



INFLUENCE OF HOT MELT ADHESIVE CONTAMINANTS ON THE MECHANICAL PROPERTIES OF RECOMPOUNDED THERMOPLASTICS

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The influence of hot melt adhesive (HMA) contaminants on the mechanical properties of thermoplastics has been studied with the aim of exploring the possibility of materials recycling of thermoplastic components containing HMA-residue. Samples of polypropylene (PP), ABS or high impact polystyrene (HIPS) containing 0-8 w% of PUR- or EVA-HMA were prepared and tested. The tensile and impact properties of the thermoplastics could be significantly affected at high contents (8 w%) of PUR- or EVA-HMA. However, at relevant contents of HMA-contamination (1-2 w%) the influence was negligible. It was shown by differential scanning calorimetry that the melt enthalpy of PP was not significantly affected by contents of up to 8 w% of PUR- or EVA-HMA. Nor was the glass transition temperature, T_g , of the PS and ANS-phase in HIPS and ABS respectively affected by contents of up to 8 w% of PUR- or EVA-HMA, thus indicating that the PUR- or EVA-residue was present as a separate phase in the thermoplastics. This could also be confirmed for the PUR-residue in PP from scanning electron microscopy (SEM) studies on samples etched in sulphuric acid. By SEM analysis it was also shown that the tensile and impact fractures were initiated at a particle of 50-100 μm size and varying shape, with no correlation to the type and concentration of HMA contaminant present in the thermoplastic. It is concluded that recycling of thermoplastic components contaminated with HMA-residue can be performed without removal of the residue.

Abbreviated title: Influence of hot melt adhesive contaminants on the mechanical properties of recompounded thermoplastics

Key words: Hot melt adhesive (HMA) ,Contaminants, Recycling

INTRODUCTION

Recycling of thermo-plastic components of a product bonded together with an adhesive is usually difficult since few separation techniques are available and since the adhesive residues will contaminate the plastic regrind. For these reasons adhesive bonding is not recommended in design of plastic products for recycling. Adhesive joints are generally considered as non-separable and are often replaced with other separable joints like snap-fits or screws^{1,2}. However, hot melt adhesive (HMA) joints are separable and show many advantages compared to traditional separable joints such as better stress distribution, faster assembly and less weight.

HMA is defined as solvent-free adhesive applied in molten state and solidifying on cooling. There are two types of HMAs, thermoplastic and reactive (crosslinked)^{3,4}. The most frequently used thermoplastic HMAs are based on ethylene-vinyl acetate copolymers. Thermoplastic types are mostly used for packaging, book-binding, non-wovens or wood-products. Reactive HMAs have higher strength and higher maximum using-temperature and are used for example in construction, flooring and automotive industry. A reactive HMA is a polyurethane (PUR), forming a semi-interpenetrating network polymer, lightly crosslinking in contact with moisture. At higher temperatures reactive types soften and thermoplastic types melt. This makes HMA-joints separable on heating in combination with mechanical action.

Thermo-mechanical methods for separation of hot melt adhesive (HMA) joints have been developed^{5,6}. Thus, the adhesive layer may be heated up to a temperature where the strength of the adhesive has decreased sufficiently to make mechanical separation possible. The heat may be generated outside and transferred to the adhesive for instance using a conventional oven. The heat may also be generated inside the adhesive using resistive heating or microwave heating. As model materials various thermo-plastics such as ABS, PP and high impact polystyrenes (HIPS) have been used in combination with different moisture curing PUR-HMAs. Adhesive will be left on the separated components causing contamination if incompatible with matrix polymer of the thermoplastic on materials recycling of the components. These contaminants may act as defects causing stress concentration and reducing the mechanical strength of the material. The stress concentrating effect will depend mainly on the size of the contaminant particles. Few publications have addressed the issue of compatibility of adhesive with thermoplastic regrind^{7,8}. Cleaning is time-consuming requiring special equipment and techniques. In this work, the possibility of incorporating PUR- or EVA-HMA-residue in ABS, high impact PS (HIPS) and PP thus avoiding the cleaning process is examined. In order to study the influence of HMA-contamination on the mechanical properties of thermoplastic regrind, virgin materials were compounded with ground HMA.

EXPERIMENTAL

Materials

Type	Grade	Supplier
ABS	Polylac F00	Polykemi AB
High impact PS (HIPS)	Polystrene 552	Polykemi AB
PP	ME210U	Borealis

Table 1. Thermoplastics

Type	Grade	Supplier	Crosslinked
Polyurethane HMA	Purfect 254	National Starch & Chemicals	Yes
Ethylene-vinyl acetate HMA	National 8686	National Starch & Chemicals	No

Table 2. Adhesives

Compounding

Compounding was carried out using a Brabender Plasticorder PL2000 twin-screw extruder. Recommended processing parameters from the suppliers were used. The polyurethane HMA was crosslinked and ground to a size of 0.5-1 mm before compounding. Samples of PP, ABS or HIPS containing 0-8 w% of PUR- or EVA-HMA were prepared, see Table 3.

Sample	Composition (w%)	Sample	Composition (w%)
ABS	100/0	HIPS/EVA-HMA	99/1
ABS/PUR-HMA	99/1	HIPS/EVA-HMA	98/2
ABS/PUR-HMA	98/2	HIPS/EVA-HMA	97/3
ABS/PUR-HMA	97/3	HIPS/EVA-HMA	96/4
ABS/PUR-HMA	96/4	HIPS/EVA-HMA	95/5
ABS/PUR-HMA	95/5	HIPS/EVA-HMA	92/8
ABS/PUR-HMA	92/8	PP	100/0
ABS/EVA-HMA	99/1	PP/PUR-HMA	99/1
ABS/EVA-HMA	98/2	PP/PUR-HMA	98/2
ABS/EVA-HMA	97/3	PP/PUR-HMA	97/3
ABS/EVA-HMA	96/4	PP/PUR-HMA	96/4
ABS/EVA-HMA	95/5	PP/PUR-HMA	95/5
ABS/EVA-HMA	92/8	PP/PUR-HMA	92/8
HIPS	100/0	PP/EVA-HMA	99/1
HIPS/PUR-HMA	99/1	PP/EVA-HMA	98/2
HIPS/PUR-HMA	98/2	PP/EVA-HMA	97/3
HIPS/PUR-HMA	97/3	PP/EVA-HMA	96/4
HIPS/PUR-HMA	96/4	PP/EVA-HMA	95/5
HIPS/PUR-HMA	95/5	PP/EVA-HMA	92/8
HIPS/PUR-HMA	92/8		

Table 3. ABS, HIPS and PP samples.

