Fracture Toughness of Interface of Polyethylene Modified in Bulk

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ABSTRACT: The fracture toughness of the interface, G_a , of the self-healed joints of poly(ethylene) (PE) was measured using the wedge method. Samples of PE modified by mixing with three additives (branched low-molecular weight PE, a graphite filler, and polypropylene oil) were investigated. The development of the strength of partially healed joints formed by several hours of contact at a welding temperature of 105°C can be represented in all cases by the linear dependence of the G_a parameter on the square root of time, in accordance with the diffusion mechanism of the interface formation. The presence of the additive in samples was found to enhance the fracture toughness of a joint for a given welding time. In the graphite composites, an induction period of welding was observed. In contrast, an instant nonzero strength occurred in joints of PE with PP oil samples. The results confirmed that the concept of the chain entanglement control of fracture toughness developed originally for the glassy polymers is well transferable to the semicrystalline PE. However, additional mechanisms due to the crystallization of PE upon cooling are also effective in the development of the joint strength. These mechanisms denoted as cocrystallization, transcrystallization, local crystallization, mechanical interlocking, etc., are substantially affected by the concentration of additives. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 74: 1009-1016, 1999

Key words: healing; polyethylene composites; adhesive energy; chain entanglement; weld lines

INTRODUCTION

Polymer–polymer interfaces are of practical importance in numerous industrial applications. An intimate contact of two pieces of solid polymer leads to a joint with certain levels of adhesive strength. In noncrosslinked elastomers the adhesion is referred to as tack. In glassy polymers heated above the glass transition temperature T_g or in semicrystalline polymers the development of strength of the joint is called healing or welding. In both cases, the wetting of molecular surfaces

first occurs followed by interdiffusion of chain segments across the wetted interface. A special example of welding occurs during processing of polymers by injection molding or extrusion, whenever separate flow fronts of material in the liquid state meet forming an area known as weld line or a knit line.

The adhesion between two nonreacting polymers is controlled by the entanglement between the two materials.¹⁻⁴ It is assumed that macromolecules cross the interface by the reptation process, i.e., the chain ends move first, and rest of chains are dragging behind. It is evident that the rate of diffusion is influenced by molecular weight of linear chains, temperature, and contact pressure. Molecular aspects of interdiffusion at a polymer-polymer interface are conveniently ex-

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pressed by simple scaling laws.² At short contact times when the chain entanglement is still small the strength of a joint is rather weak, but in the limiting case of long welding time the strength of the interface formed may reach the cohesive strength of the bulk material. The mutual solubility of two polymers enhances the number of entanglements across the interface and interface thickness; generally, the broad interface results in a strong adherence.

Although the glassy or semicrystalline polymers are welded at enhanced temperature the testing of strength of resulting joints is performed at room temperature. The available tests of the strength of polymer adhesion are essentially fracture tests. Fracture mechanics is a topic well developed for bulk polymer materials,^{3,4} and can be applied to polymer interfaces under the critical condition that the crack must propagate along the interface. In linear fracture mechanics the fracture toughness of material is expressed by G_a , the critical value of strain energy release rate. This quantity can be measured, for example, by asymmetric double cantilever beam test (wedge test).^{2,5-7} In this simple method the crack is formed by inserting a razor blade at the interface, and its length ahead of the blade is measured. From the data measured, the fracture toughness of the interface G_a can be calculated using the expressions derived from finite elasticity theory.

The mechanisms of the strength development at polymer–polymer interface have been largely studied for glassy polymers only. The same factors should be relevant for fracture toughness of semicrystalline polymers, but in addition, influence of the crystalline micro- and macrostructure in the bulk and especially near the interface have to be considered.^{7–9} The role of crystallites in adhesion becomes even more pronounced in the case of semicrystalline polymers containing additives. An additive, depending on its nature, may behave as a nucleating agent or as a plasticizer.

