specimens as shown in Fig. 1, on a PS40E5ASE precise injection molding machine, with a temperature profile of 180, 190, 215, and 220 from the feeding zone to the nozzle, and the injection pressure and the holding pressure were both about 65 MPa.

The EWF specimens, of varying ligaments were prepared from injection-molded parts. Initial notches were made perpendicularly to the tensile direction with a fresh razor blade, obtaining at least 17 specimens with ligament lengths varying between 2 and 15 mm for each set. The ligament lengths and the thickness were measured before the test using a reading microscope and a vernier caliper.

Static tensile tests on DDENT specimens were performed on an Instron electrical universal testing machine series IX at 25 °C and the crosshead speed was 5 mm/min. Tests with specimens of the pure LDPE and glass bead filled LDPE composites were carried out. The load-displacement curves, and the elongation to break at the same time, were recorded, and the absorbed energy until failure was calculated by computer integration of the load-displacement curves.

The load displacement curves (not shown) during DENT tests on the samples as a function of ligament lengths were similar in shape. For pure LDPE and LDPE composites filled with glass beads of different average diameter and different content, there is a linear elastic region in the initial stage. On further loading, two plastic zones were generated at the tips of both cracks, the size of which increased on further loading. The load eventually reached a maximum value, and the two plastic zones continue to propagate. After the peak, a sudden drop in load occurred, and the yielded ligament began to form a neck. As is known, the neck stabilization ability of LDPE is quite poor, so the load-displacement curves drop quickly after the yield point. In this period, the specimen fractured quickly.

The specific essential work of fracture, w_e, was obtained from the linear extrapolation of specific total work of fracture (w_f) against the ligament length. The slope of the w_f vs. ligament length l plot gave the nonessential work of fracture or plastic work, βw_p. The effect of dimensions (thickness, width, and gauge length), geometry, and test rate on the w_e and βw_p values was not considered in this study. Fig. 2 shows the plot w_f, of samples filled with glass beads of different average diameter, against the ligament length l and the specific essential work of fracture, and the plastic work results are summarized in Table I. From the values listed in Table I, it is clear that the specific plane stress fracture toughness is lower for the composites than that of pure LDPE, indicating that the crack resistance of the composites was lower with the addition

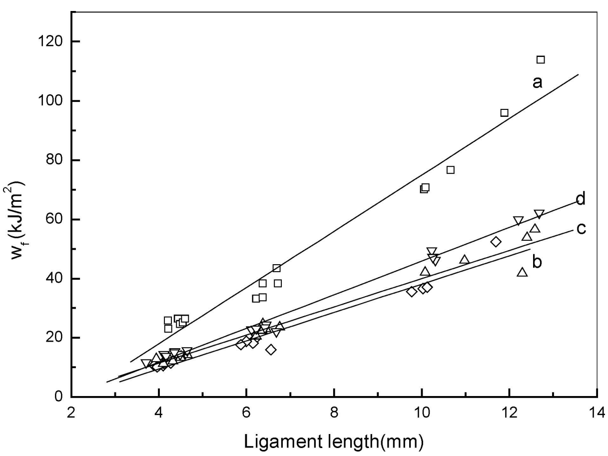

Figure 2 Total specific energy to fracture, w_f, of samples filled with GBs of different average diameter, against the ligament size, l. (□) curve a, Pure LDPE; (◯), curve b, LDPE + 15%GB with an average diameter of 2.15 μm; (□), curve c, LDPE + 15%GB of 1.23 μm; (□), curve d, LDPE + 15%GB of 0.71 μm.

TABLE I Fracture parameters obtained from EWF test for pure LDPE and the composites of LDPE and glass bead of different average diameter

LDPE composites	w_e (kJ/m^2)	βw_p (MJ/m^3)	R^2
Pure LDPE	31.8	2.55	0.940
85%LDPE + 15% 2.15 μm GB	15.2	1.48	0.926
85%LDPE + 15% 1.23 μm GB	18.7	1.33	0.914
85%LDPE + 15% 0.71 μm GB	19.5	1.55	0.961

of glass bead, which may be due to retarded molecular mobility in the presence of glass beads of the matrix. The plastic work, which is the energy dissipated in the process zone, also decreased with the addition of glass beads, indicating that less energy is absorbed during the fracture process for the composites. This may also be attributed to the retarded molecular mobility of LDPE molecules and the quick fracture of the composites.

