

Recycled Multilayer Cartons as Cellulose Source in HDPE-Based Composites: Compatibilization and Structure-Properties Relationships

Maurizio Avella, Roberto Avolio, Irene Bonadies, Cosimo Carfagna, Maria Emanuela Errico, Gennaro Gentile

Istituto di Chimica e Tecnologia dei Polimeri, Consiglio Nazionale delle Ricerche, Pozzuoli 80078, NA, Italy

Received 23 April 2009; accepted 9 June 2009

DOI 10.1002/app.30913

Published online 16 July 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: In this article, an alternative mechanical recycling of multilayer carton scraps (MC), consisting in the use of MC without the physical separation of its different fractions (cellulose fibers and low-density polyethylene, 80/20 wt/wt), is proposed. In particular, MC was considered as a source of cellulose fibers in the obtainment of high-density polyethylene (HDPE)-based composites. Composites containing up to 60 wt % of milled MC were prepared by reactive processing, i.e., by adding different amount (5 and 10 wt %) of a linear low-density polyethylene grafted with maleic anhydride (coupling agent, maleated linear low-density polyethylene) during HDPE/MC mixing. Then, structure/

properties relationships were deeply investigated as a function of MC and coupling agent content. The coupling agent was able to induce a complete polymeric covering of cellulose fraction as well as a strong HDPE/cellulose interfacial adhesion. As a consequence, a significant improvement of mechanical properties at low and high deformation rates as well as a very interesting response of composites to water vapor permeability was obtained. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 114: 2978–2985, 2009

Key words: compatibilization; composites; interfaces; mechanical properties; recycling

INTRODUCTION

Plastic manufacturing, including flexible films and rigid containers, has quickly dominated the packaging industry because of their interesting performances (optical, thermal, barrier, and mechanical properties, low density, easy processability) and effective cost with respect to other materials.

Such packaging materials are becoming the major component of the plastic waste stream also because of the short-life cycle, thus seriously contributing to overburden the management of the urban solid waste disposal. Recycling of polymeric residues in blends or even in composites has found a great interest in the market representing the most promising waste disposal strategy.^{1–3}

Nowadays, besides neat plastic resins, the use of multilayer containers is growing for food and beverage packaging, and their recycling process is even more complicated.⁴

Within this class of packaging materials, multilayer cartons (MC), mainly constituted by cellulose (80 wt %) and low-density polyethylene (LDPE, 20 wt %), are among the most used. For aseptic cartons, an aluminium sheet is also added to improve UV and oxygen barrier. Temperature is responsible

for gathering all of the layers, so that no glue or hot melt is added.

Unfortunately, the production of MC gives rise to a large amount of industrial scraps, mainly due to defected, not well-welded containers and scraps remaining from the cutting process. As a matter of fact, the set up of recycling strategies of multilayer preconsumer residues is a very interesting challenge also from the industrial point of view.⁴

Actually, MC recycling essentially involves the separation of paperboard from other layers. The cellulose separation is performed through a wet process leading to a cellulose pulp that can be reused to produce industrial paper products, such as corrugated cartons, and consumer products, such as paper towels. Then, polymer and aluminium residues are recycled following three different approaches: (1) generation of energy through incineration in a biomass reactor; (2) recovery of aluminium in pyrolysis oven; (3) processing of LDPE and aluminium to realize composites used as injection molded parts for low-cost housing materials (EcoAllene[®], Lecce Pen Verona S.r.L., Castelnuovo del Garda, Italy, Marahlene[®], Tetra Pak Italiana S.p.A., Rubiera, Italy).⁵ All these recycling processes are very complex and expensive.

In this article, an alternative mechanical recycling of MC, consisting in the use of MC without the physical separation of its different fractions is proposed. In particular, preconsumer MC industrial

Correspondence to: M. E. Errico (bors@ictp.cnr.it).

scraps, constituted by 80 wt % of cellulose and 20 wt % of LDPE, have been used as a source of cellulose fibers in the realization of high-density polyethylene (HDPE)-based composites.

Composites based on polymers filled with natural fibres are attracting the attention of many industrial sectors, such as the automotive industry. Natural fibers are renewable resources readily available in many countries and exhibit relatively low cost and good specific properties.^{6,7} Recently, the use of recycled cellulose to produce composites has been proposed, in some case with promising results.^{8–10}

However, it is well known that different surface properties between fiber (highly polar) and polymer matrix such as polyolefin (non-polar and hydrophobic) require the set up of a proper compatibilization strategy. Without such a strategy, weak interfaces are generated, likewise a poor ability of the polymer to completely wet fiber surface and consequently overall properties of the composite are almost substandard.