In the present study we have examined the effects of concentration of various additives (X) on fracture toughness of the adhesive joints formed by poly(ethylene) (PE). We have studied the symmetrical interfaces formed between two plates of the same polymer either at PE self-healing (PE/PE) or at healing of PE modified in bulk by an additive (PE+X/PE+X). The basic material parameter, fracture toughness G_a , was determined by the wedge method for partially healed joints formed by several hours of welding at an elevated temperature. The time dependence of the joint

strength and its variation on the welding temperature and method of specimen preparation was evaluated. It was found that the increase in the concentration of additives leads to a growth of the joint strength of PE due to a combination of mechanisms of chain interpenetration and (co)crystallization close to interface and crystallization in the bulk.

EXPERIMENTAL

Commercial-grade low-density polyethylene (PE), (Bralen RA 2-19, Slovnaft Bratislava, Slovakia) with a density of 916 kg/m³ and a melt flow index of 2.0 g/10 min (210°C, 21.6 N), a melting temperature of 112°C, and a crystalline portion of about 52% (DSC) was used in the experiments. In composite sheets of PE modified in bulk, three different types of additives were used with the concentration up to 30 wt %: (1) a special grade of branched low-molecular weight polyethylene (LPE, SA 308-22, Slovnaft Bratislava, Slovakia), with a melt flow index of 305 g/10 min and a density of 919 kg/m³; (2) synthetic graphite (G) (EG-10, SGL-Carbon, UK), mean particle size of 31 μ m, surface area of 0.28 m²/cm³; and (3) PP oil (oPP) (Z 800, Slovnaft Bratislava, Slovakia) with a viscosity of 4000 mPa \cdot s corresponding to a molecular weight of 765–820. These three types of additives serve to model: (1) a mixture of polyethylenes differing in chain length and architecture, (2) a composite of PE with a particulate filler, and (3) a PE plasticized by a viscous liquid plasticiser.

All specimens of PE without or with additives were prepared by mixing in a 50-mL mixing chamber of a Brabender PlastiCorder PLE 331 (Brabender, Germany) for 8 min at 35 rpm at a temperature of $T_{\rm mix} = 130$ °C. The mixed material was compression molded at 120°C into 2 mmthick sheets from which the strips with dimensions $15 \times 50 \times 2$ mm were cut. Two strips in sandwich arrangement were subsequently welded in the press SRA-100 (Fontijne, The Netherlands) under light pressure to achieve good contact between two plates. Thin aluminium sheets were placed between the hot press and a sandwich specimen. For PE/PE specimens the welding temperature T_w was chosen in the range of 100-109°C, whereas for PE+X/PE+X specimens only the temperature of $T_w = 105 \pm 0.5$ °C was used. After the desired period of welding time (several hours) the specimens were cooled between metal

Sample	E [MPa]	$k = dG_a/dt^{1/2}$ [J · m ⁻² /h ^{1/2}]	$(G_a)_{t=5}^{\mathbf{a}} [\mathbf{J} \cdot \mathbf{m}^{-2}]$	T_m [K]
PE (unmixed)	234.1	3.22 ± 0.25	51.4	384.0
PE (mixed)	112.7	0.62 ± 0.05	12.3	388.5
PE-LPE (10%)	149.8	2.05 ± 0.27	40.5	387.0
PE-LPE (20%)	129.5	2.65 ± 0.23	54.7	384.0
PE-oPP (5%)	200.0	1.41 ± 0.26	33.1	_
PE-oPP (10%)	162.3	1.60 ± 0.43	45.8	_
PE-oPP (15%)	149.0	1.91 ± 0.27	56.7	_
PE-G (9.1%)	140.7	4.93 ± 0.29	62.8	_
PE-G (33.3%)	306.5	6.96 ± 0.71	87.9	_

Table I Young's Modulus and Interface Adhesion Parameters of PE Samples Modified by Mixing (See Text) and by Additives (wt %, Low Molecular Weight PE—LPE, Polypropylene Oil—oPP, Graphite—G) Welded at 105°C

^a At welding time of 5 h.

plates, cut into smaller stripes of dimensions of 5 \times 50 \times 2 mm, and mounted by a glue on a rigid support (glass, wood).

The test samples were stored at room temperature for at least 24 h prior to fracture. The fracture toughness of each sample was measured by an asymmetric double cantilever beam test (wedge test).¹⁻⁸ A blade was inserted at the interface and pushed into the sample. The crack length between the tip of the blade and visually determined end of the crack was measured after 24 h to allow for the sample relaxation. In preliminary experiments we have found that after this period of time the crack attains on average about 98% of its equilibrium length.

