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Recycling of a plastic car component having a multilayer structure: Morphological and mechanical analysis

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The recycling of two types of dashboards having a multilayer structure presently available on the market and hence on the scrap cars has been extensively investigated. In both the cases the recycled material showed a strong worsening in the mechanical and impact properties. This effect was attributed to the incompatibility of the different materials constituting the item and to the occurrence of degradation phenomena taking place during processing. To overcome these limitations suitable polymeric additives were added during the recycling. By this procedure materials with improved mechanical properties able to be reused for the same or for similar applications within the car have been obtained.

1. Introduction

Recently much attention has been paid to the recycling of materials from durable goods. Automobiles figure prominently in this category. In fact, during the last twenty years plastics have been largely employed in the car industry for replacing metals due to their easier processability, more versatile design, lighter weight and corrosion resistance. This has led a growing interest to recycle the plastics in the cars at the end of their life cycle.

Actually a limited amount of expensive engineering plastics have been separated and recycled at a considerable economic cost. But for the majority of plastic components disassembly and separation are too difficult to be cost-effective. This has raised the question of whether the total mixed plastic can be recycled economically into useful materials [1–6].

In the present contribution the recycling of a multilayer internal component of the vehicle, such as dashboard, has been investigated. This item represents the more complex part of the car to be recycled owing to the large number of constitutive elements and because of the different polymeric materials, often mutually incompatible, used in its construction, which require a preliminary separation of the various components before the recycling process. All these aspects make the dashboard recovery not economically and industrially feasible. On the other hand, the weight of this component is very large with respect to the total amount of plastic present in the vehicle, so that, its recycling may represent a target of considerable technological relevance.

In this work two kinds of dashboards, which are presently available on the market and hence on the scrap cars, have been considered. The former is a two layers structure made by a support in acrylonitrile-butadiene-styrene (ABS) and a covering layer in polyvinyl chloride (PVC). The latter is a three layers structure constituted by a rigid support in ABS reinforced with glass fibers, an intermediate layer in polyurethane foam and a skin layer in ABS/PVC mixture.

The aim of the work was to optimise the recycling of each of these multilayer items by means of an economically feasible process, which does not involve the separation of the different layers. Moreover, to improve the end-properties of the resulting material, suitable polymeric additives, have been added during the recycling process. A morphological analysis by scanning electron microscopy (SEM), and a detailed mechanical characterisation by tensile and impact tests have been performed on the recycled materials.

2. Experimental

2.1. Materials

The two types of multilayer dashboards investigated were kindly supplied by the Research Centre of Fiat-Elasis (Naples-Italy). The first was made by an ABS support and a leather-like covering in PVC with the internal part in foamed PVC. The composition was:

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ABS 75 wt % and PVC 25 wt %. The second type was a three layers structure having an ABS support reinforced with 20 wt % of glass fibres, an intermediate layer of foamed polyurethane (PU) and a final leather-like of PVC/ABS mixture 50/50 by weight. The weight ratio of the constituents was: ABS support 70 wt %, polyurethane 20 wt % and PVC/ABS 10 wt %.

For both the dashboards as polymeric additive was used an ABS directly provided from Fiat-Elasis with a trade name ABS 95.150. It was the same employed in the construction of the dashboard support. The ABS 95.150 has an acrylonitrile (AN) content of 19.5 wt % and a melt flow index of 6 g/10 min.

2.2. Techniques

The dashboard was first granulated and then the recycling process was conduced by using a Haake 600 internal mixer at a temperature of 200 °C, roller speed of 32 r.p.m and for a mixing time of 5 min. Previous results showed that under such conditions thermomechanical degradative processes were significantly reduced.

The material so obtained was compression moulded to produce sheets of two different thicknesses (1 and 4 mm) at 200 °C and a pressure of 150 bars in a heated press. The 1 mm thick sheets were cut using a suitable hollow punch in dumb-bell shaped specimens on which tensile mechanical tests were performed. The 4 mm thick sheets were cut using a mill to obtain rectangular specimens 12 mm wide and 60 mm long for performing Charpy impact tests. Prior to testing the specimens were notched at the middle point of their length using a machine with a V-shaped tool. The value of notch depth was measured after fracture using an optical microscope.

Tensile mechanical tests were performed using an Instron machine model 4505 at ambient temperature and at a cross-head speed of 2 mm/min. The modulus, the stress and elongation at rupture were calculated from stress-strain diagrams on an average of six specimens.

Fracture tests were carried out at an impact speed of 1 m/sec using a Charpy instrumented Pendulum of Ceast DASWIN 4000. For all the materials examined a set of five samples were broken at ambient temperature and with a span of 48 mm. Curves of energy and load against time or displacement were recorded for each test.

Fracture surfaces of notched specimens were examined by using a Philips scanning electron microscope (SEM) mod. XL20. Prior to examination the surfaces were coated with a thin layer of a gold-palladium alloy in order to improve conductivity and prevent charging.

Dynamic-mechanical measurements at 1 Hz were made in the tensile mode using a Polymer Laboratories apparatus in the temperature range −150 to 200 °C and at a heating rate of 3 °C/min.

3. Results and discussion

3.1. Two layers dashboard

A preliminary analysis of the layers constituent the dashboard was performed. In Fig. 1A the tan δ curve as a function of temperature for the PVC covering layer is reported. A single broad relaxation process is observed in the whole temperature range investigated. The maximum of the tan δ peak occurs at about 17.5 °C. Generally, the tan δ curve of PVC exhibits two distinct relaxation processes referred to as β and α in order of increasing temperature. The β transition region is located at about −26 °C while the α is centred at 80–85 °C. According to literature data [7, 8] the α transition corresponds to the activation of relatively long-range motions of the PVC chains, involved in the glass transition phenomenon (T_g). On the other hand, the β transition is associated with the occurrence of molecular interactions among the chains. Therefore, the existence of a single relaxation process, exhibited by the PVC layer as well as its low T_g value strongly suggest the presence of a large amount of plasticiser. In fact, it has been found that the incorporation in PVC of a plasticiser, such as dioctylpthalate [7], leads to a lowering in the T_g and an increasing in the β transition. In particular, for an amount of plasticiser close to 30 wt %, the two transitions merge into one. These observations explain the breath and the low T_g value exhibited by the tan δ curve of the PVC layer. A further indication of the extensive plasticisation of the PVC component, arises from the shape of the tensile stress-strain diagram showed in Fig. 1B. It is noted that the behaviour