As a matter of fact, in this paper a proper coupling agent (maleated linear low density polyethylene) has been selected and HDPE based composites containing up to 60 wt % of MC scraps have been prepared through reactive processing. Structure/properties relationships have been deeply investigated as a function of the coupling agent and MC amount through morphological, thermal, and mechanical analysis as well as water vapor permeability (WVP) tests.

EXPERIMENTAL

Materials

MC constituted by cellulose and LDPE (80/20 wt/wt) were kindly supplied by Tetra Pak Italiana S.p.A. (Latina, Italy), as production scraps.

Before using, scraps were milled in a Retsch SM100 cutting mill equipped with a bottom sieve (conidur holes, 1 mm in diameter). The obtained powder was dried under vacuum at 90°C overnight and kept in a desiccator. After milling the fiber, average length and diameter, measured by scanning electron microscope (SEM), were 500 and 15 μm , respectively.

HDPE (Alathon M6580, density 0.965 g/cm^3 , melt index 8.2 g/10 min at 190°C and 2.16 kg) was supplied by Equistar (Texas).

Maleated linear low-density polyethylene (MAPE), trade name AGRIMAL-HD, density 0.92 g/cm^3 , was supplied by Agricola Imballaggi (Pagani, Italy). The content of maleic anhydride grafted onto polyethylene is 1% by weight, evaluated by titration.¹¹

Composites preparation

HDPE/MC composites were prepared by melt mixing in an internal mixer (Rheocord EC of HAAKE, New Jersey).

HDPE was mixed with different amounts of MAPE (0, 5 and 10 wt %) at 175°C for 10 min. then the mixture was cooled and pelletized. Afterwards, each mixture was mixed with different amount of dried MC powder (20, 40, and 60 wt %) at 175°C for 10 min. Composites and neat HDPE were successively compression molded at 180°C to obtain films (thickness 100 \div 200 μm) and sheets (thickness 1 and 3.5 mm).

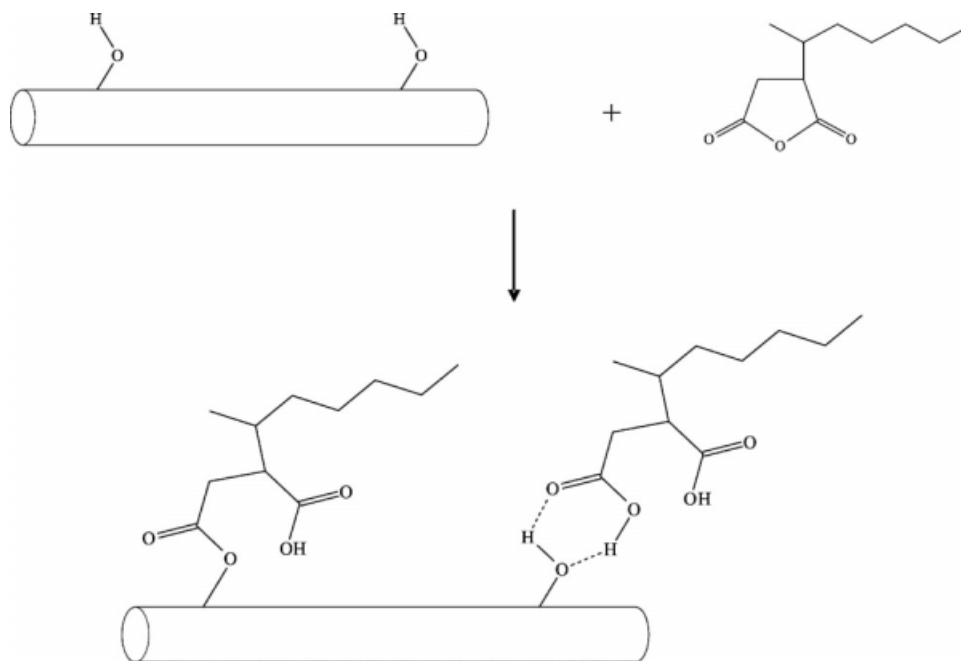
Techniques

Morphological analysis of composites was performed by using a scanning electron microscope (SEM), Philips XL20 series, on cryogenically fractured surfaces and on fractured surface of samples after impact test. Before the observation, samples were coated with an Au-Pd alloy with a SEM-coating device (SEM BALTEC MED 020).