In asymmetric arrangement of the wedge test only the upper plate is bent by the wedge insertion. For this case the fracture toughness G_a is calculated by the relation derived from the finite elasticity theory.^{2,5,6}

$$G_a = (3Ed^3b^2) / \{8a^4(1 + 0.64d/a)^4\}$$
(1)

where E is Young's modulus, d is the thickness of the upper plate, b is the thickness of the blade, and a is the crack length.

A blade of the thickness of 0.42 mm was used in the experiments. The G_a values were not significantly affected by using the blades of thickness in the range of 0.2–1.2 mm. Each experimental point was determined by an average of at least five values of the fracture energy. In general, the reliability of the wedge method is good for small and intermediate values⁶ of G_a up to about 100 J/m². Higher values of G_a correspond to a very short crack lengths. Because this quantity enters into eq. (1) in its fourth power, any error in its magnitude results in a large error in G_a values.

Young's moduli E of the materials were determined for dumbell-shape specimens by a universal tensile tester Instron TM 4301 at a deformation rate 5 mm/min. The DSC method was used for determination of the melting temperatures measured by a Perkin-Elmer DSC II apparatus in a dynamic mode at a heating rate 10 K/min.

RESULTS

Young's Modulus

Fracture toughness of an interface is controlled by the local mechanisms around the crack tip and by the mechanical properties of the bulk material represented in eq. (1) through Young's modulus *E*. The tensile moduli *E* determined from the initial slope of the stress-strain curves are given in Table I for virgin PE and composite samples. Table I also includes the data for PE prior to mixing. It is seen that kneading in the mixing chamber results in a reduction of the modulus of PE by about 50%, evidently due to the mechanochemical degradation of PE. In two mixtures, PE+LPE and PE+oPP, the addition of a low amount of the second component causes a substantial increase of the Young's modulus relative to PE, because the additives apparently reduce the shear at mixing and protect the chain against scisson. A further increase in the concentration of the additive, a mechanically "soft" element, results in the re-

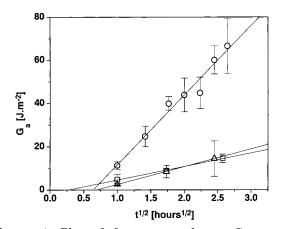


Figure 1 Plot of fracture toughness G_a versus square-root of time for unmixed PE (\bigcirc), and PE mixed at 130°C (\square) and 140°C (\triangle).

duction of the overall modulus of the mixture. In composite sheets of PE with graphite, the considerable reinforcing effect of the filler on the modulus dominates over a possible modulus reduction due to the mechanochemical degradation.

Variations in G_a with the Sample Preparation

The vast majority of energy associated with G_a is consumed by necessary plastic or viscous processes that occur around a crack tip. In the case of symmetrical interface of two identical PE materials it can be assumed that only the deformation by the "opening mode I" is operative at the crack tip.⁵ A stable crack growth along the interface was observed for test specimens. However, the crack lengths were considerably affected by the history of the samples and the method of their preparation.

From various variables involved in sample preparation, the plastification of PE by mixing brought the largest effect on the values of fracture toughness. This observation is illustrated in Figure 1, which shows the time development of the strength of the joints of PE untreated by mixing and PE samples mixed at either 130 or 140°C. The mechanical mixing of PE before compression molding results in reduction of fracture toughness of the interface compared to the joint formed by the virgin PE. In other words, a modification of the bulk mechanical properties of PE (evident from the modulus E in Table I) strikingly affects the interface adhesion. On the other hand, the increase in the temperature of mixing from 130 to 140°C has a negligible effect on the strength development in the interface. Both mixing temperatures used are well above the melting point T_m of PE samples, which is about 111–115°C. Therefore, all samples containing additives were mixed at 130°C.

The time development of the strength of joints is shown in Figure 1 and subsequent figures by using the square-root of time as a variable. Such type of dependence is suggested by the experimental results available for welding of identical or compatible pairs of polymer sheets, and is rationalized² by the theoretical considerations on the formation of molecular entanglements in the interface by reptation diffusion. For very long welding times the fracture toughness of the interface should approach the limiting value of the fracture toughness of bulk PE.