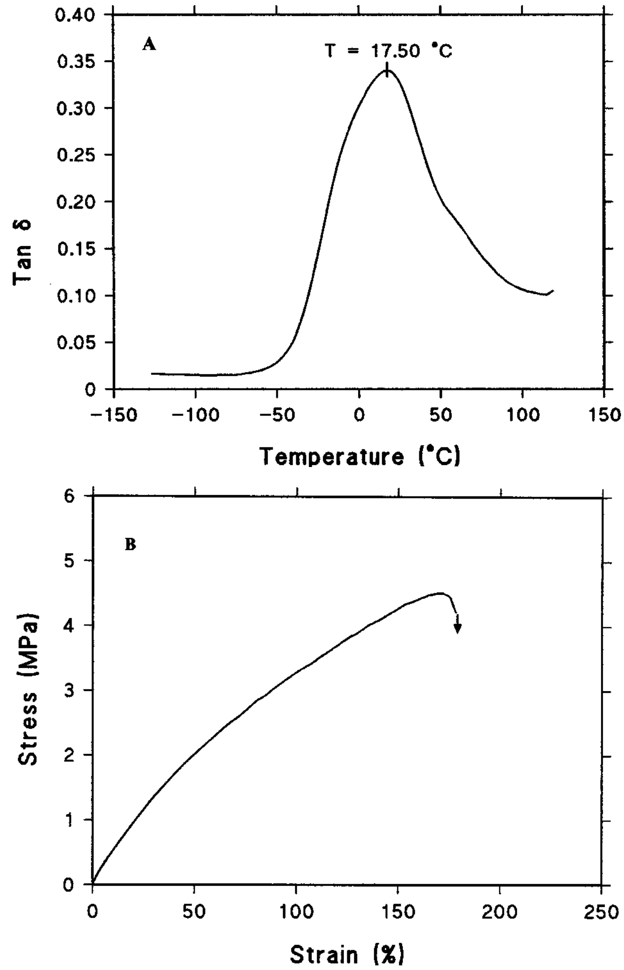


Figure 1 Tanδ curve as a function of temperature (Fig. 1A) and tensile stress-strain diagram (Fig. 1B) of the PVC covering layer of the two layers dashboard.

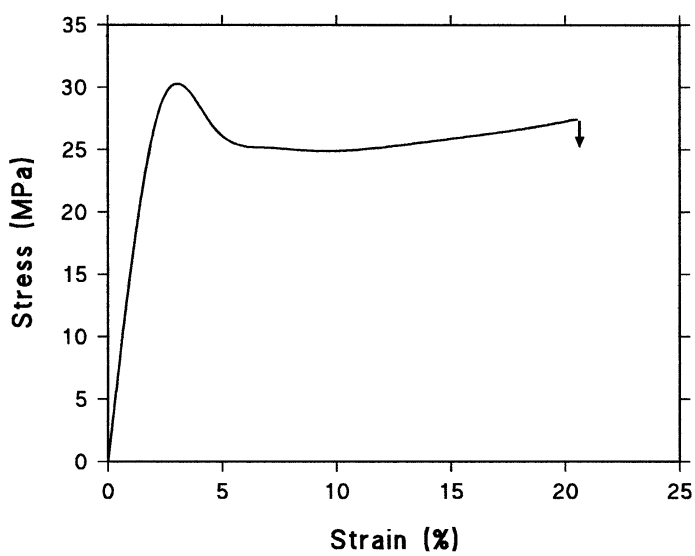
closely resembles that of a rubber-like material. The elastic modulus is low (about 48 MPa) and the elongation at break is high (about 180%) if compared to the unplasticised PVC.

The tensile stress-strain diagram of the ABS support is shown in Fig. 2A. As it can be expected, the behaviour is that of a toughened material. There is a well defined yield stress at about 30 MPa followed by strain softening to a lower yield stress of 25 MPa. The stress then rises slowly until the sample fractures at a strain of about 20%. The deformation process is accompanied by stress whitening and a small reduction of the cross-sectional area, indicating that shear yielding is the dominant mechanism occurring in this multiphase polymer.

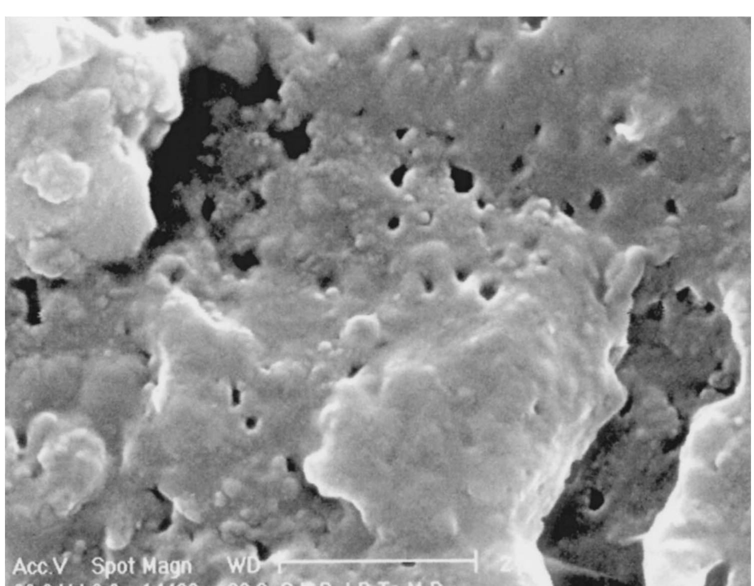
Previous studies [9–11] have shown that in ABS samples the operative deformation mechanism, namely shear yielding and/or crazing, is dependent on the size of rubber particles, rate of deformation and testing temperature. In particular it was found that, when small particles of about 0.1 μm in diameter are present, shear deformations are the major toughening mechanism. On the other hand, large particles of about 1.5 μm in diameter favour craze initiation. For intermediate dimensions both these mechanisms may occur simultaneously.

The average size of the rubber particles of the ABS support was evaluated by a SEM analysis performed on a fractured surface obtained by breaking a notched sample in liquid nitrogen. As it is evident from the SEM micrograph of Fig. 2B, most of the rubbery particles appear as circular cavities over the fracture surface, with a size range from 0.2 to about 0.6 μm. The matrix is very slightly deformed since the material has been fractured at a temperature lower than the T_g of the rubbery component (polybutadiene). From the above considerations on the size of the rubber domains, the shear yielding should be the main deformation mechanism occurring in the case of the ABS support.