Differential scanning calorimetry (DSC) analyses were carried out using a Mettler-Toledo DSC 822 in N_2 flux with the following temperature program: heating from 25°C to 180°C, cooling from 180°C to 0°C, and heating from 0°C to 180°C. The heating/cooling rate was set to 10°C/min, and the values of enthalpy were calculated from the II heating run. The crystallinity content was calculated considering the effective polymeric content (sum of the HDPE, LDPE, and MAPE fractions) and using the enthalpy of fusion of PE in the fully crystalline state of 293 J/g.¹²

Tensile tests were performed on dumb-bell specimens (4 mm^2 cross-section, 1-mm thickness, 25-mm gage length) at room temperature and cross-head speed of 5 mm/min by using a Instron machine model 4505, according to ASTM D638 test method. Young's modulus (E) and stress at break (σ_r) were calculated as average values over 10 tested samples. Fracture tests were carried out with a Charpy Ceast Resil Impactor equipped with a DAS 4000 Acquisition System, using an impact energy of 3.6 J and an impact speed of 1 m/s. Samples (10.0-mm wide, 3.5-mm thick, and 60-mm long) with a notch depth to width ratio of 0.3 and a span length of 48.0 mm were fractured at room temperature according to ASTM D256 test method. For each material, five specimens were tested and the average values of resilience and peak force were calculated.

Before mechanical tests, samples were conditioned at 30°C and 50% relative humidity (RH) for 24 h. WVP was measured according to the ASTM E96 standard, using the upright cup test. Film specimens were conditioned for 48 h in a chamber at $T = 30^\circ\text{C}$ and 50% before the test. Films were sealed on cups containing distilled water. Test cups were placed in a conditioned environment at $T = 30 \pm 1^\circ\text{C}$ and $\text{RH} = 50 \pm 2\%$. The permeation rate of the water vapor



Scheme 1 Scheme of interaction between anhydride groups and cellulose hydroxyl groups.

across the film was measured gravimetrically on three specimens for each sample, by weighting the cup every 24 h until the permeation rate reached a constant value.

RESULTS AND DISCUSSION

The enhancement of performances in multicomponent polymer-based materials is often ascribed to interactions between different phases because the property of transferring takes place through the interfacial region. Therefore, the set up of a proper compatibilization strategy plays a critical role to obtain high-performance composites.

In this article, HDPE/MC composites containing up to 60% by weight of MC were prepared by reactive mixing at process conditions compatible with thermo-oxidative stability of the cellulose fraction and polymer phases (HDPE, LDPE) whose starting degradation temperatures were previously evaluated by means of dynamic thermogravimetric analysis, thus preventing eventual degradation phenomena during composite preparation.

The reactive mixing consisted in the addition of different amount (5 and 10 wt %) of LLDPE grafted with 1 wt % of maleic anhydride (MAPE), as coupling agent, during processing. This coupling agent is able to interact both with cellulose fraction and HDPE, thus acting as a linkage phase. In fact, hydroxyl surface groups of cellulose can interact with MAPE carboxyl moieties through covalent and/or hydrogen bonds, according to the simplified Scheme 1. At the same time, LLDPE backbone can

intermingle with polymer matrix. In Table I, composition and codes of prepared materials are reported.

The effectiveness of the coupling agent on the HDPE/MC interfacial adhesion and fibre dispersion was investigated by morphological analysis (SEM) performed on the cryogenically fractured surface of samples.

In Figure 1, morphologies of uncompatibilized composites containing the lowest (a) and the highest (b) content of MC are reported. As shown, fibers do not present any preferential orientation and their distribution results almost homogeneous. Moreover, fibers appear uncovered by the polymeric fraction, and no adhesion between phases seems to be present, as demonstrated by fiber tracks. In Figure 1(c,d),

TABLE I
Composition and Relative Codes of Prepared Materials

HDPE + MAPE (%)	MC (%)	MAPE/HDPE weight ratio (%)	Code
100	0	0	HDPE
80	20	0	HDPE/MC 80–20
60	40	0	HDPE/MC 60–40
40	60	0	HDPE/MC 40–60
100	0	5	HDPE/M5
80	20	5	HDPE/M5/MC 80–20
60	40	5	HDPE/M5/MC 60–40
40	60	5	HDPE/M5/MC 40–60
100	0	10	HDPE/M10
80	20	10	HDPE/M10/MC 80–20
60	40	10	HDPE/M10/MC 60–40
40	60	10	HDPE/M10/MC 40–60