Variations in G_a with the Welding Temperature

In semicrystalline polymers the welding temperature T_w is the primary factor affecting the energy of adhesion. For PE samples investigated T_w was always selected under the melting temperature T_m of the respective material as determined from the DSC curve. The development of a joint strength of PE plates welded at four temperatures in the range of 103–107°C is shown in Figure 2. A linear plots of G_a vs. $t^{1/2}$ are observed for all four temperatures, and their slopes $k = dG_a/dt^{1/2}$ steeply increase with the increase of welding temperature. The welding at 109°C for the time over 10 h yields the joints with appreciable strength over 100 J/m². However, welding at temperature close to T_m becomes inconvenient because of the difficulties in the handling of the

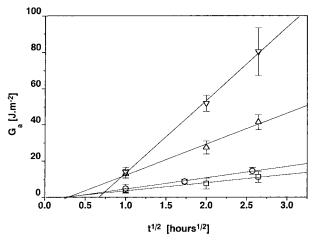


Figure 2 Plot of fracture toughness G_a versus square-root of time for PE welded at temperature $T_w = 103 \ (\Box), \ 105 \ (\odot), \ 107 \ (\triangle), \ and \ 109^{\circ}C \ (\bigtriangledown).$

160 140

G [J.m⁻²]

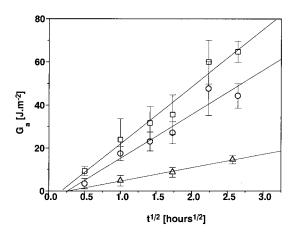


Figure 3 Plot of fracture toughness G_a versus square-root of time for PE mixture with LPE (wt %): 0 (\triangle), 10 (\bigcirc), and 20 (\square).

t1/2 [hours1/2]

Figure 4 Plot of fracture toughness G_a versus square-root of time for PE composite filled with graphite (wt %): 0 (\diamond), 9.1 (\triangle), and 33.3 (\bigtriangledown).

sandwich specimens and in the identification and localization of the interface between plates. Consequently, all joints formed from the modified PE were welded at $T_w = 105^{\circ}$ C.

Variations in G_a with the Additive Concentration

The toughness of bulk glassy or semicrystalline polymers is frequently enhanced by an addition of a suitable modifier. In our experiments we have focused on the related question of the influence of modifiers on the interface toughness. The first composite material studied, of the type PE+X, was a polymer mixture PE+LPE, where the standard low-density PE was modified by the addition of 10 or 20% (by weight) of the polyethylene with much lower molecular weight. The results in Figure 3 show a considerable increase in fracture toughness of the interfaces formed between plates of the PE+LPE in comparison to the PE/PE samples. This increase seems to be larger for the increment of the first 10% addition of LPE than for the second increment from 10 to 20%. The addition of LPE in mixture results in a slight reduction of T_m of the material (Table I).

Next, the adhesion properties of PE composites with a particulate filler were investigated. The development of the strength of adhesion as a function of $t^{1/2}$ for two concentrations of graphite in PE is shown in Figure 4. A particularly high fracture toughness was observed for the higher concentration of filler (33.3% wt). In this case, the G_a values of PE-G/PE-G joints welded for more than 7 h are by one order of magnitude larger than G_a values of unmodified PE. The proportionality of G_a to modulus E according to eq. (1) suggests that

considerable reinforcement of modulus E of composites (Table I) with increasing concentration of graphite (by almost 300%) can be a major reason of the observed enhancement of the interface fracture toughness. Interestingly, a long induction period in the development of adhesion between plates of graphite composites is observed (Fig. 4). Besides the decrease of the PE chain mobility, the limitations on heat transfer from the hot press to the sample may contribute to this phenomenon.