Injection moulding

A Battenfeld Plus 250/050 injection moulding machine was used to prepare tensile test specimens (type M-1, ASTM D 638 M) with a thickness of 3.15 mm for tensile and tensile impact testing, see Table 4.

Parameter	ABS	HIPS	PP
Injection speed (cm ³ /s)	16.5	16.5	38.5
Mold temperature (°C)	25-40	25-40	30
Cylinder temperature profile (°C)	220, 225	220, 225	230, 235
Holding pressure (MPa)	40	40	40
Holding pressure time (s)	5.5	5.5	20
Cooling time (s)	28-30	28-30	30

Table 4. Processing parameters

Tensile testing

An Instron 5566 tensile testing machine provided with an external video extensometer was used to determine the modulus, the yield strength and the strain at fracture. The measurements were performed using a cross-head speed of 5 mm/min for ABS and HIPS and 50 mm/min for PP in accordance with ASTM D 638 M.

Tensile impact testing

An Amsler RKP300 Charpy impact tester equipped with a tensile impact device was used for tensile impact testing. Tests of PP specimens were performed at a striking velocity of 2.07 m/s and an initial potential energy of 23.51 J. Tests of ABS and HIPS specimens were performed at a striking velocity of 2.7 m/s and an initial potential energy of 40.14 J.

Thermal analysis

A Mettler-Toledo 820 DSC was used for thermal analysis at a heating rate of 10 °C/min in nitrogen atmosphere.

Scanning electron microscopy

The fracture surfaces were sputtered with gold-palladium and studied using a JEOL JSM 5400 scanning electron microscope. Etching of PP/PUR-HMA-samples in conc. sulphuric acid was performed at 20°C for 7 h to identify particles of PUR-HMA.

RESULTS

The effect of HMA residue on the mechanical properties of recompounded PP, ABS and HIPS was studied using typical levels of HMA residue in a plastic regrind. These levels are usually below 1 w% and rarely more than 2 w%^{7,8,9}.

Tensile testing

Modulus. The modulus of PP, ABS and HIPS as a function of the amount of PUR- or EVA-HMA is shown in Figures 1-3. The moduli of PP and HIPS decrease with increasing PUR- or EVA-HMA content. Addition of 8 w% PUR-HMA to PP or HIPS lowers the modulus by approximately 40 %. The corresponding figure for EVA-HMA is 25%. However, the loss of modulus at 2 w% HMA is only 10 and 20 % respectively. The modulus of ABS is not affected by PUR- or EVA-HMA contents up to 8 w%. Due to the expected data scatter (average standard deviation $\pm 5-10$ %) any attempt to fit the data to theoretical modulus-content expressions is meaningless.

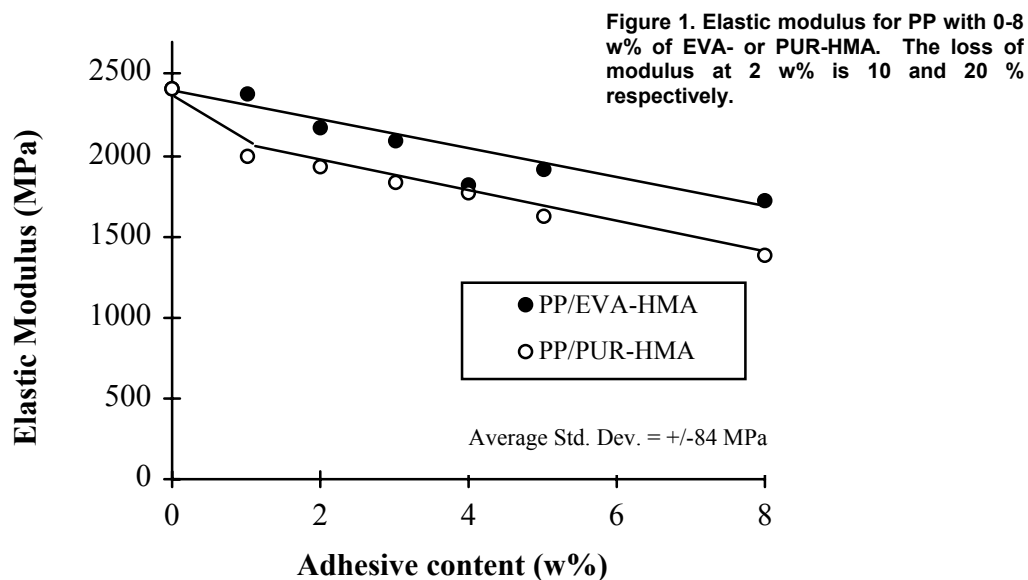


Figure 1. Elastic modulus for PP with 0-8 w% of EVA- or PUR-HMA. The loss of modulus at 2 w% is 10 and 20 % respectively.