On the other hand, the obtained values of w_e show an increasing trend as the average diameter of the glass bead becomes smaller while the βw_p do not show a clear changing trend for the filled materials, which indicates that average diameter of glass bead does influence the specific essential work of fracture significantly, while its influence on the βw_p is still not clear now.

The evaluation of essential work of fracture by means of COD was also adopted in this work. To obtain COD, the extension to break values, e_f, were plotted against ligament length shown in Fig. 3, and a linear dependence exists between the two parameters as shown in Equation 6.

The intercept value e_o has been identified as the COD of the advancing crack tip and the slope, *e*p, as the plastic contribution to extension; these parameters were included in Table II.

Here, the COD of the advancing crack tip, e_o, is larger for the composites than that of pure LDPE while the plastic contribution to extension, *e*p, decreased with the addition of glass beads. For the filled composites, e_o shows a significant increase as the average diameter of glass bead decreases to a certain value (which can not be obtained here) and then level off; e_p, on the other hand, don't show much difference for the filled composites.

To evaluate the effect of glass bead content on the essential work of fracture of glass beads filled LDPE, composites with the glass bead concentration of 5, 10, 15, 20 and 40% by weight were prepared. The plot of w_f vs. l of these samples was recorded in Fig. 4, and the essential work of fracture results is summarized in Table III. It can be seen that w_e of the composites also shows a great decrease with the addition of glass bead. For the composites filled with different content of glass bead, w_e shows an increase first, reaching a peak at 15 wt%, and then decreases, which indicates that the addition of glass bead with proper average diameter and content into LDPE matrix, presents a kind of toughening effect. On further increasing the content

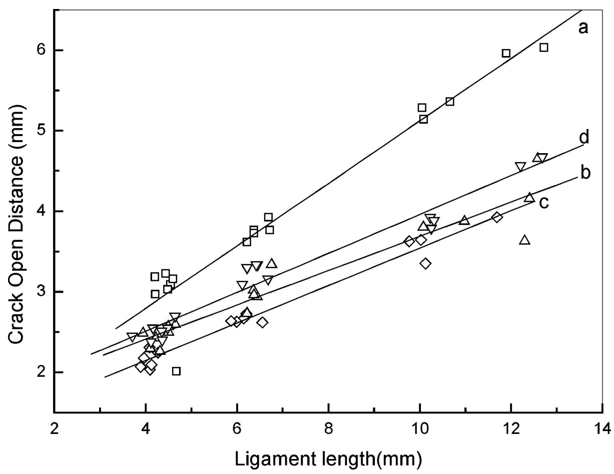

Figure 3 Crack open distance, *COD*, of samples filled with GBs of different average diameter, against the ligament size, *l*. (□) curve a, Pure LDPE; (○), curve b, LDPE + 15%GB of 2.15 μm; (□), curve c, LDPE + 15%GB of 1.23 μm; (□), curve d, LDPE + 15%GB of 0.71 μm.

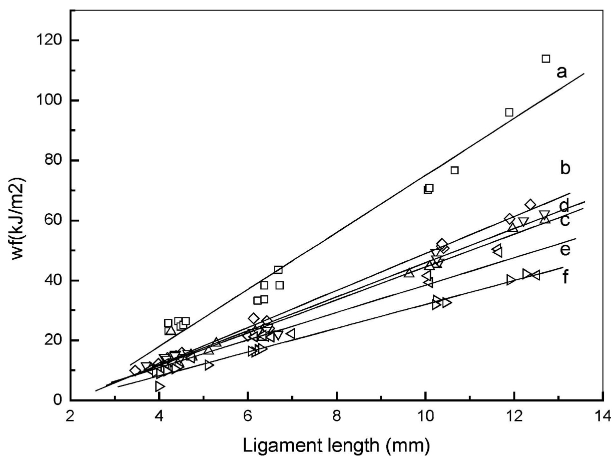

Figure 4 Total specific energy to fracture, w_f, of samples filled with GB of different content, against the ligament size, *l*. The average diameter of glass bead used here is 1.23 μm. (□) curve a, Pure LDPE; (○), curve b, LDPE + 5%GB; (□), curve c, LDPE + 10%GB; (□), curve d, LDPE + 15%GB; (◁), curve e, LDPE + 20%GB; (▷), curve f, LDPE + 40%GB.