The third additive investigated, PP oil, served as an example of viscous liquid plasticizer. In analogy with the bulk toughness the interface toughness also increases with the concentration of PP oil in the composites (Fig. 5). In this system the cyclic phase separation of PP oil into the surface can be expected in several stages of the experiment. At first, the high-temperature mixing

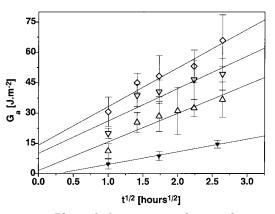


Figure 5 Plot of fracture toughness G_a versus square-root of time for PE plasticized with PP oil (wt %): 0 (∇), 5 (\triangle), 10 (∇), and 15 (\Diamond).

and compression molding of the PE+oPP mixture yields a homogeneous material. However, the cooling of the mixture to room temperature reduces the miscibility, and results in segregation of PP oil into the plate surface. In the next step, heating to the welding temperature should again bring the homogenization of the PE+oPP composite in test samples. The final cooling to ambient temperature for the fracture test presumably leads to the partial segregation of PP oil into the partially healed interface where it can act as a plasticizer. This description is supported by additional experiments where the PP oil segregated after cooling to the surface of the molded plates was removed by repeated wiping out by a cotton cloth. After welding of wiped samples their fracture toughness closely resemble that of the virgin PE.

The wetting of surface of PE by PP oil should be also responsible for the positive intercept on the ordinate axis in Figure 5, where a nonzero fracture toughness is found by the extrapolation to time t = 0. This observation can be attributed to the instant "wetting adhesion" on contact of two plates wetted by PP oil, before any chain interpenetration across the interface occurs.

DISCUSSION

The fracture toughness of the interface, expressed by the critical value of the strain energy release rate G_a represents the nonrecoverable energy expended in a unit increase in the crack length. The high values of G_a imply the materials with an efficient ability to stop the propagation of the cracks and, thus, to suppress the complete separation of crack surfaces at the failure. In the process of fracture of interfaces, two major mechanisms can be always identified on the molecular level: the deformation of coiled chains⁹ combined with the chain pullout, and the chain scission. The values of G_a obtained by the wedge method are closely related to quantities for joint strength, which provide the other tests of adhesion such as the various peel tests.^{5,10-12}

In our study of interfaces of PE modified in bulk we have focused on the partially healed joints. Thus, for example, the samples welded for 5 h give values of G_a in the range of $12-88 \text{ J} \cdot \text{m}^{-2}$ (Table I). Our results as well as other reports^{7,8} indicate that the wedge method can be employed in semicrystalline polymers for interfaces of the weak or intermediate strength. The complication due the mixed-mode effects stemming from the difference in the interface failure and bulk failure and ensuing tendency of cracks to grow away from the bond line into the lower crazing-stress material were eliminated by the use of identical polymers in the sandwich specimens.

The slopes $k = dG_a/dt^{1/2}$ of the straight lines in Figures 1–5 give the rate of the formation of the joints. From Table I it can be deduced how this rate is influenced by the kneading of PE or by the presence of additives. The observed straight-line plots of fracture toughness G_a vs. $t^{1/2}$ can be taken as a serious support for the diffusion nature of the welding in semicrystalline PE. In the reptation diffusion model the strength development² in PE joints is proportional to the number of bonds crossing the interface, and the latter quantity is proportional to the interfacial thickness. Evidently the length of molten PE chains is sufficient to secure chain entanglement and the formation of the interface bond. The critical molecular weight for PE entanglement can be estimated in good approximation from the characteristic ratio and monomer molecular weight M_{o} by the equation² $M_c = 30C_{\infty}M_o$, which, for PE, gives M_c = 5600. The more precise evaluation² shows some variation in M_c , depending on the effective chain cross-section and chain architecture. When the thickness of the interface attains approximately a coil size, the strength of joined material should approach the bulk strength. The bulk fracture energy of PE in the range of 1–10 kJ/m² was deduced,^{11,12} i.e., one or two orders of magnitude higher than the values of G_a of the partially healed interfaces we have measured. However, in the strength of bulk semicrystalline polymers the mechanism of local plastic yielding in crystalline domains is considered¹¹ to be the dominant factor.