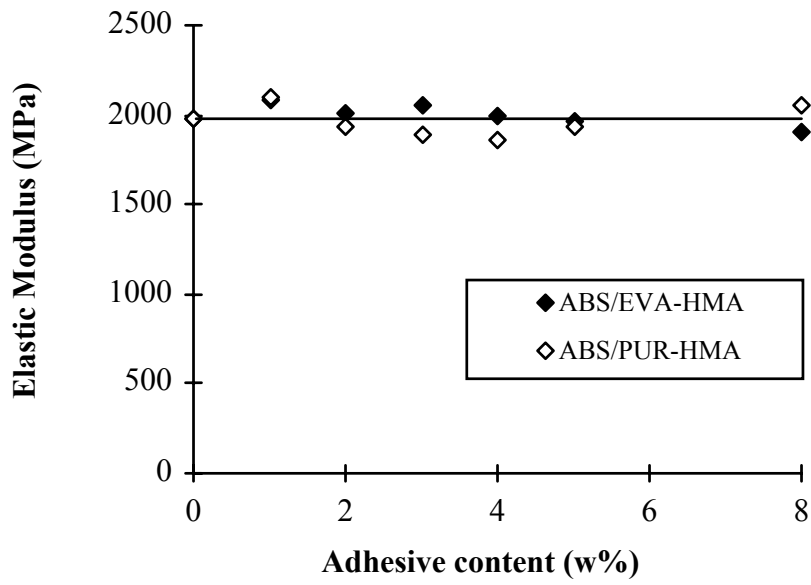


Figure 2. Elastic modulus for ABS with 0-8 w% of EVA- or PUR-HMA. No loss of modulus at 2 w% of HMA is observed.

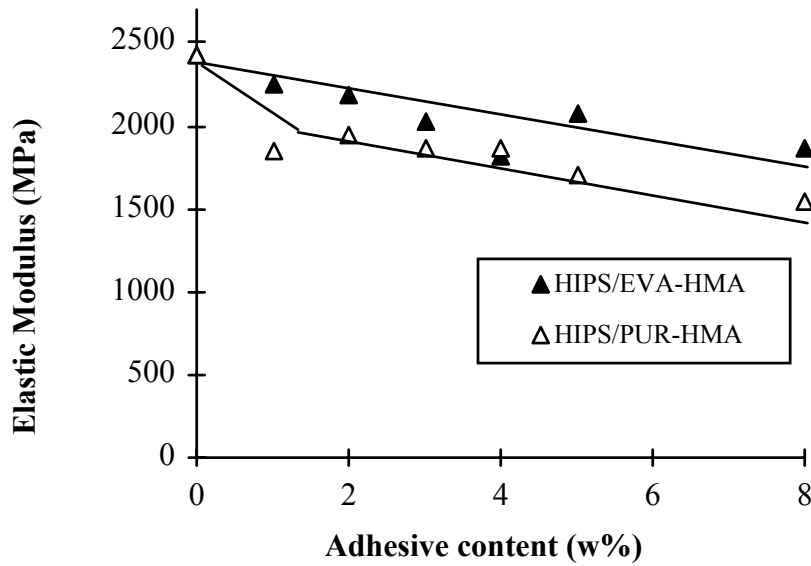


Figure 3. Elastic modulus for HIPS with 0-8 w% of EVA- or PUR-HMA. The loss of modulus at 2 w% HMA is 14 and 20 % respectively.

Yield strength. The yield strength of PP, ABS and HIPS as a function of the amount of PUR- or EVA-HMA is shown in Figures 4-6. The yield strength of PP is lowered by less than 20 % on addition of 8 w% HMA, whereas the yield strengths of ABS and HIPS are basically unaffected. Thus, the loss of yield strength at 2 w% HMA is very small for PP (5-8 %) and negligible for ABS and HIPS.

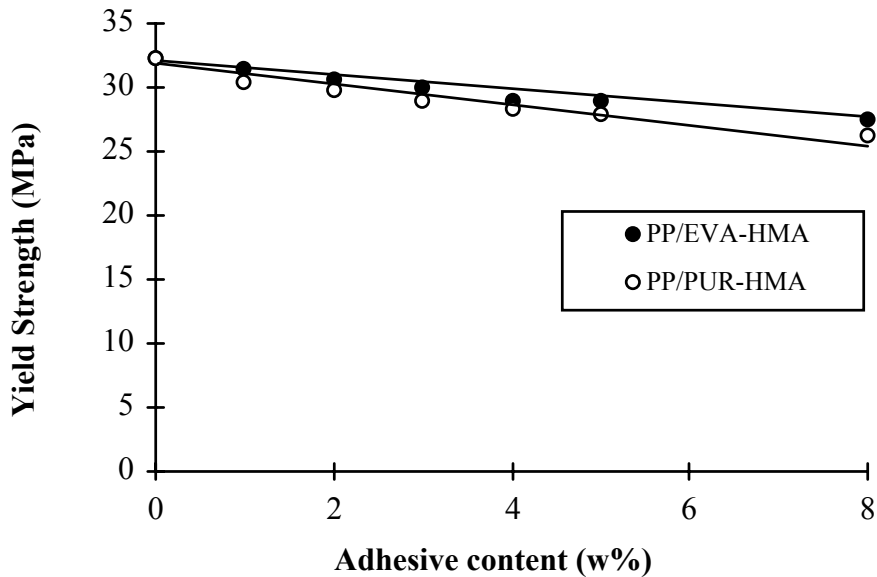


Figure 4. Yield strength for PP with 0-8 w% of EVA- or PUR-HMA. The loss of yield strength at 2 w% HMA is 5 and 8 % respectively.

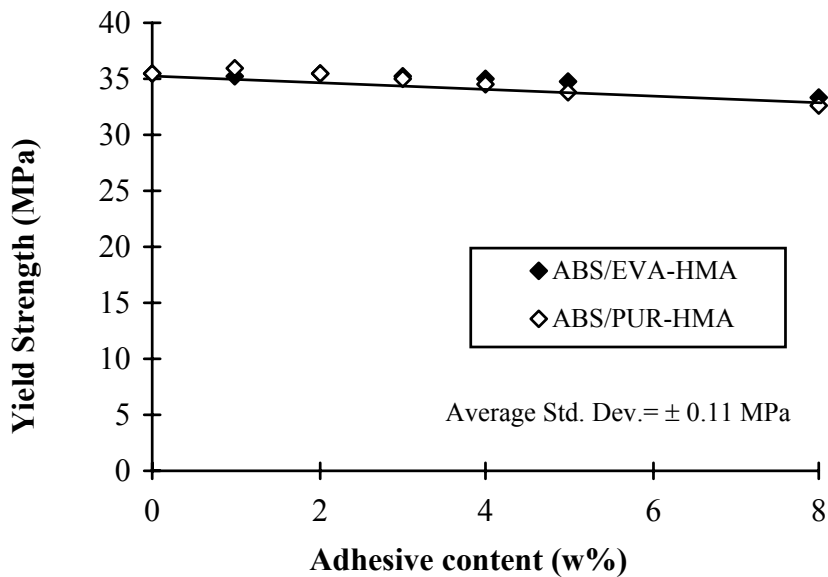


Figure 5. Yield strength for ABS with 0-8 w% of EVA- or PUR-HMA. No loss of yield strength at 2 w% HMA is observed.