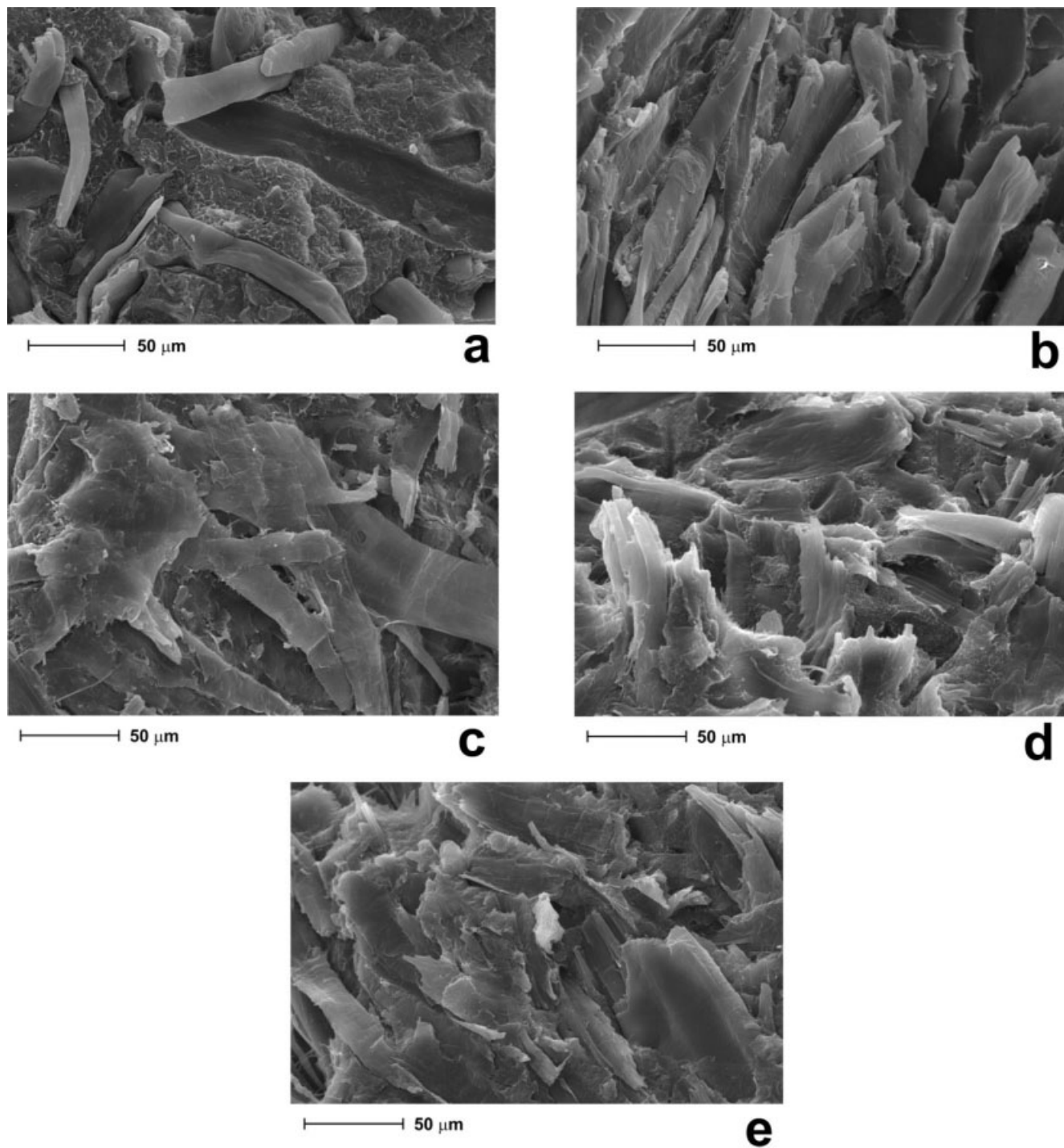


Figure 1 SEM micrograph of cryogenically fractured surface: (a) HDPE/MC 80/20; (b) HDPE/MC 40/60; (c) HDPE/M5/MC 80/20; (d) HDPE/M5/MC 40/60; (e) HDPE/M10/MC 40/60.