Successively, the whole dashboard has been recycled following the procedure reported in the experimental



(A)



(B)

Figure 2 Tensile stress-strain diagram (Fig. 2A) and SEM micrograph of the ABS support of the two layers dashboard (Fig. 2B).

section. The tensile stress-strain diagram of the recycled material is shown in Fig. 3. As in the case of the ABS support, there is a pronounced yield stress at about 24 MPa, but it is followed by a rapid rupture. Thus, unlike the tensile behaviour of the single components, the mechanical response of the recycled product is typical of a brittle material. This result seems to be in contrast with the fact that ABS and PVC are considered to be compatible polymers. In fact, it has been reported that the PVC is miscible with Styrene-Acrylonitrile copolymers, SANs [12, 13], owing to molecular interactions formed between the electron-deficient α-hydrogen of vinyl-chloride groups of PVC and the electron rich nitrile group of the SAN [14–16]. Thus, in the case of ABS/PVC blends, a two-phase system is realized, consisting of a matrix made by a SAN/PVC miscible blend and a polybutadiene dispersed phase.

However, the literature contains conflicting results on this point [17, 18]. Several factors are responsible for this debate. First, in systems involving copolymers, miscibility is strongly affected by copolymer composition [19–21] and for some of the studies involving

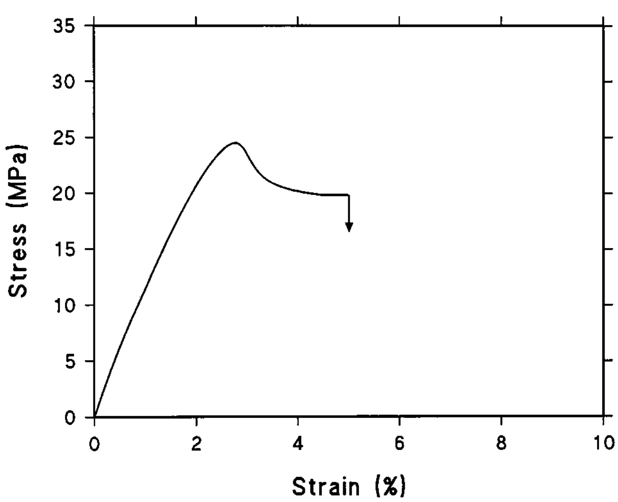


Figure 3 Tensile stress-strain diagram of the recycled two layers dashboard.

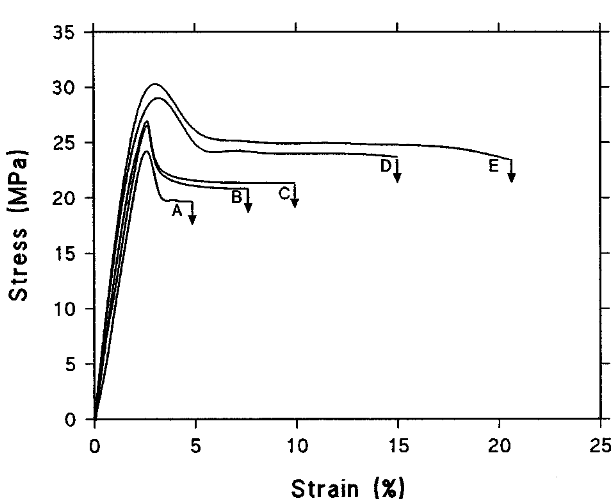


Figure 4 Tensile stress-strain diagrams of the recycle two layers dashboard processed with different amount of ABS: curve A, dashboard with 0 wt% of ABS; curve B, dashboard with 20 wt% of ABS; curve C, dashboard with 30 wt% of ABS; curve D, dashboard with 50 wt% of ABS; curve E, neat ABS resin.