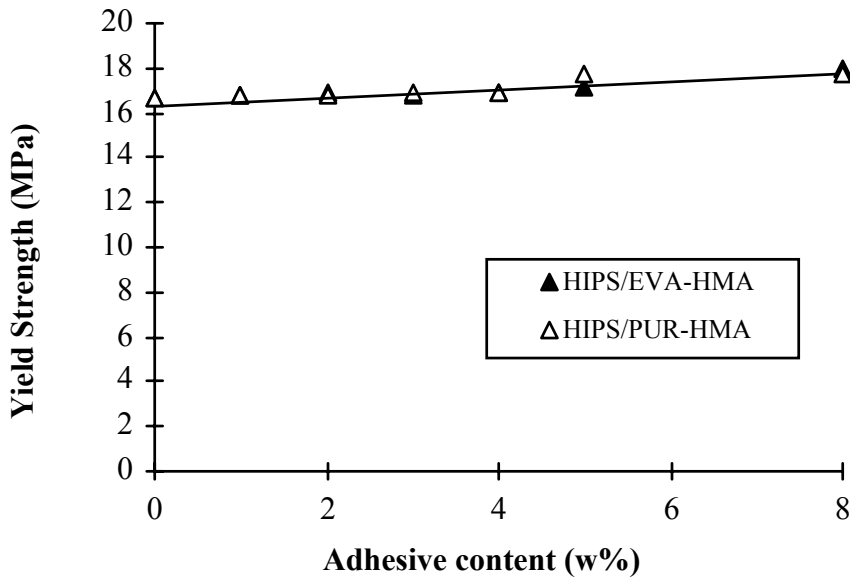


Figure 6. Yield strength for HIPS with 0-8 w% of EVA- or PUR-HMA. No loss of yield strength at 2 w% HMA is observed.

Strain at fracture. The strain at fracture of PP, ABS and HIPS as a function of the amount of PUR- or EVA-HMA is shown in Figures 7-9. On addition of 8 w% HMA the strain at fracture for PP is in fact increased by a factor of approximately two, for ABS it is lowered by 20-50 % and for HIPS it is lowered by 50 % (EVA-HMA) or unaffected (PUR-HMA). The loss of strain at fracture at 2 w% HMA for PP, ABS and HIPS is negligible with the exception of the small loss observed for ABS/EVA-HMA.

For details on tensile properties, see Table 5.

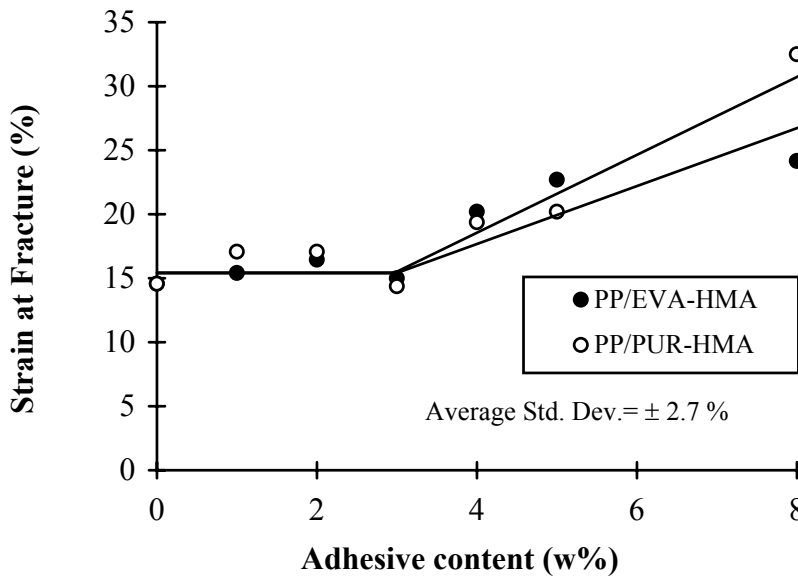


Figure 7. Strain at fracture for PP with 0-8 w% of EVA- or PUR-HMA. No loss of strain at fracture at 2 w% HMA is observed.

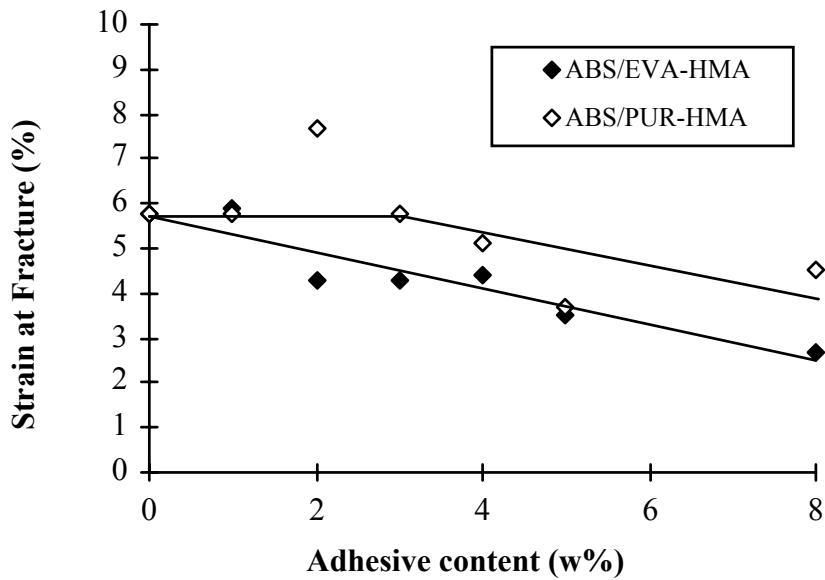


Figure 8. Strain at fracture for ABS with 0-8 w% of EVA- or PUR-HMA. The loss of strain at fracture at 2 w% HMA is 0 and 15 % respectively.