TABLE II Fracture parameters of the extension to break of samples obtained from EWF test for pure LDPE and the composites of LDPE and glass bead of different average diameter

LDPE composites	e_o	e_p	R^2
Pure LDPE	1.24	0.39	0.968
85%LDPE + 15% 2.15 μm GB	1.21	0.23	0.986
85%LDPE + 15% 1.23 μm GB	1.56	0.21	0.956
85%LDPE + 15% 0.71 μm GB	1.54	0.24	0.979

TABLE III Fracture parameters obtained from EWF test for pure LDPE and the composites of LDPE and glass bead (0.71 μm) with different filling content

Filling content (wt%)	w_e (kJ/m^2)	βw_p (MJ/m^3)	R^2
0	31.8	2.55	0.940
5	17.3	2.01	0.964
10	17.4	1.79	0.966
15	19.5	1.55	0.961
20	17.3	1.52	0.967
40	14.8	0.942	0.953

TABLE IV Fracture parameters of the extension to break of samples obtained from EWF test for pure LDPE and the composites of LDPE and glass bead (0.71 μm) with different filling content

Filling content (wt%)	e_o	e_p	R^2
0	1.24	0.388	0.968
5	1.37	0.284	0.991
10	1.54	0.237	0.975
15	1.54	0.242	0.979
20	1.52	0.215	0.977
40	1.15	0.169	0.969

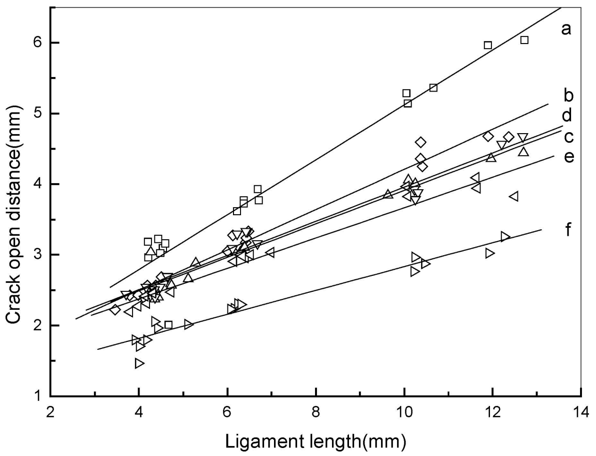

Figure 5 Crack open distance, *COD*, of samples filled with GB of different content, against the ligament size, *l*. The average diameter of glass bead used here is 1.23 μm. (□) curve a, Pure LDPE; (○), curve b, LDPE + 5%GB; (□), curve c, LDPE + 10%GB; (□), curve d, LDPE + 15%GB; (◁), curve e, LDPE + 20%GB; (▷), curve f, LDPE + 40%GB.

of glass bead, the retardance of the LDPE molecular mobility is greater, and w_e decreases correspondingly. But for βw_p, the composites show a roughly linear decreasing with increasing of the filling content of glass beads.

Also, the essential work of fracture by means of COD was evaluated, and the results were included in Fig. 5 and Table IV. In this case, the e_o values for the composites were all larger than that of pure LDPE except for 40 wt% glass bead filled composites, different from the changing trend of w_e shown in Table III, while the e_p values were all smaller than that of pure LDPE, similar to the changing trend of βw_p shown in Table III.

In conclusion, it is found that w_e and βw_p are lower for the composites than that of pure LDPE, and for the filled composites, w_e is increasing with the decreasing of the average diameter of the glass bead while the βw_p do not show a clear changing trend. For the composites filled with different content of glass bead, w_e shows an increase first, after reaching a peak and then decreases, presenting a kind of toughening effect. On further increasing the content of glass bead, w_e decreases. But for βw_p, the composites show a roughly linear decrease with increasing of the filling content of glass beads. By means of COD, the obtained e_o is larger for the filled composites than that of pure LDPE while *e*p, decreases with the addition of glass beads. For the filled composites, e_o shows a significant increase as the average diameter of glass bead decreases to a certain value. For the composites filled with different content of glass bead, the e_o values were larger than that of pure LDPE, while the e_p values were all smaller than that of pure LDPE.

Acknowledgements
The authors gratefully acknowledge the financial support of National Natural Science Foundation of China (Grant No. 20044001 and No. 50373027), Doctoral Research Foundation granted by the National Ministry of Education, China (Grant No. 20020610006) and the financial support on the Fundamental of Application Research Project from Sinopec Corp

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Received 29 January 2004
and accepted 25 February 2005