The chain entanglement is a mechanism operative in the amorphous phase in both PE sheets at welding. The welding temperature was chosen close to the PE melting point to secure the destruction of a considerable part of crystallites. Remaining crystallites at T_w should not essentially affect the chain interdiffusion. However, in case of crystallisable interfaces such as PE, the interface strength can be controlled by an additional mechanism: on the cooling of the sandwich sample the molecules crystallize. The macromolecules can joint the growing crystals located in one sheet in the vicinity of interface and considerable disentanglement of chain segments from the melt interface can occur. At the same time molecules lying on either side of the interface can join the same crystallite,¹⁰ i.e., can cocrystallize. The crystallites that involve molecular strands from both sides of the interface are torn apart at the wedge test. It is interesting that the welding time seems to play a different role at these two mechanisms: the chain interpenetration results in a function G_a vs. $t^{1/2}$, while the crystallization is primarily affected by the mode and rate of cooling regardless of healing time. In incompatible semicrystalline interfaces such as polyethylene/polypropylene the related effect of "local crystallization" was proposed.¹³ The volume contraction due to crystallization of spherulites in the vicinity of the interface can result in breakup of the original interface plane with a subsequent influx of melts across the interface and formation of a mechanically interlocked interface. Evidently the growth of crystals near the interface can have positive and negative effects on the strength of interface.

Details of the crystalline micro- and macrostructure in the bulk and especially near the interface can control the measured values of fracture toughness. The degree of crystallinity and the crystal morphology in the bulk may differ substantially from the analogous features close to interface. By modifying the experimental procedure for making the joints, the crystal organization and, consequently the adhesive properties, can change. The role of crystallites in adhesion becomes even more pronounced in the case of semicrystalline polymers containing additives. Deciphering the mechanisms by which an additive affects the fracture toughness G_a is very difficult. Depending on the nature of the additive, it may act as nucleating or reinforcing agent or as a plasticizer and softener for a bulk material. It is known that under appropriate conditions a highly oriented layer, known as the transcrystallization layer is developed¹⁴ close to the surface, especially in fiber-reinforced semicrystalline polymers. This distinct morphology seems to be a consequence of a high nucleation ability of fibers, and affects the polymer/fiber interface strength.¹⁴ In our samples, the presence of an additive in all cases enhanced both the fracture toughness of a joint for a given welding time (Table I) and the rate of development of adhesion strength (the slope k). Only in the case of graphite is the direct proportionality found between the effects of the additive on G_a and on E, i.e., the increase in fracture toughness with concentration of the filler correlates with the reinforcement of the modulus.

In bulk polymers the influence of additives on fracture toughness was investigated mainly by the impact tests⁴ with the aim to increase the impact strength and the energy absorption. The related experiments focused on the interface toughness of polymers containing additives are rare. From the peel measurements data Novak¹⁵ determined the strength of adhesion of polypropylene (PP) foils modified by several low-molecular compounds (softeners) such as oleic acid. Their presence enhanced the strength of the adhesive joint. Interestingly, however, a maximum was observed on the dependence of the adhesive strength on the concentration of the additives in the sample.

In interfaces of PE of low or intermediate strength, a delicate control of the processing history is a necessary requirement for optimization of the adhesion at welding. This condition applies even more to the strong interfaces formed during the processing of this commodity thermoplastics. The internal weld lines,¹⁶ formed in the injectionmolded PE specimens whenever separate flow fronts of the material in the liquid state meet, represent a close analogy to the joints of our welded sandwich specimens. The presence of weld lines has been observed to reduce the mechanical properties of moldings of various polymers. Similarly, during the melt processing of polymer powders or pellets the interface weld lines occur. which completely heal provided a sufficient time for diffusion is allowed. The understanding of weld lines behavior is important for filled polymers and composites, and the rationalization of their strength should include all the factors related to crystallinity and additives discussed above for self-healed PE joints.

CONCLUSIONS

The fracture toughness of the interface G_a of the self-healed joints of poly(ethylene) (PE) was measured using the wedge method. The samples of PE modified by mixing of the three prototypes of additives were investigated. By the welding at the temperature close under the PE melting point the results show that the concepts derived for interface fracture toughness of glassy polymers are well transferable to the semicrystalline PE. In the partially healed joints of weak and intermediate strength, the presence of an additive in all cases enhanced the fracture toughness of a joint for a given welding time and the rate of development of

adhesion strength. The interface strength seems to be controlled by two mechanisms: the chain interpenetration represented by the function G_a vs. $t^{1/2}$, and (co)crystallization influenced primarily by the rate of cooling.

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