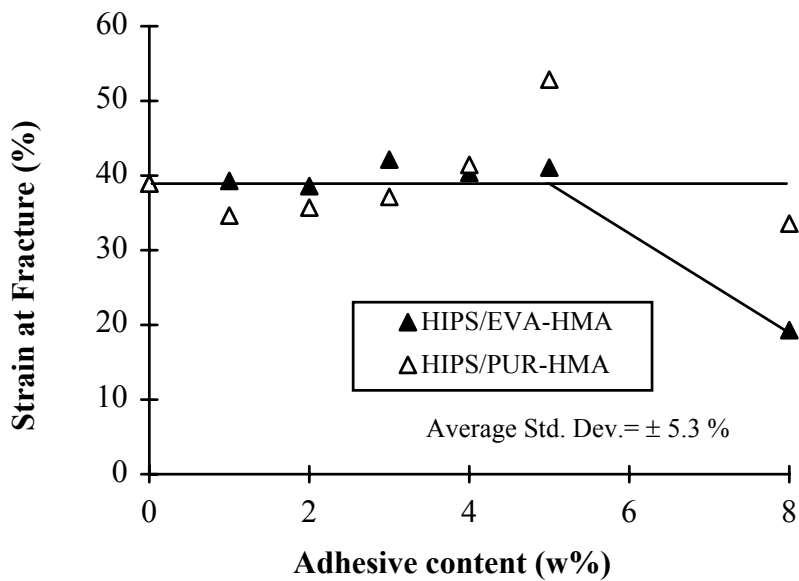


Figure 9. Strain at fracture for HIPS with 0-8 w% of EVA- or PUR-HMA. No loss of strain at fracture at 2 w% HMA is observed.

