

# Effects of adhesive type and polystyrene concentration on the shear strength of bonded polystyrene/high-density polyethylene blends

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## Abstract

The lap-shear strengths of adhesively bonded polystyrene (PS), high-density polyethylene (HDPE), and their blends, were studied as a function of adhesive type and blend composition. The performance of virgin and recycled polymer systems was examined. The lap-shear strength depended strongly on the amount of PS in the blend and the type of adhesive, and the acrylic adhesives demonstrated the best performance for all compositions. Bonded strengths of HDPE increased by approximately 50% when HDPE was blended with 34% PS, the co-continuous composition. The results indicate that structural elements made from PS/HDPE immiscible blends may be effectively bonded with adhesives without expensive surface treatments.

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## 1. Introduction

Low surface energy is a well-known property of most polymers, particularly compared to metals and ceramics. As a general rule, the lower the surface energy of the substrate, the greater the difficulty to develop an adhesive bond. Typical surface energies of various materials are given in Table 1 [1–3]. Due to the low surface energy and chemical inertness of polyethylene, expensive surface treatments are generally performed to increase adhesive performance. All such treatments aim to form oxygen-containing functional groups at the surfaces, a state that can be achieved by exposure to ultraviolet radiation, plasma or corona discharge, or by flame or acid treatment [4,5]. Epoxy adhesives have also been used to bond polyethylene, but elaborate surface preparations are required that include surface etching with a sodium sulfuric-dichromate acid solution at an elevated temperature. Flame treatment, corona dis-

charge, and plasma treatment have also been effectively used. Due to its aromatic nature, polystyrene (PS) is sensitive to aromatic and chlorinated solvents, a feature that has led to the conventional process of solvent bonding. However, if the PS surface is sufficiently abraded and solvent cleaned, polyurethane and epoxy adhesives will also provide a good bond [6,7].

However, due to the increasing usage of immiscible co-continuous blends of PS with high-density polyethylene (HDPE) and/or polypropylene in structural applications, a commercial need exists for bonding these plastics with minimal surface preparation. In particular, work in our laboratory has identified regions in the PS/HDPE system where co-continuity and enhanced mechanical properties occur. Based on well-established rheological models, these co-continuous regions are usually near 35% PS for common commercial blends. These composites, which are stiff and durable, are an intimate micron-scale mixture of the two immiscible polymers.

Hence, in this study, we evaluated the lap-shear strength of commonly used adhesives on abraded but otherwise untreated surfaces of virgin and recycled

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PS/HDPE blends, including the co-continuous compositions and the neat end member polymers. An additional goal of this work was to examine the effect of PS concentrations in PS/HDPE blends. The premise guiding this part of the study is that the PS component of the blend will provide enhanced bonding of the composite, thus precluding the need for expensive adhesives or elaborate surface preparations.

## 2. Materials

Adhesives were selected to represent a variety of polymers from traditional and inexpensive to pricier, state-of-the-art, multi-part adhesives designed for polymers having low surface energies (Table 2). Substrates for virgin composition were prepared from pure polymer pellets of general purpose PS supplied by GE polymers under the name, Espree CPS7GP, and HDPE supplied by CP Chem under the name, Marlex HHM5202BN.

Recycled materials were also studied, due to the growing commercial production of railroad ties and bridge beams from blends of recycled PS/HDPE. The raw materials for these commercial structural elements are obtained from post-industrial scrap and are highly variable in composition. To obtain recycled materials for this study, samples representative of annual produc-

tion materials of both PS and HDPE were obtained from a major manufacturer of structural elements (Polywood incorporated, Edison, NJ). These materials were received as clean polymer flakes about 3–5 mm in size, which from visual inspection appeared to be shredded packaging materials of PS and HDPE. The PS flakes were slightly pigmented and contained some foil and paper impurities. The HDPE flakes were a mixture of colored and clear flakes originating mainly from blow-molded containers.

Blends were prepared by pellet blending of 20% and 34% virgin PS with HDPE and 20% and 34% recycled PS with recycled HDPE. Adhesives were applied to substrates of identical compositions. Five specimens were prepared and tested for each adhesive/polymer combination.