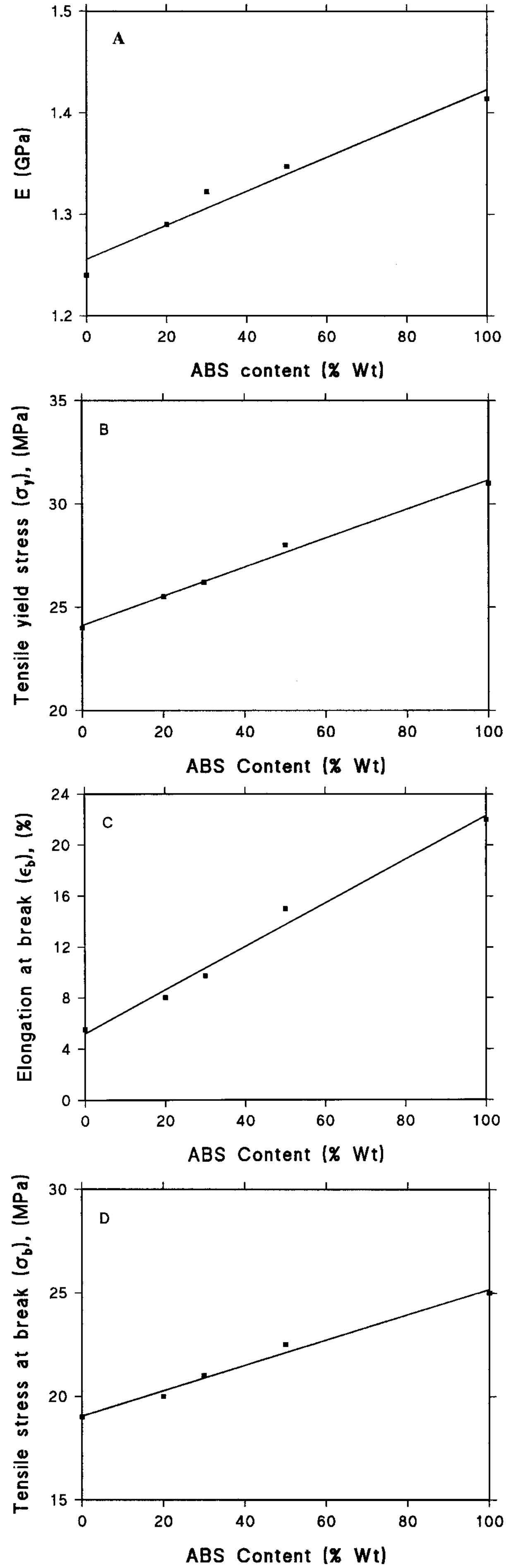
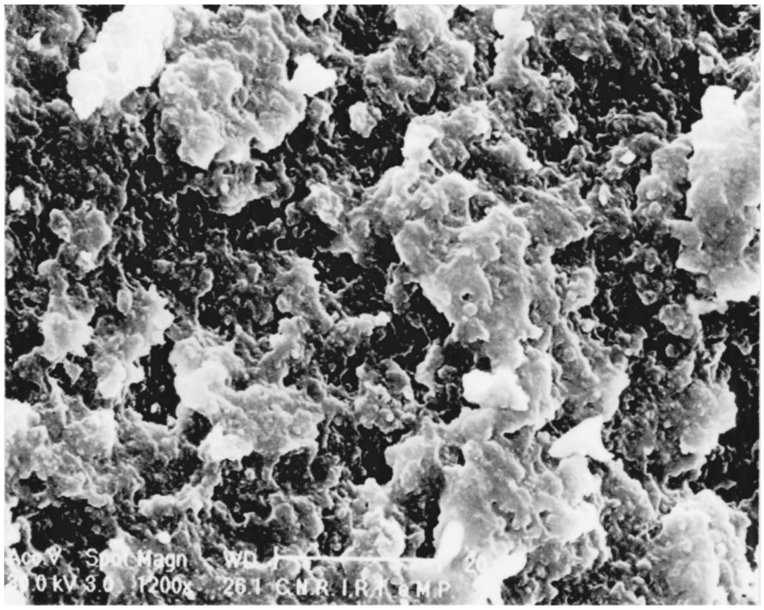
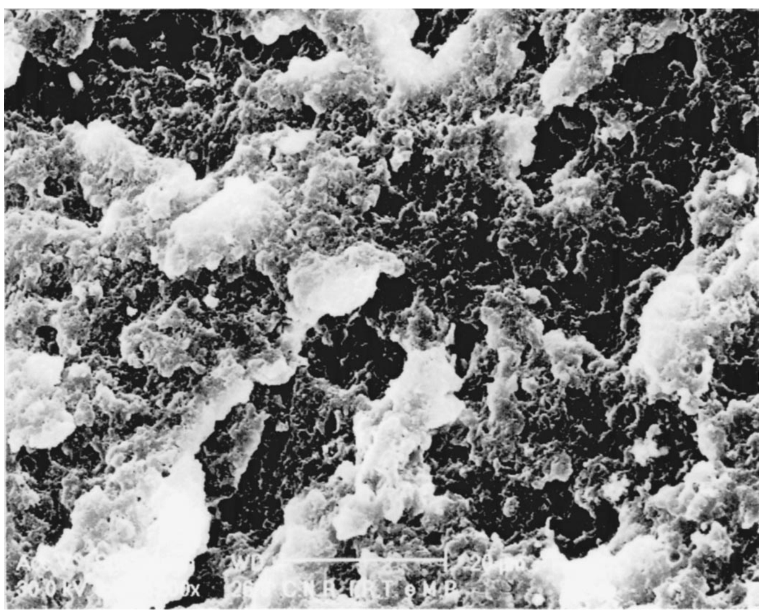


Figure 5 Tensile mechanical parameters evaluated from the stress-strain curves of Fig. 4, as a function of the ABS content: Fig. 5A, elastic modulus; Fig. 5B yield stress (σ_b); Fig. 5C elongation at break (ε_b); Fig. 5D stress at break (σ_b).



(A)



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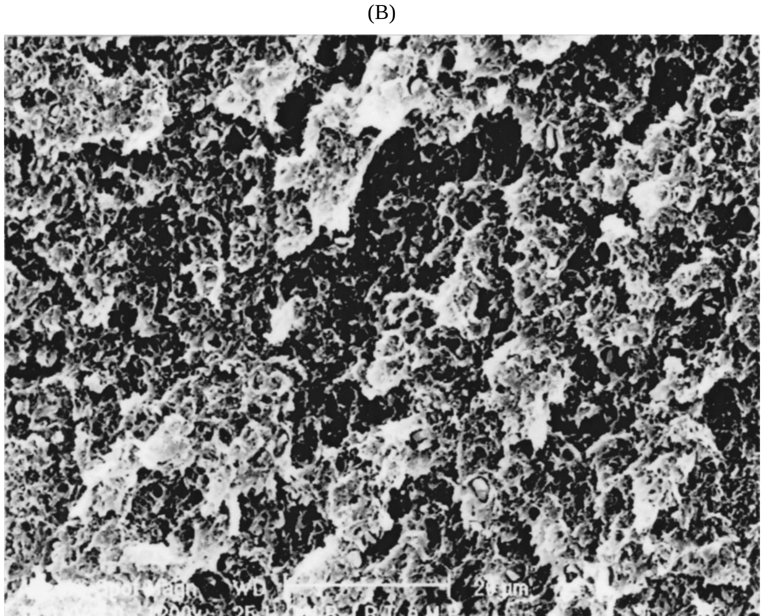


(C)

Figure 6 SEM micrographs of the tensile fractured surfaces of the two layers dashboard: A, unmodified dashboard; B, dashboard with 30 wt % of ABS; C, dashboard with 50 wt % of ABS.

SAN copolymers the composition is not known or not reported.

Second, in polymers blends, the degree of miscibility is influenced by the preparation procedure [22–24]. For example, two-phase blends may result for systems that are miscible when a too high temperature is used in melt mixing, owing to lower critical solution temperature (LCST) behaviour. In solvent casting procedures the so-called solvent effect may also induce phase separation of otherwise miscible components [19, 25].

For the PVC/ABS systems, the majority of works reported in the literature have established that the SAN composition plays a primary role in determining the degree of miscibility. In particular, results based on glass transition temperature measurements and on LCST behaviour, have indicated that for SAN copolymers having an acrylonitrile (AN) content ranging from about 11 to 26 wt %, miscibility with PVC is observed in the continuous phase. Thus, the reports on the immiscibility of PVC/ABS blends probably reflect either SAN copolymers having a composition outside this range, and/or melt mixing at temperatures above the phase boundary caused by LCST behaviour.