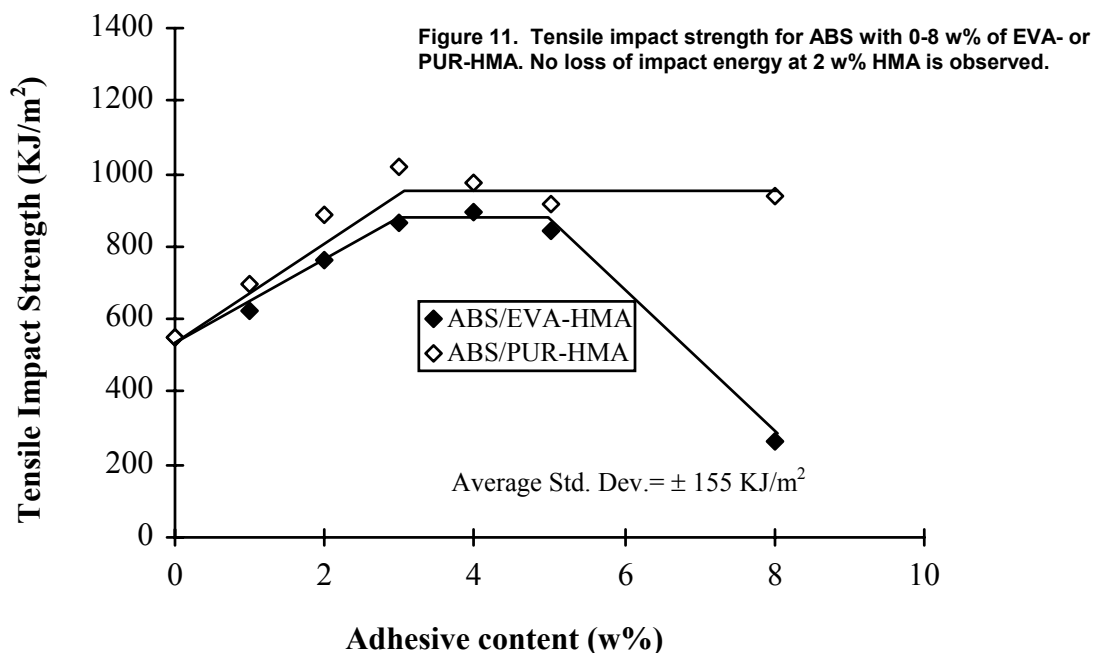
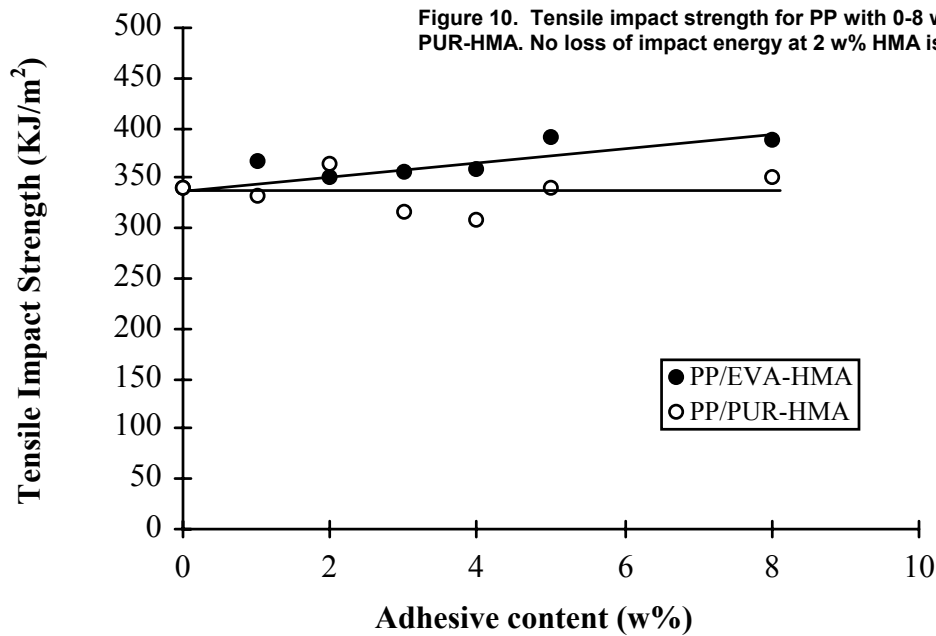
Sample	Modulus (MPa)	Yield strength (MPa)	Stress at fracture (MPa)	Strain at fracture (%)	Tensile impact strength (kJ/m ²)
PP virgin resin	2274	32.1	26.7	12.5	353.7
PP regrind [*]	2405	32.3	26.3	14.6	339.5
PP / 1 w% PUR-HMA	1997	30.4	22.9	17.1	331.4
PP / 2 w% PUR-HMA	1933	29.7	22.0	17.1	364.4
PP / 3 w% PUR-HMA	1839	28.9	20.9	14.4	316.9
PP / 4 w% PUR-HMA	1760	28.3	15.0	19.3	309.3
PP / 5 w% PUR-HMA	1627	27.8	12.3	20.2	341.5
PP / 8 w% PUR-HMA	1387	26.3	10.0	32.4	350.7
PP / 1 w% EVA-HMA	2379	31.4	25.5	15.4	366.1
PP / 2 w% EVA-HMA	2162	30.6	24.5	16.4	349.8
PP / 3 w% EVA-HMA	2094	29.9	24.2	15.1	356.6
PP / 4 w% EVA-HMA	1818	28.9	12.7	20.3	359.3
PP / 5 w% EVA-HMA	1913	28.9	15.0	22.8	390.1
PP / 8 w% EVA-HMA	1724	27.5	11.1	24.1	387.0
ABS virgin resin	1969	34.9	31.7	3.3	753.2
ABS regrind [*]	1985	35.5	32.3	5.8	556.4
ABS / 1 w% PUR-HMA	2091	36.0	32.3	5.8	705.6
ABS / 2 w% PUR-HMA	1936	35.4	30.6	7.7	906.3
ABS / 3 w% PUR-HMA	1890	35.1	29.0	5.8	1041.3
ABS / 4 w% PUR-HMA	1858	34.5	28.1	5.1	995.1
ABS / 5 w% PUR-HMA	1929	33.9	27.5	3.7	936.7
ABS / 8 w% PUR-HMA	2050	32.6	25.7	4.5	954.7
ABS / 1 w% EVA-HMA	2082	35.2	31.5	5.9	634.1
ABS / 2 w% EVA-HMA	2016	35.4	32.0	4.3	778.3
ABS / 3 w% EVA-HMA	2050	35.2	31.5	4.3	882.2
ABS / 4 w% EVA-HMA	1988	35.1	31.3	4.4	910.3
ABS / 5 w% EVA-HMA	1967	34.7	30.9	3.5	857.3
ABS / 8 w% EVA-HMA	1910	33.4	30.8	2.7	265.5
HIPS virgin resin	2169	17.6	19.8	39.3	751.1
HIPS regrind [*]	2431	16.7	18.8	38.9	717.5
HIPS / 1 w% PUR-HMA	1856	16.8	18.1	34.5	676.1
HIPS / 2 w% PUR-HMA	1942	16.8	18.4	35.7	524.9
HIPS / 3 w% PUR-HMA	1859	16.9	18.3	37.3	812.9
HIPS / 4 w% PUR-HMA	1863	16.9	18.4	41.4	667.0
HIPS / 5 w% PUR-HMA	1709	17.7	19.6	53.0	811.1
HIPS / 8 w% PUR-HMA	1543	17.7	18.1	33.4	767.3
HIPS / 1 w% EVA-HMA	2257	16.7	18.2	39.2	723.9
HIPS / 2 w% EVA-HMA	2193	16.9	17.9	38.4	685.4
HIPS / 3 w% EVA-HMA	2017	16.8	18.3	42.0	555.4
HIPS / 4 w% EVA-HMA	1819	16.9	18.2	40.3	780.6
HIPS / 5 w% EVA-HMA	2078	17.2	18.2	40.9	528.6
HIPS / 8 w% EVA-HMA	1858	18.0	16.1	19.2	361.8

*) Recompounded without addition of PUR- or EVA-HMA

Table 5. Tensile and tensile impact data for ABS, HIPS and PP with 0-8 w% of EVA- or PUR-HMA.

Tensile impact testing

The tensile impact strength of recompounded PP, ABS and HIPS as a function of the amount of PUR- or EVA-HMA is shown in Figures 10-12. On addition of 8 w% HMA the impact strength of PP is unaffected or even slightly increased (EVA-HMA) while the impact strength of HIPS is unaffected (PUR-HMA) or lowered by approximately 50 % (EVA-HMA). The impact strength of ABS increases on addition of HMA leveling out above 3 w% but decreasing by 50 % at 8 w% of EVA-HMA. No loss of impact strength is obtained at 2 w% HMA for PP, ABS or HIPS, in fact for ABS an increase is observed. As expected a considerable data scattering is noticed, particularly for ABS and HIPS (average standard deviations from 10 to 25 % were obtained). However, the general conclusions are not affected. For details on tensile impact strength values, see Table 5.