## 3. Experimental

ASTM Type-1 tensile bars were injection-molded with a 55 ton laboratory injection molding machine (Negri-Boss v55-200). The tensile bars were cut into two equal halves and the bonding surfaces were abraded with a 180-grit paper until no evidence of surface gloss was visible and then were wiped clean with a dry cloth. The bars were bonded in a lap-shear geometry. The bonded area of adhesion was nominally  $10 \times 13$  mm and pressure was applied to the lap joint during the curing cycle by two large binder clips. Prior to testing, the bonded specimens were cured at room temperature and ambient humidity for 72 h.

The lap-shear strength for each adhesive/substrate was determined on a computer-controlled tensile testing machine (MTS Q Test/25) with TestWorks software. During this test, shear stress was applied across the adhesive bond, and the bonded materials were forced to slide over each other with the adhesive bond layer providing the resistive force. Prior to each test the bonded area of each specimen was measured and recorded. The specimens were tested at a crosshead speed of 1.3 mm/min as per ASTM specifications and the peak load in Newtons was noted [8]. The lap-shear strength in megapascals was calculated as the measured peak load divided by the bonded area. The reported test values are the average of five measurements.

## 4. Results and discussion

The results of mean lap-shear strength measurements for joints composed of each adhesive and the various polymer substrates are shown in Table 3. The goal of the lap-shear test is to determine the level of stress needed to exceed the cohesive strength of the adhesive, the shear

Table 1  
Typical surface energies of polymers and metals [1,2]

Material	Surface energy (mJ/m <sup>2</sup> )
Teflon	20
Paraffin wax	26
Polyethylene	33
Polystyrene	42
Iron	1360
Gold	1500

Table 2  
Adhesives selected for the study

Type of adhesive	Details	Brand
Acrylic	Structural adhesive. Two-part proprietary adhesive formulation	3M's DP 8010
Polyurethane	Commercial	Nail Power
Solvent/elastomeric	Popular construction adhesive	Macco Liquid Nails
Epoxy	Two-part	LN 601 Loctite Epoxy

Table 3  
Shear strength values for various adhesives bonding PS/HDPE blends (values are averages of five replications)

Substrate	Adhesive							
	Acrylic		Polyurethane		Epoxy		Solvent/elastomer	
	Shear strength (MPa)	Failure mode	Shear strength (MPa)	Failure mode	Shear strength (MPa)	Failure mode	Shear strength (MPa)	Failure mode
HDPE 100%	1.34	TYPE D	0.46	TYPE D	0.70	TYPE D	0.62	TYPE D
Recycled HDPE100%	1.64	TYPE D	0.35	TYPE D	0.45	TYPE D	0.42	TYPE D
80%HDPE/ 20%PS	2.12	TYPE D	0.92	TYPE D	0.70	TYPE D	0.70	TYPE D
Recycled 80%HDPE/ 20%PS	1.52	TYPE D	0.55	TYPE D	0.45	TYPE D	0.68	TYPE D
66%HDPE/ 34%PS	1.96	TYPE D	0.96	TYPE D	0.89	TYPE D	0.77	TYPE D
Recycled 66%HDPE/ 34%PS	1.53	TYPE D	0.82	TYPE D	0.81	TYPE D	0.73	TYPE D
PS100%	4.76	TYPE C	2.39	TYPE D	1.51	TYPE D	1.24	TYPE D
Recycled PS100%	2.93	TYPE C	2.94	TYPE C	1.01	TYPE D	1.46	TYPE D

Note: Failure type refers to the illustrations in Fig. 1. An example of Type D failure is shown in Fig. 2.

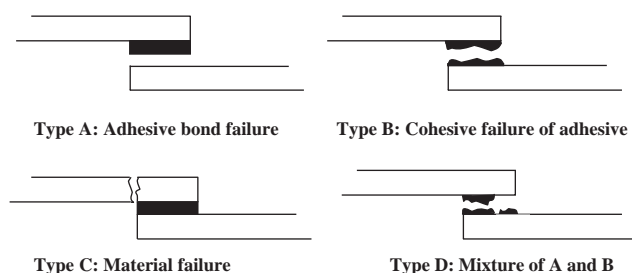


Fig. 1. Typical failure modes during lap-shear adhesion test.

strength of the substrate, or the adhesive strength of the bond [9]. Naturally, some combination of these mechanisms can occur as well. Typical failure modes during lap-shear tests are shown in Fig. 1 [10]. In the present study we observed four failure modes, the three aforementioned and a combination of adhesive and cohesive failure, as depicted in Fig. 1: Type D. The mixed adhesive and cohesive failure mode was observed quite often. The morphology of this mode consisted of small patches of cohesively failed adhesive bonded to the substrate and surrounded by areas of bare substrate where the interface between the adhesive and substrate had failed. Fig. 2 illustrates an example of this Type D failure.