From the previous observations and considering the fact that the AN content in the ABS support is about 20 wt%, the recycling of the dashboard should give a compatible system, in the sense specified above, and hence the worsening of the mechanical properties showed by the recycled item, may be ascribed to the thermo-mechanical degradation taking place during processing. The main contribution arises from the PVC skin layer, which, although representing the minor component (about 25 wt %), has a very poor thermal stability compared to the ABS support.

It has been established that the thermal degradation of PVC starts at the glass transition temperature by elimination of hydrogen chloride, which is the main volatile product [26–28]. The intramolecular dehydrochlorination proceeds leading to conjugated double bonds (polyene sequences) in the polymer chain which cause secondary reactions, e.g. intramolecular cyclization and crosslinking with neighbouring PVC chains.

The extent of the thermal degradation process of PVC is strongly dependent on the processing conditions and in particular it is enhanced by increasing the temperature, the mixing time and the mechanical stresses. Moreover, in the case of ABS/PVC mixtures, the presence of degraded PVC may also induce thermo-oxidative mechanisms in the rubbery phase of the ABS component. Thus, the reprocessing of PVC or items containing PVC should be carried out in presence of heat stabilisers. Most of these additives are organic metal salts which act as hydrogen chloride acceptors to prevent its catalytic effect on the process of degradation.

In general, a relatively high concentration of heat-stabilisers is employed when the PVC is first processed. Consequently, at the end of the first life-cycle the material should still contain a certain amount of stabilisers for reprocessing, although further addition of these agents is often recommended.

In our case we have tried to minimize the PVC thermo-mechanical degradation through a procedure in which, during the recycling process, different amount of virgin ABS were added in order to reduce the total percentage of PVC in the system.

In Fig. 4 are reported the stress-strain diagrams of the dashboard recycled with 0, 20, 30 and 50 wt % of ABS 95.150 (curves A, B, C and D respectively). On the same figure is also shown, for comparison, the stress-strain diagram of the neat ABS 95.150 (curve E).

It can be observed that there is a general improvement of the tensile mechanical behaviour by increasing the ABS content. An amount of 50% of virgin ABS leads to a stress-strain curve very close to that of the pure ABS. The effect of the ABS addition on the tensile mechanical parameters is better seen in Fig. 5 where the elastic modulus (Fig. 5A), the yield stress (Fig. 5B) and ultimate properties such as the stress (Fig. 5C) and the elongation at break (Fig. 5D) are reported as a function of the ABS content. It is found that both the low (elastic modulus) and the large strain properties (yield stress and stress and elongation at rapture) increase linearly with enhancing the ABS concentration. These results indicate a substantial reduction of the degradation process which can be attributed essentially to the fact that the ABS reduces the amount of PVC in the system and hence its unfavourable effect on the mechanical properties. In particular, as it is shown in Fig. 5D, for a formulation containing 50 wt % of ABS the elongation at break, which is the parameter more sensitive to degradation, increases more than three times with respect to the recycled dashboard.

This improvement in the mechanical behaviour was confirmed by a morphological analysis performed by SEM on the tensile fractured surfaces. In Fig. 6 are shown the SEM micrographs of the recycled dashboard (Fig. 6A) and of the same material containing 30 and 50 wt % of ABS (Fig. 6B and C respectively). Although the morphology of the recycled item appears quite homogeneous, features distinctive of a plastic deformation mechanism are scarcely visible. When virgin ABS is added, a stress whitening during the tensile deformation process is generally observed. This phenomenon is clearly shown in the corresponding fracture surfaces in which plasticity and tearing are evident. The extent of such an effect increases with enhancing the ABS content (see Fig. 6B and C). The above observations account for the improved tensile mechanical properties and, at the same time, give further evidence that the shear yielding is the dominant deformation mechanism.

Mechanical measurements were also carried out under impact conditions in order to evaluate the fracture toughness of the recycled material under rapid loading.

The corresponding values of the impact strength as a function of the ABS content are shown in Fig. 7. The fracture toughness increases linearly with increasing the amount of ABS and, as in the case of the tensile parameters, a substantial enhancement is achieved for a formulation containing 50 wt % of ABS.

A SEM examination of the fracture surfaces (see Fig. 8A and B) reveals a morphology quite different from that observed in tensile tested samples, although the fractographic analysis has been conduced at lower magnification. In particular, at higher deformation rate the fractured surfaces appear smoother and the shear

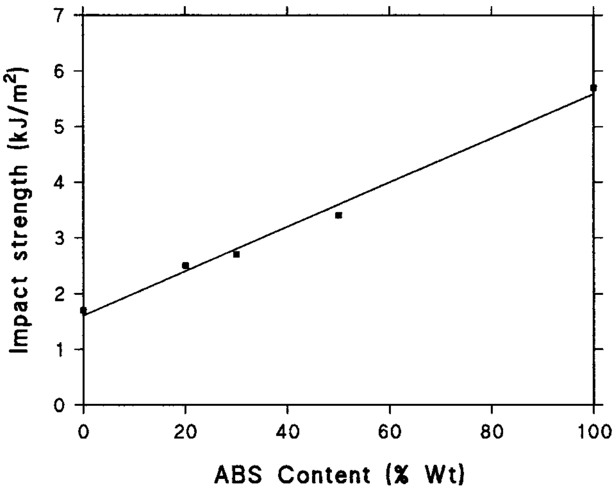
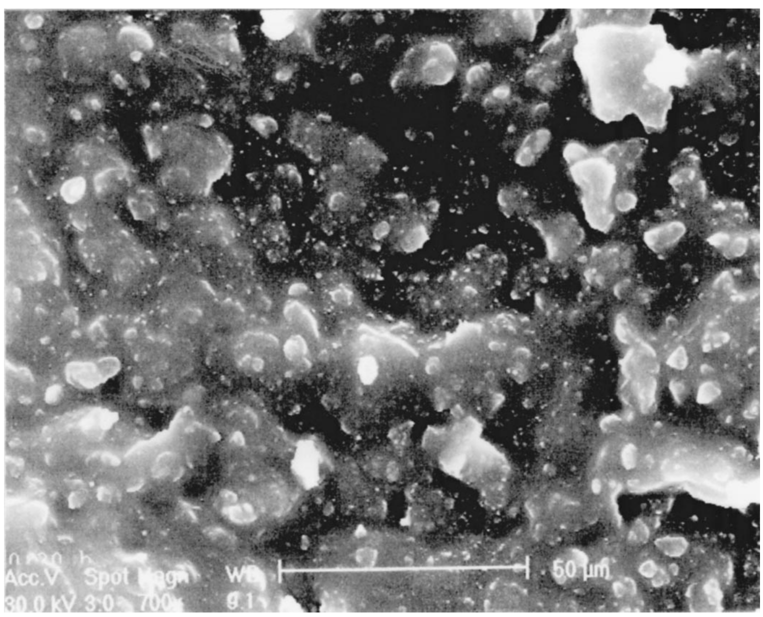


Figure 7 Impact strength of the two layers dashboard as a function of the ABS content.