SEM micrographs of composites containing 5 wt % of MAPE and the lowest (20 wt %) and the highest (60 wt %) MC amount are reported, respectively. Fibers emerge completely embedded and bonded to the polymeric phase, no pull out and debonding phenomena take place after the applied load. These evidences remark a strong interconnection between HDPE and MC, as a consequence of interactions

generated during the reactive mixing. Nevertheless, it must be highlighted that the addition of 5 wt % of MAPE is not enough to guarantee a complete polymeric wetting of the fibrous phase at the highest MC content. In fact, a small amount of uncovered cellulose fibers is distinguishable, although a strong interconnection between phases is achieved. On the contrary, the morphology of composite at 60 wt % of

TABLE II
Results of DSC Analysis: Crystallization Temperature (T_c), Melting Temperature (T_m), and their Relative Crystallinity Content

Composite code	T_c (°C)	x_c (%)	T_m (°C) (II Run)
HDPE	116	67	138
HDPE/MC 80–20	115	64	140
HDPE/MC 60–40	116	60	139
HDPE/MC 40–60	116	58	137
HDPE/M5	117	64	136
HDPE/M5/MC 80–20	114	64	139
HDPE/M5/MC 60–40	117	59	137
HDPE/M5/MC 40–60	117	53	136
HDPE/M10	116	63	136
HDPE/M10/MC 80–20	115	62	138
HDPE/M10/MC 60–40	115	56	138
HDPE/M10/MC 40–60	117	53	134

MC is restored with the addition of 10 wt % of MAPE, see Figure 1(e).

In Table II, results of DSC analysis are reported. MC phase does not influence the temperature of main phase transitions. However, composites exhibit lower crystallinity than neat HDPE as a function of MC content. This can be ascribed to the hindering effect of the fibrous fraction on the HDPE crystallization.^{13–16} Moreover, MAPE induces a further crystallinity decrease, slightly correlated to its amount, mainly depending on HDPE/MC interactions that reduce the molecular mobility of polyethylene.

Mechanical analysis

Mechanical analysis of neat HDPE- and HDPE-based composites was performed at low (tensile) and high (impact) deformation rates.

As concerning the tensile test, no yielding phenomena occur, the specimens break down at very small elongation due to the presence of an unde-

formable phase according to a brittle mechanism. A significant increase in the elastic modulus was recorded as a function of MC content for both compatibilized and uncompatibilized systems, as reported in Table III. The extent of this improvement is up to 200% in the case of uncompatibilized materials, whereas it results slightly lower and correlated to the MAPE amount for compatibilized composites. These results can be justified considering that the modulus of a composite is strongly dependent on the modulus of components but only slightly sensitive to the interfacial adhesion. In fact, it is measured at very small deformation, when simple physical contact among components is sufficient to transfer the stress. As a matter of fact, the inclusion of a rigid phase, such as cellulose fibers, is able to increase the polymer stiffness. On the contrary, the lower increase of Young's modulus as a function of MAPE content could be ascribed to the intrinsic lower MAPE modulus and to the above discussed effect of the coupling agent on the HDPE crystallization. Conversely to the modulus, stress at break is very sensitive to the interfacial adhesion and for this reason it can be used to probe the strength of polymer/filler interactions. In fact, this parameter refers to not negligible deformations, so that the interface plays a crucial role in transferring the stress from the matrix to the fiber phase.¹⁷ Stress at break undergoes a drastic decrease (up to 50%) as a function of MC content for uncompatibilized materials (Table III), whereas in the presence of MAPE it remains almost unchanged with respect to HDPE even at the highest MC content. Then, the interfacial adhesion has been quantitatively estimated through the calculation of the interfacial parameter B , following the method developed by Turcsányi et al.¹⁸ The composite stress at break (σ_b) can be expressed as follows:

$$\sigma_b = \sigma_{b0} \frac{1 - \varphi_f}{1 + 2.5\varphi_f} \exp(B\sigma_b\varphi_f) \quad (1)$$

TABLE III
Results of Tensile and Impact Tests: Young's Modulus (E), Stress at Break (σ_b), Resilience and Peak Force Values