The relationships between bond strength and virgin substrate composition for each adhesive are shown in Fig. 3. Virgin PS has higher bond shear strength

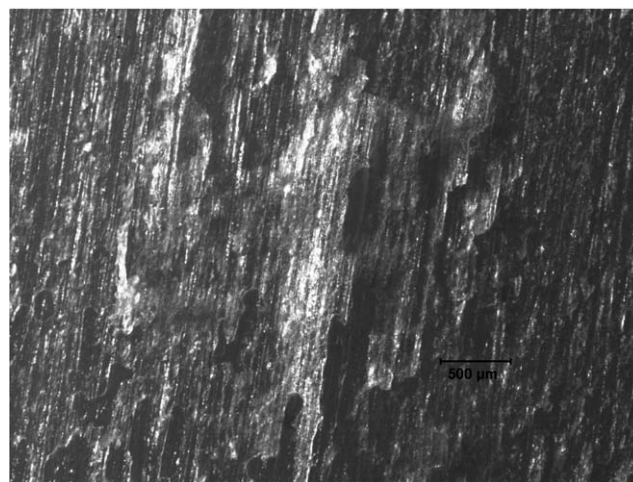


Fig. 2. Optical micrograph of polyurethane bond failure interface of 34% PS/HDPE virgin composite. Note evidence of adhesive and cohesive failure, Type D.

compared to virgin polyethylene for all adhesives tested and blends show approximately a rule of mixtures behavior. The structural acrylic adhesive was the superior adhesive on all surfaces. In fact, several of the PS substrates broke during test, suggesting that the actual bond shear strength was higher, perhaps much higher, than the value reported. The mode of substrate failure seems to result from an induced flexural moment.

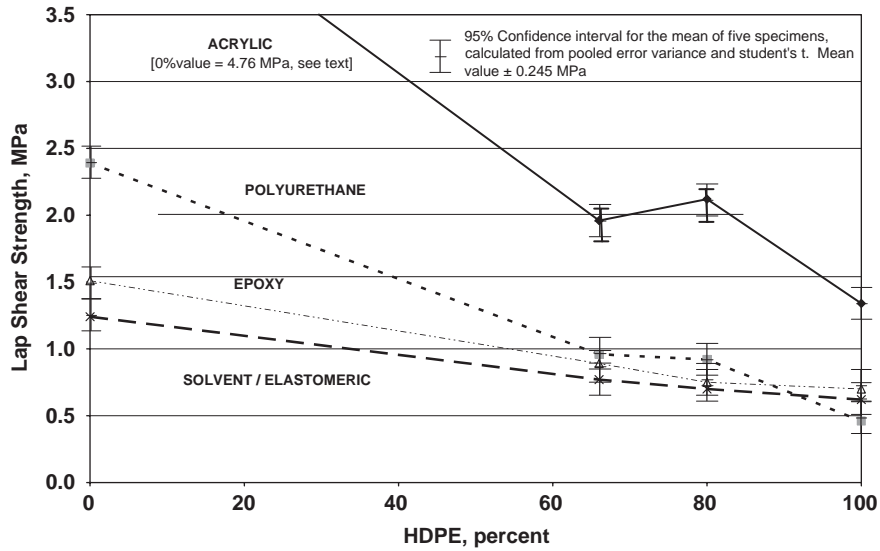


Fig. 3. Lap-shear strength in the virgin PS/HDPE system.