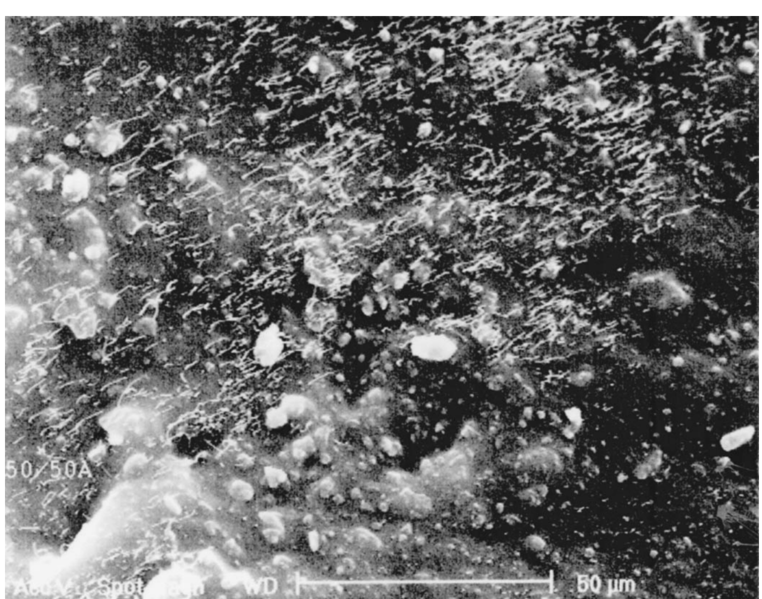
yielding mechanism seems to be partially suppressed and replaced by crazing. In fact, as shown in Fig. 8B, the plastic deformation features visible on the fracture surface of the recycled dashboard containing 50 wt % of ABS are probably due to the occurrence of craze formation during the fracture process. This mechanism is completely absent for the unmodified dashboard (Fig. 8A) which exhibits the fracture morphology of a brittle material.

3.2. Three layers dashboard

As in the previous case an analysis of the various layers constituent the dashboard was performed before the recycling process. In Fig. 9 are reported the tensile stress-strain diagrams of the covering ABS/PVC layer (Fig. 9A) and of theABS support (Fig. 9B). The former (Fig. 9A) shows a stress-strain curve typical of



(A)



(B)

Figure 8 SEM micrographs of the impact fractured surfaces of the two layers dashboard: A, unmodified dashboard; B, dashboard with 50 wt % of ABS.

TABLE I Tensile mechanical parameters of the three layers dashboard

Sample	Elastic modulus (GPa)	Stress at break (MPa)	Strain at break (%)
ABS/PVC layer	0.22	9.5	110
ABS support	2.3	37.0	2.0
Recycled dashboard	1.5	9.1	1.3

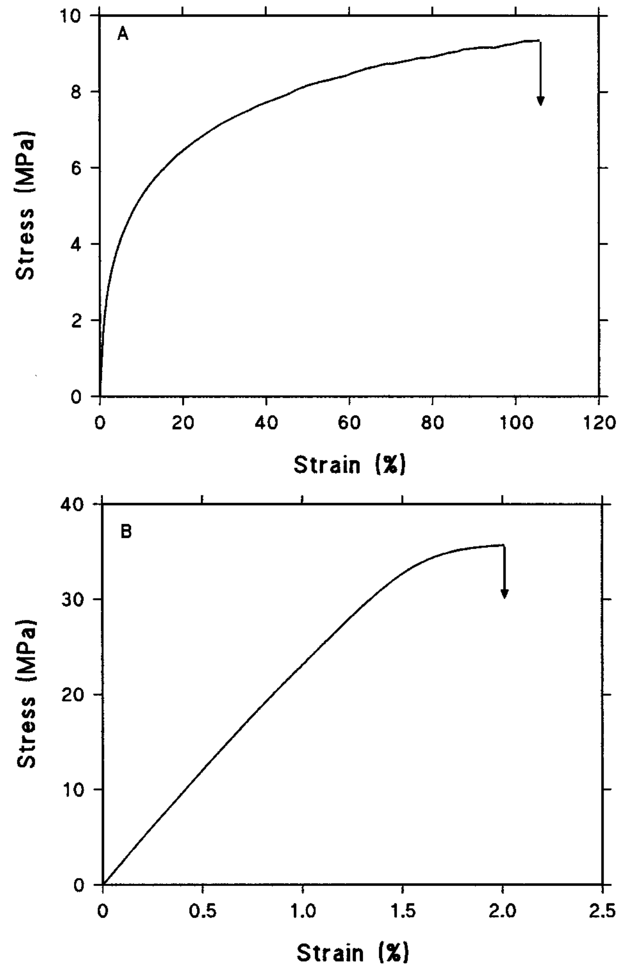


Figure 9 Tensile stress-strain diagrams of the ABS/PVC covering layer (curve A) and of the ABS support (curve B) of the three layers dashboard.

a ductile material with a high deformation at rupture. The corresponding values of modulus, stress (σ_b) and deformation (ε_b) at break are reported in Table I. A visual inspection, during the test, has evidenced that the yielding phenomenon is not accompanied by a localised necking but the deformation process proceeds through a gradual and uniform reduction of the cross-sectional area of the sample before fracture. The absence of a well defined yield stress, which is normally exhibited in ABS and PVC sample, as well as the observed low value of the modulus can be ascribed to the presence of a high amount of plasticiser present in the PVC component.