Composite code	E (MPa)	σ_b (MPa)	Resilience (KJ/m ²)	Peak Force (N)
HDPE	1288 ± 53	29 ± 1	1.6 ± 0.1	60 ± 2
HDPE/MC 80–20	1705 ± 67	21 ± 1	1.6 ± 0.2	55 ± 1
HDPE/MC 60–40	2166 ± 77	16 ± 2	1.4 ± 0.3	59 ± 4
HDPE/MC 40–60	2647 ± 131	13 ± 2	1.4 ± 0.3	59 ± 3
HDPE/M5	1185 ± 58	29 ± 1	–	–
HDPE/M5/MC 80–20	1530 ± 34	31 ± 1	2.5 ± 0.1	63 ± 5
HDPE/M5/MC 60–40	2059 ± 78	33 ± 2	3.4 ± 0.3	85 ± 3
HDPE/M5/MC 40–60	2476 ± 59	26 ± 2	3.8 ± 0.5	93 ± 4
HDPE/M10	1048 ± 22	27 ± 1	–	–
HDPE/M10/MC 80–20	1410 ± 36	29 ± 1	3.2 ± 0.1	69 ± 3
HDPE/M10/MC 60–40	1854 ± 55	32 ± 2	4.4 ± 0.4	86 ± 6
HDPE/M10/MC 40–60	2329 ± 95	26 ± 2	3.7 ± 0.3	86 ± 10

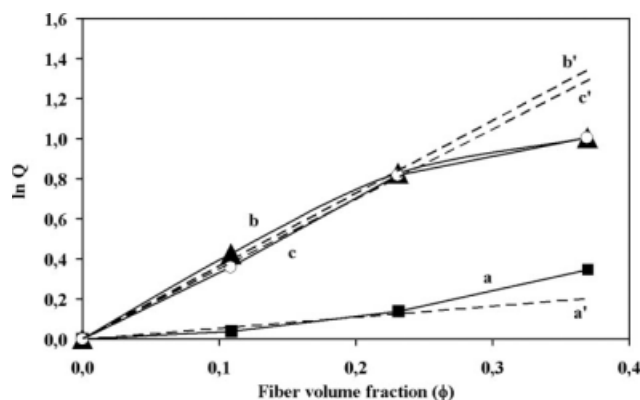


Figure 2 Plot of $\ln Q$ versus fiber volume fraction: (a) HDPE/MC (■); (b) HDPE/M5/MC (▲); (c) HDPE/M10/MC (○). Dashed lines (a', b' and c', respectively) represent the linear fitting of experimental data in the linear range.

where σ_{b0} is the matrix stress at break, ϕ_f is the filler volume fraction, and $B_{\sigma b}$ is the interaction parameter. The fiber volume fraction was calculated considering a density of 1.50 g/cm^3 for cellulose fibers and 0.92 g/cm^3 for LDPE in MC.^{19,20}

In this equation, the fractional term accounts for the effective load-bearing cross section while the exponential term refers to the matrix-filler interaction. The interaction parameter $B_{\sigma b}$ depends on surface area and density of the filler as well as on thickness and strength of the interphase. As a matter of fact, $B_{\sigma b}$ increases with the increasing of polymer/filler adhesion.²¹ It can be calculated by plotting eq. (1) in the following form:

$$\ln Q = B_{\sigma b} \phi_f \quad (2)$$

where

$$Q = \left(\frac{\sigma_b}{\sigma_{b0}} \frac{1 + 2.5\phi_f}{1 - \phi_f} \right) \quad (3)$$

The graph of $\ln Q$ versus ϕ_f is shown in Figure 2 for uncompatibilized (curve a) and compatibilized (curves b and c) composites. As it can be observed, curves present a linear trend up to 40 wt % of MC, whereas the highest fiber content induces a deviation from the linear correlation between $\ln Q$ and ϕ_f . This evidence can be explained considering that at 60 wt % of MC, fiber/fiber interactions are not negligible while $B_{\sigma b}$ only takes into account matrix/fiber interactions.¹⁷ However, comparing the slope of curves a, b and c in the linear range, a relevant increase of the interaction parameter for compatibilized ($B_{\sigma b} = 3.5$, curves b and c) with respect to uncompatibilized materials ($B_{\sigma b} = 0.5$, curve a) can be evidenced, thus confirming the effectiveness of the compatibilization strategy. Moreover, this trend is almost independent from the amount of MAPE. In fact, the curves corresponding to compo-

sites containing 5 wt % and 10 wt % of MAPE are practically overlapped, as shown in Figure 2 (curves b and c). This suggests that 5 wt % of compatibilizing agent is enough to induce strong interactions between phases.