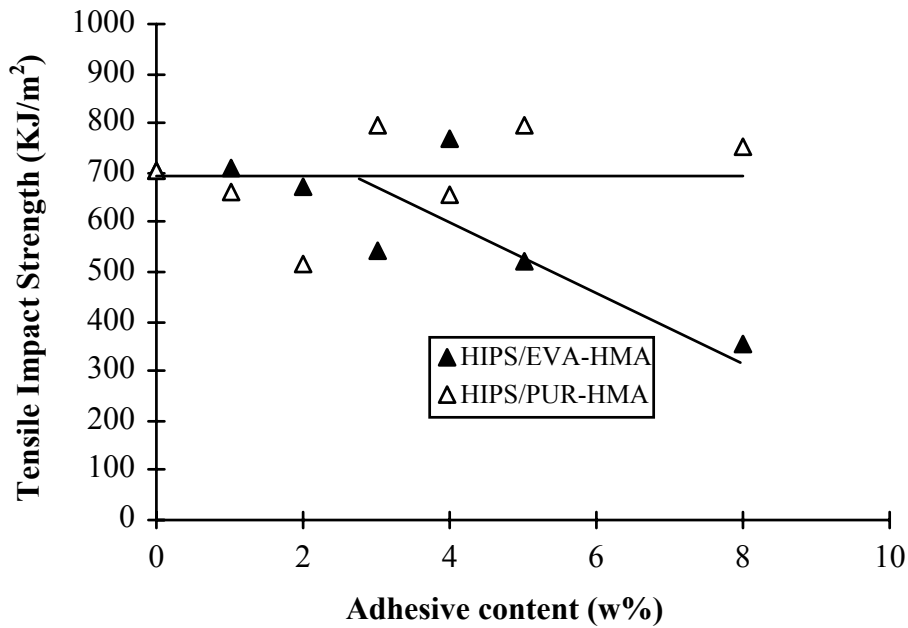


Figure 12. Tensile impact strength for HIPS with 0-8 w% of EVA- or PUR-HMA. No loss of impact energy at 2 w% HMA is observed.

Thermal analysis

PP. The melt enthalpy of PP containing PUR- or EVA-HMA was measured to decide if the presence of HMA affects the crystallization of PP. The results show that the melt enthalpy of PP is not significantly affected by the addition of up to 8 w% of PUR- or EVA-HMA, see Figure 13.

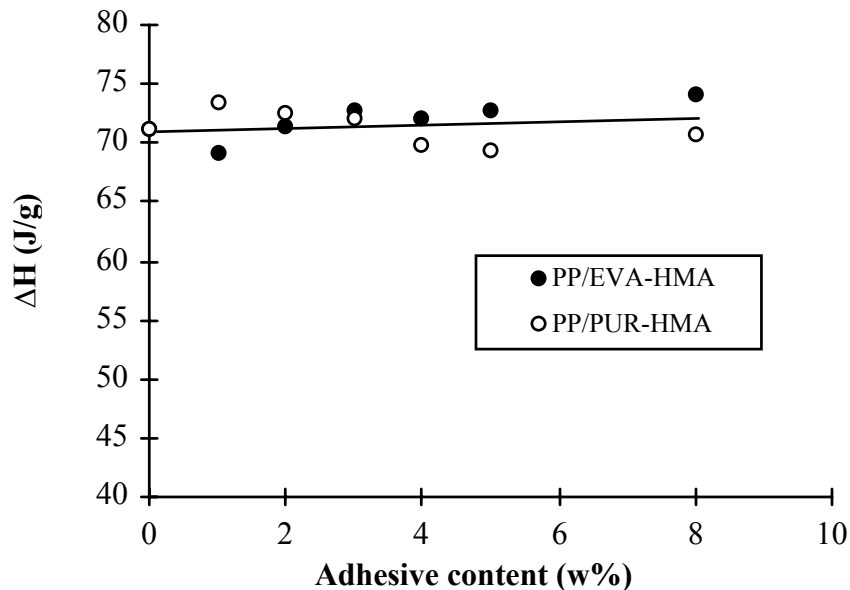


Figure 13. Melt enthalpy for PP with 0-8 w% of EVA- or PUR-HMA.

HIPS/ABS. The glass transition temperature, T_g , of the PS and ANS-phase respectively is not affected by contents up to 8 w% of PUR- or EVA-HMA, see Figures 14-15.

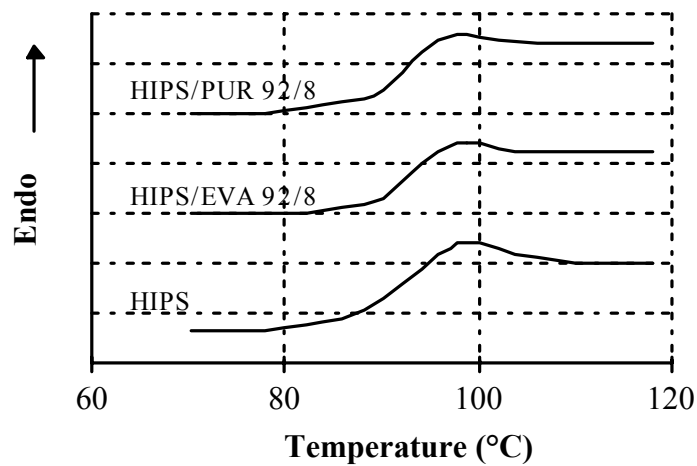


Figure 14. DSC - thermograms of HIPS containing 0 and 8 w% of PUR- or EVA-HMA.

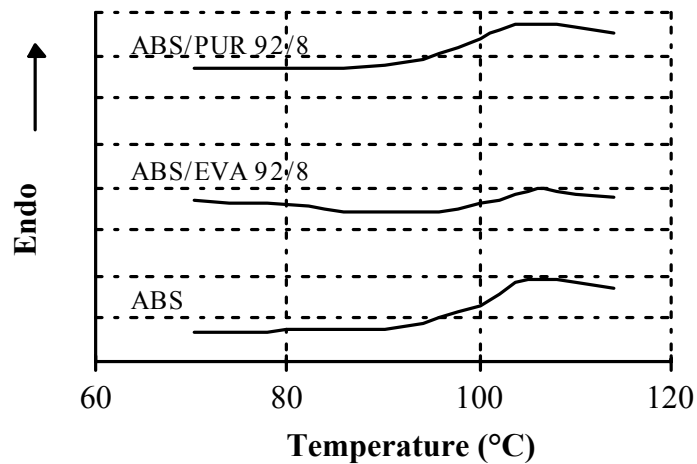


Figure 15. DSC - thermograms of ABS containing 0 and 8 w% of PUR- or EVA-HMA.