On the contrary, the tensile stress-strain curve of the ABS support (see Fig. 9B) resembles that of a brittle material. The fracture takes place at an elongation not exceeding 2.0% and as soon as the yielding starts. The other mechanical parameters evaluated from Fig. 9B are summarised in Table I. The observed fragile behaviour, as will be shown from the SEM analysis, can be ascribed to the presence of glassy fibres embembed in the ABS support as reinforcement. The glassy fibres acting as stress concentrators induce the formation of cracks at their ends, which propagate along the fibre sides due to the shear deformation of the matrix. As a consequence of the occurrence of these cracks, the load capability of the fibres is much reduced, and the ABS matrix is required to support a much higher stress. The matrix cracks grow from the interfacial cracks just prior to the failure. These interfacial cracks and matrix cracks leads to a catastrophic fracture of the composite in contrast with the ductile behaviour generally shown by the unfilled ABS (see Fig. 2A).

The mechanical tensile behaviour of the recycled dashboard is shown in Fig. 10A. As in the previous case, the recycling leads to an overall worsening of the mechanical properties compared to the constituent layers. This effect is well evidenced in Table I where the modulus and the ultimate parameters of the recycled material are reported. In this case, unlike the recycling of the two components dashboard, the observed deterioration in the mechanical properties cannot be attributed exclusively to the poor thermal stability of the PVC component, which constitutes only about 5.0 wt % of the entire weight of the item, but the major contribution arises from the presence of the foamed polyurethane (PU) layer. This thermosetting material is highly incompatible with ABS and PVC and, due to its cross-linked structure, cannot be re-melted. Thus, during the recycling, the PU component is only crushed into fine particles as is shown from the SEM micrograph of Fig. 11. Since the interfacial adhesion of these PU particles is very poor, they can act as weak points in the material, favouring a rapid formation and growth of cracks and consequently a premature fracture.

The above considerations on the unfavourable effect of the polyurethane layer are confirmed by the shape of the stress-strain curve of Fig. 10B which shows the tensile behaviour of the dashboard recycled in absence of the foamed polyurethane layer. It can be observed that there is an overall improvement in the mechanical properties. This effect is also shown when the material is tested under impact conditions.

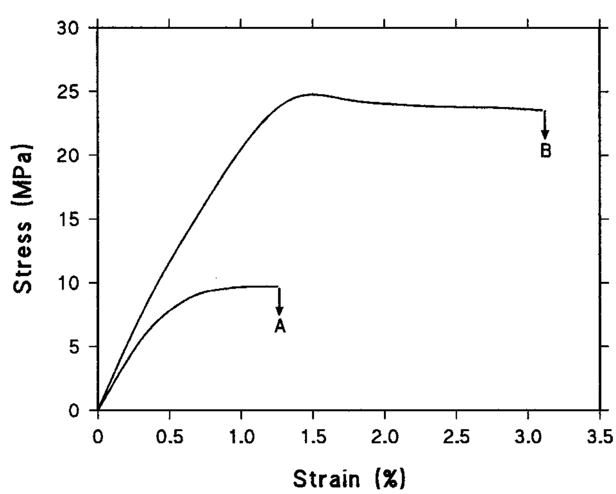


Figure 10 Tensile stress-strain diagrams of the recycled three layers dashboard (curve A) and of the same recycled in absence of the foamed PU layer (curve B).

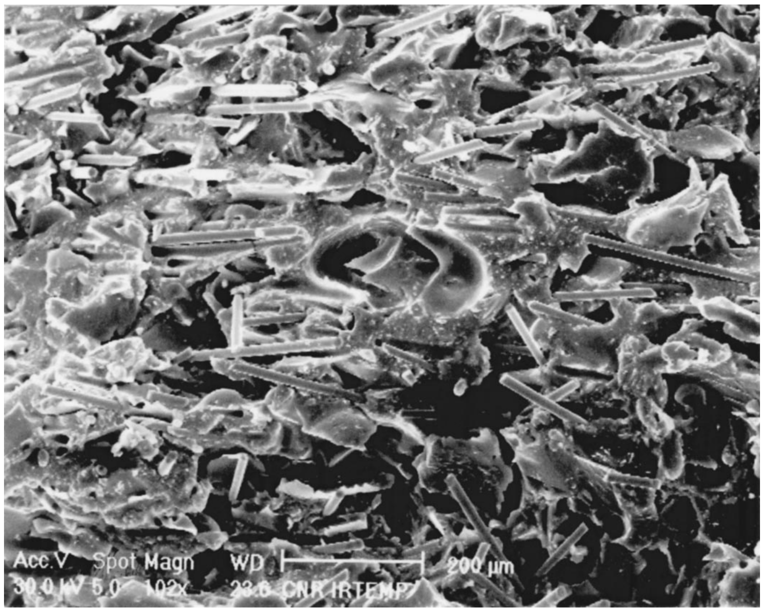


Figure 11 SEM micrographs of the fractured surface of the recycled three layers dashboard.

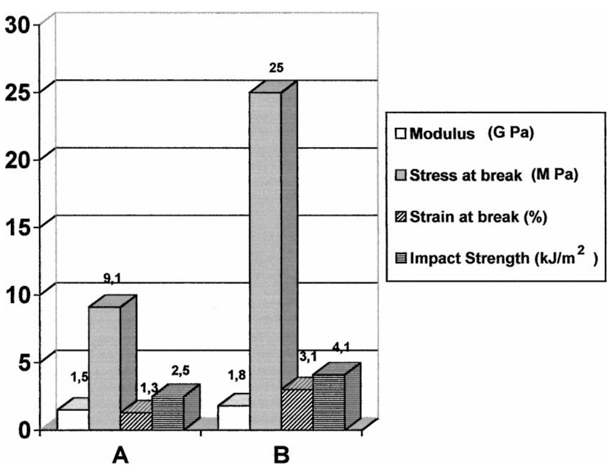


Figure 12 Bar-graph of the tensile and impact parameters of the three layers dashboard: A, dashboard recycled in presence of the PU layer; B, dashboard recycled in absence of the PU layer.