As concerning impact parameters, resilience and peak force values are reported in Table III. The MC phase does not seem to deteriorate the toughness of HDPE, leaving almost unchanged the resilience and peak force values in the case of uncompatibilized composites. On the contrary, the addition of MAPE induces a significant increase of these parameters up to 280% for the resilience and to 150% for the peak force, thus indicating an outstanding improvement of the toughness.

Rigid fillers, such as cellulose fibers, mainly used to improve the stiffness of a polymeric matrix, in some cases are also able to improve the resilience.^{22,23} Several fracture mechanism can be activated depending on particle size or aspect ratio and surface adhesion²⁴; as an example, weak interface could represent a preferential path for the crack growth.

The toughening mechanism in fiber reinforced composites can be qualitatively described by considering that the energy dissipated by the fracture propagation can be evaluated as the sum of three main components: matrix fracture energy, fiber fracture energy, and an interaction term accounting for debonding and pull-out phenomena.²⁵ The fiber fracture energy and the interaction term are negligible for short or weak fibers. In this case, the overall toughness of the composite is worsened independently from the interfacial adhesion. On the contrary, both these terms can exceed the matrix contribution by adding long and/or strong fibers thus justifying an improvement of the toughness as a function of the interfacial strength.

In HDPE/MC uncompatibilized composites, the fracture mechanism involves evident fiber pull out and debonding phenomena due to a poor adhesion between the phases, as shown in Figure 3(a). In this condition, fibers are not able to bear the applied stress because slipping out phenomena occur. However, it must be underlined that pull out phenomena adsorb a minor part of the involving fracture propagation energy. This can partially compensate the effect of the weak interfacial adhesion on the mechanical response, thus allowing only a slight decrease of impact parameters. On the contrary, in the presence of MAPE, the fracture mechanism prevalently regard the breaking of fibers that anyway remain well welded to the polymer matrix [Fig. 3(b)]. Therefore, the fiber breaking occurs as a result of the stress transferring from matrix to fiber and requires a large amount of energy. These evidences explain the significant enhancement of the

toughness and peak force in the case of compatibilized materials.

Water vapor permeability

The hydrophilic nature of cellulose is responsible for its strong tendency to adsorb water. This is one of the main drawbacks that limit the use of cellulose fibers in polymer composites. In fact, the water adsorption/desorption compromises the dimensional stability of fibers and thus overall composite properties. As a matter of fact, the evaluation of the WVP is particularly relevant to verify potential applications of prepared materials, as an example in the packaging sector.

The WVP was calculated from the slope (G) of a linear regression of weight loss versus time.

$$\text{WVP} = \frac{G \cdot x}{A \cdot \Delta P} \quad (4)$$

where x is the film thickness, A is the area of exposed film, and ΔP is the differential water vapor