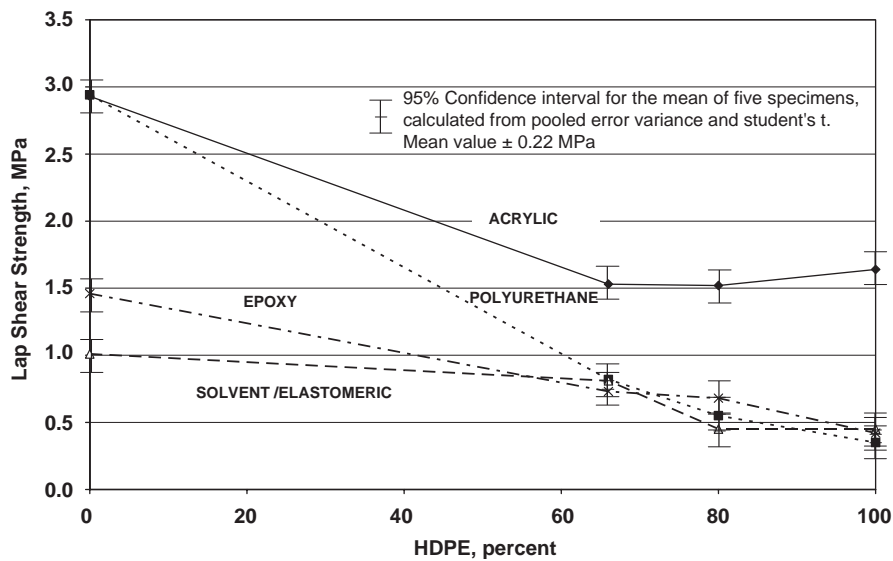


Fig. 4. Lap-shear strength in the recycled PS/HDPE system.

The influence of PS in the PS/HDPE blends in raising the bonding capability of these blends is also evident in Fig. 3. PS levels of ~30% increase the shear bond strengths of the joints by ~50% over virgin HDPE, although no significant differences ( $p > 0.05$ ) between 20% and 34% PS were measured. On a technical performance basis, the solvent/elastomer adhesive and epoxy demonstrate inferior adhesion as compared to acrylic. Although cost performance was not assessed in this study, there may be numerous applications in which shear strength of 0.7 MPa (100 psi) is satisfactory and the low-cost solvent/elastomer adhesive (Liquid Nails) will give greater cost performance than the higher performing adhesives.

The performance of adhesives on recycled PS and HDPE is shown in Fig. 4. These data show a trend similar to their virgin counterparts, although the polyurethane adhesive demonstrates greater bond strength with the recycled PS compared to the virgin material (2.9 versus 2.4 MPa) and the acrylic adhesive provided lower lap-shear strength values for recycled PS (2.9 versus 4.7 MPa). These exceptions to the trend appear to arise from a combination of effects. Impurities in the recycled PS may alter the surface energy and promote enhanced polyurethane bonding. Likewise, the recycled PS is mechanically weaker than the virgin material and numerous substrate failures occurred (see Table 3) which produced low values for both the

acrylic- and polyurethane-bonded PS substrates. Hence, the reported bond strengths between recycled PS and acrylic or polyurethane adhesives may be lower than would be observed if the substrates had not failed.

PS has an exceptional ability to bond to itself and to many common adhesives, in part due to the softening and plasticizing role of many solvents present in adhesives. Such performance makes PS an attractive additive to otherwise difficult-to-bond systems, such as polyethylene. Generally only solvent cleaning and abrasion are necessary for surface preparation of PS [7]. This characteristic of PS played an important role in improving the bonding capability of polyethylene, an otherwise inert polymer. Although elucidation of the bonding mechanisms of these adhesives is well beyond the scope of this paper, the general theory of adhesive bonding via interdiffusion seems relevant in these systems. As the glass transition temperature ( $T_g$ ) of PS, or any other material, is reduced below ambient temperature by solvents, the bonding of such materials with other materials that are also above  $T_g$ , e.g. HDPE, is enhanced [11,12]. In addition to these physical/chemical effects, the roughness of the surface is also important. Brittle thermoplastics like PS can produce rough surfaces through brittle fracture [13,14]. In the present investigation, the topography of the roughened test specimen surfaces was examined under an optical microscope. Micrographs of these surfaces revealed a high level of surface roughness on all specimens. These surface imperfections serve as locations where the adhesive can enter and mechanically bond with the adherend [9] thus increasing the lap-shear strength.

## 5. Summary and conclusions

Lap-shear tests of adhesively bonded virgin PS and HDPE and their blends have shown that bond strengths increase with increasing PS content for all four adhesives studied: acrylic, polyurethane, epoxy, and a solvent/elastomer system. Compared to neat polyethylene, the addition of ~30% PS (the amount needed to form a co-continuous blend for the polymer system used) resulted

in an average increase in joint strength of ~50%. Of the adhesive types, the acrylic adhesive performed the best, followed by polyurethane, epoxy, and the elastomer system, although for the blend containing 34% PS, all of the non-acrylic adhesives performed similarly. Recycled materials behaved nearly identically to the virgin materials, except for a slight improvement in the polyurethane adhesive bonded to virgin PS.

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