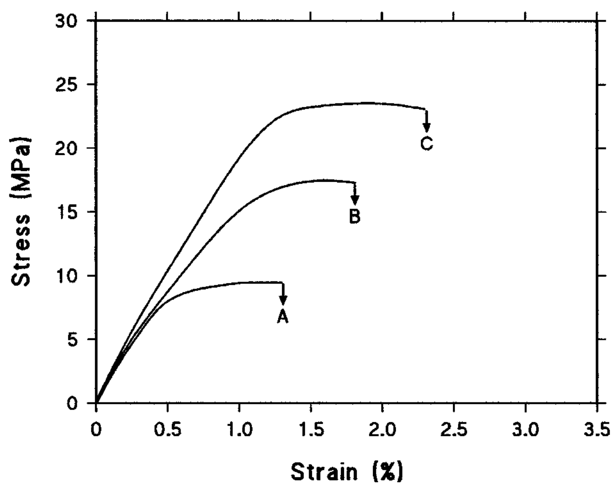


Figure 13 Tensile stress-strain diagrams of the recycled three layers dashboard processed with different amount of ABS: curve A, dashboard with 0 wt % of ABS; curve B, dashboard with 10 wt % of ABS; curve C, dashboard with 20 wt% of ABS.

The results of the tensile and impact tests are reported in the bar-graph of Fig. 12 and are compared with those of the dashboard processed in presence of the PU layer. This comparison shows that, in absence of PU, a material with satisfactory mechanical properties can be obtained. In fact, the modulus increases by about 20%, the stress and the elongation at break increase by about 175% and 130% respectively and the impact strength increases by more than 60%.

However, as already mentioned, the separation of the layers which constitute the dashboard may render the entire recycling process not economically favourable. Therefore for enhancing the mechanical performances of the recycled dashboard we have used, as in the previous case, a procedure based on the addition during the recycling process of virgin unfilled ABS.

In Fig. 13 are shown the stress-strain diagrams of the recycled dashboard, taken as reference material, (curve A) and of two formulations containing 10 and 20 wt % of ABS (curve B and C respectively).

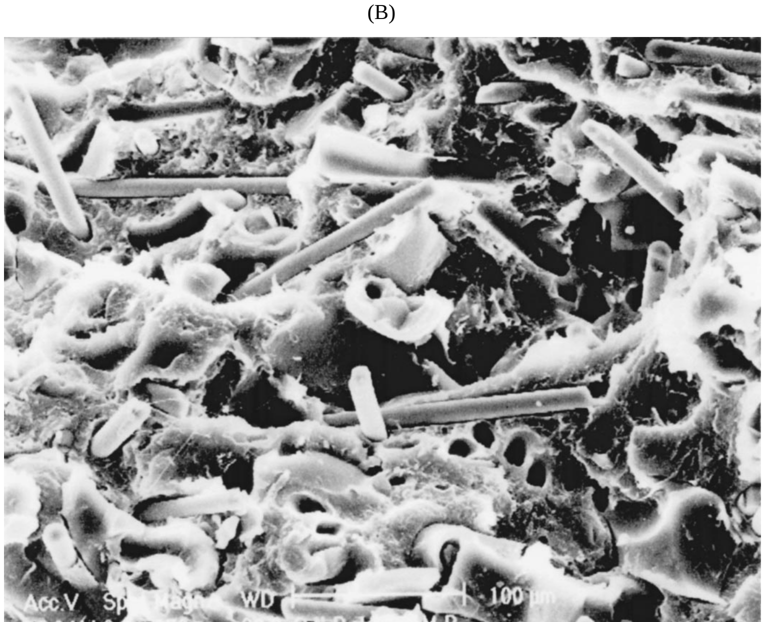
One notes that, the tensile behaviour improves considerable with enhancing the ABS content approaching to that of the dashboard recycled in the absence of PU.

In particular, by increasing the amount of ABS, a substantial enhancement in the ultimate parameters is achieved while the elastic modulus remains almost constant. An improvement in the impact strength is also detected as shown in Table II where the results of the above tensile measurements are also reported.

Quantitatively the addition of 20 wt% of ABS increases the stress and the elongation at break of about 150% and 75% respectively and the impact strength of about 110%. These results are in good agreement with the fractographic analysis performed by SEM on such materials. In Fig. 14 the SEM micrographs after impact tests of the unmodified and the modified dashboard containing 10 wt% and 20 wt% of ABS are reported. For the unmodified resin (Fig. 14 A), besides the already noted presence of PU particles, the fracture surface exhibits

(A)

(B)



(C)

Figure 14 SEM micrographs of the impact fractured surfaces of the three layers dashboard: A, unmodified dashboard; B, dashboard with 10 wt % of ABS; C, dashboard with 20 wt % of ABS.

TABLE II Tensile and impact parameters of the three layers dashboard recycled in presence of ABS

Sample	Elastic modulus (GPa)	Stress at break (MPa)	Strain at break (%)	Impact strength (kJ/m^2)
Recycled dashboard	1.5	9.1	1.3	2.5
Recycled dashboard with 10 wt % of ABS	1.5	17.0	1.8	3.8
Recycled dashboard with 20 wt % of ABS	1.6	23.0	2.3	5.2

a brittle morphology without any evidence of plastic flow, in accordance with its poor mechanical properties. The addition of ABS (Fig. 14 B and C) changes substantially the fracture morphology which shows features of plastic deformation whose extent increases with enhancing the amount of ABS in the formulation. Most probably the observed plastic deformation may be ascribed to a shear yielding mechanism which represents the main source of energy dissipation before fracture.

From the above results it may be concluded that the addition of ABS strongly reduces the unfavourable effect of the foamed PU layer, giving rise to a recycled material with balanced properties in terms of modulus and impact strength, depending on the amount of additive. Thus, by such a procedure, materials with properties suitable to be reused within the car can be obtained.

4. Conclusions

In the present contribution we have reported on the recycling of the dashboard, which is one of the more complex internal car components having a multilayer structure. In particular, two types of dashboards have been recycled without a preliminary separation of the various layers. In both cases the recycled material showed very poor mechanical and impact properties. This phenomenon has been attributed for the two layers dashboard to the thermo-mechanical degradation of the PVC skin, while for the three layers dashboard essentially to the presence of the foamed PU component which, being infusible and highly incompatible with ABS and PVC, leads to a material with a low degree of homogeneity.

A substantial enhancement in the mechanical and impact behaviour has been achieved by adding, during processing, different amount of virgin ABS. It has been demonstrated that, by an appropriate ABS content, it is possible to obtain materials with balanced mechanical properties in terms of modulus and toughness, capable to be reused for the same or similar applications within the car, as for instance in the manufacture of door panels or roof covers.

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