Scanning electron microscopy

Initiation of fracture. Examination of a large number of tensile and impact fracture surfaces revealed that fracture was always initiated at a defect, 50-100 μm in size. Furthermore, no correlation was found between the size and shape of the defect and the type and content of HMA. Thus, it can be concluded that initiation of fracture is not primarily caused by defects consisting of HMA residue.

Morphology. No distinct phases of PUR- or EVA-HMA could be identified in the fracture surfaces of PP, ABS and HIPS when observed as is by SEM. After etching of PP containing 8 w% of PUR-HMA using sulphuric acid, PUR-particles could be identified, see Figure 16. The distribution of PUR-particles was estimated from a great number of micrographs and a representative picture is given in Figure 17.

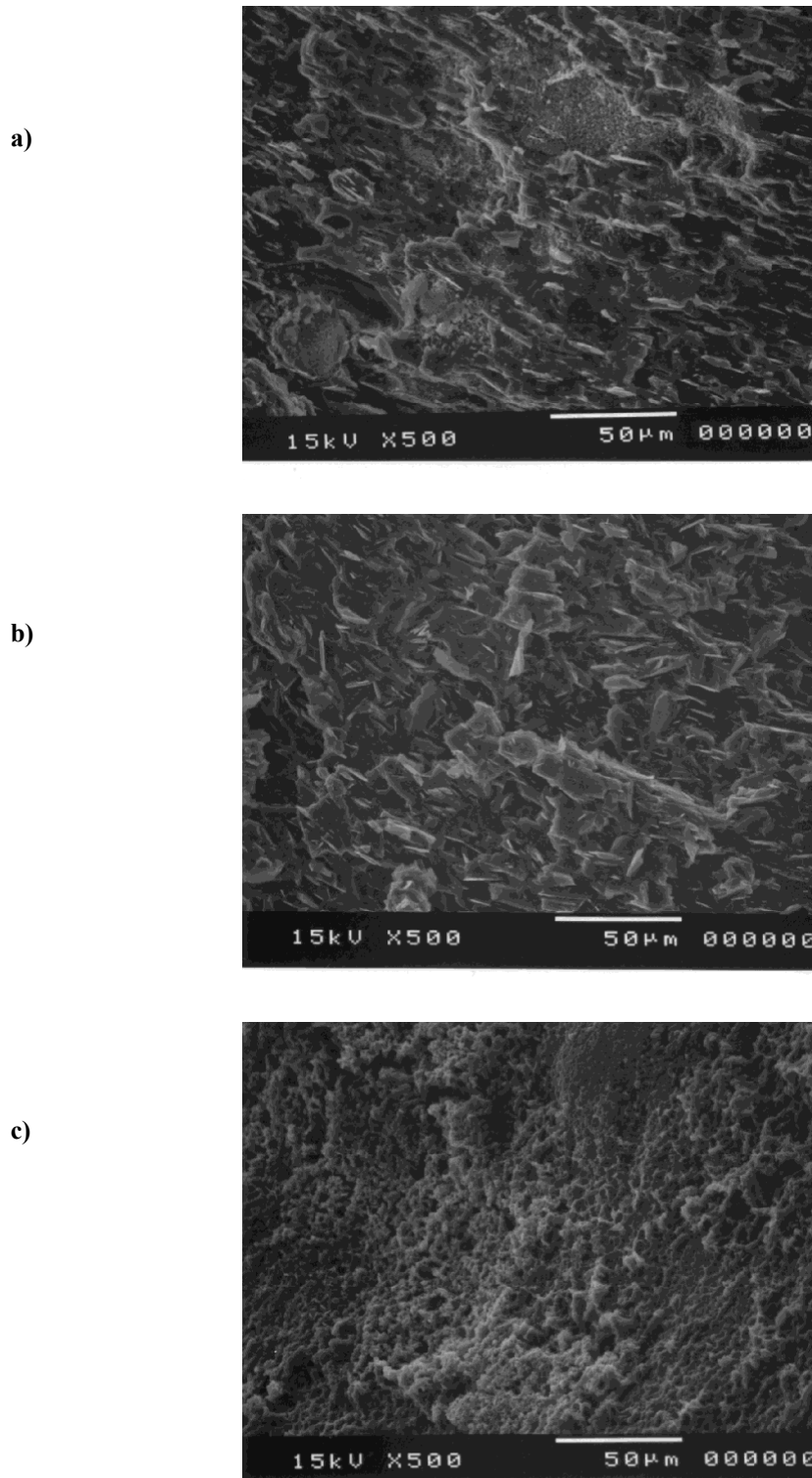


Figure 16. a) PP containing 8 w% PUR-HMA after etching in conc. sulphuric acid at 20°C
b) PP after etching in conc. sulphuric acid at 20°C.
c) PUR-HMA after etching in conc. sulphuric acid at 20°C.



Figure 17. Distribution of PUR-HMA particles (white) in PP (grey) in PP/PUR-HMA 92/8.

CONCLUSIONS

No significant loss of tensile or impact properties of PP, ABS and HIPS could be observed on addition of small amounts (0-2 w%) of PUR- or EVA-HMA. The melt enthalpy of PP is not significantly affected by addition of up to 8 w% of PUR- or EVA-HMA, indicating an unaffected crystallization of PP on addition of PUR- or EVA-HMA. The glass transition temperatures, T_g , of the PS-phase in HIPS and the ANS-phase in ABS are not affected by addition of up to 8 w% of PUR- or EVA-HMA. These results indicate that HMA exists as a separate phase in PP, HIPS and ABS. Furthermore, SEM-micrographs confirm the presence of PUR-HMA as a separate phase in the PP-matrix. From the SEM-micrographs it can also be concluded that fracture is initiated at particles of different shape showing no correlation to the type and content of HMA. Thus, fracture is generally caused by other defects than HMA-particles. It can be concluded that materials recycling of thermoplastic components of PP-, ABS- and HIPS-type can be performed without the separation of contaminating HMA-residue.

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