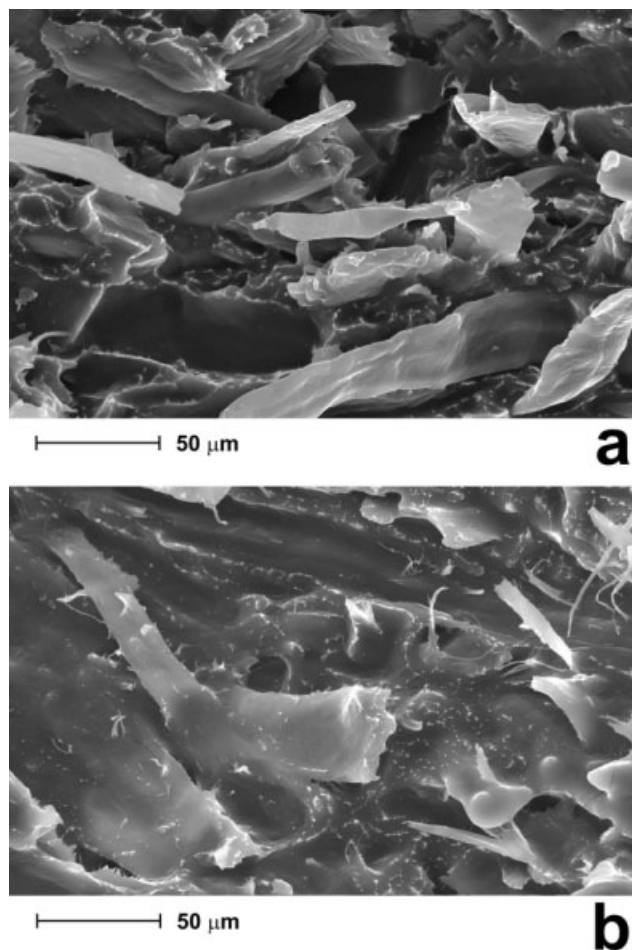


Figure 3 SEM micrograph of impact fractured surface of (a) HDPE/MC 60/40 (b) HDPE/M5/MC 60/40.

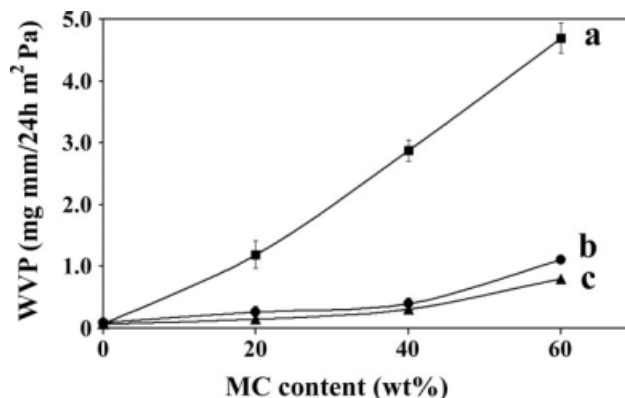


Figure 4 Water vapor permeability as a function of MC content: (a) uncompatibilized composites; (b) HDPE/M5/MC composites; (c) HDPE/M10/MC composites.

pressure across the film. Test results are shown in Figure 4.

As expected, composites are more permeable to water vapor with respect to HDPE and the extent of this phenomenon depends on the MC amount and compatibilization strategy. As concerning uncompatibilized composites, the WVP drastically increase at the highest amount of MC (40 and 60 wt %), up to values two order of magnitude greater than HDPE. This behavior can be clearly attributed to the fact that fibers are not well embedded into polymer matrix. In this condition, fiber surfaces result totally exposed and easily accessible to diffusive water vapor.

On the other hand, the addition of MAPE allows to reduce by one order of magnitude the water permeability with respect to uncompatibilized materials. This result can be explained taking into account that fibers appear completely covered by the polymer matrix that protects them from the water. Moreover, WVP is a function of MAPE content. This finding can be supported considering that only the addition of 10 wt % of MAPE is able to guarantee a complete coverage of fibers at the highest amount of MC.

CONCLUSIONS

In this article, an alternative mechanical recycling of MC industrial scraps is proposed. In particular, MC have been considered as a direct source of cellulose fibers to realize HDPE-based composites. Composites have been prepared by a reactive approach consisting in the addition of different amount of maleated polyethylene (MAPE) as coupling agent during the mixing. The analysis of structure/properties relationships has clarified the role of the coupling agent and demonstrated the effectiveness of the compatibilization strategy on the improvement of composite final properties.

Cellulose fibers appear completely wetted and well welded to HDPE phase as well as a good

interfacial adhesion between the components is reached with the addition of MAPE. A significant increase of the Young's Modulus as a function of MC content has been recorded, independently from the reactive process. On the contrary, stress at break values drastically decrease in the case of uncompatibilized composites while they are comparable with that of HDPE with the addition of the coupling agent. The calculation of an interaction parameter has permitted to quantitatively evaluate the interfacial adhesion, thus confirming the effectiveness of the coupling agent. Moreover, a very interesting improvement of the composite toughness has been pointed out and correlated to the strong interfacial adhesion. The WVP of composites has been drastically reduced in the presence of the coupling agent. This finding is justified considering that a complete polymeric covering of the fiber surface protects them from water absorption/desorption phenomena.

On the basis of the above discussed results, these materials can be considered as a valid alternative for common application sectors in which fibre-reinforced polyolefins are already in use. Moreover, the reduced WVP obtained through compatibilization lets to suppose possible applications for packaging.

The authors thank Mr. Vincenzo Di Liello and Mr. Giuseppe Narciso for their precious technical assistance on mechanical and morphological characterization, respectively. Agricola Imballaggi is gratefully acknowledged for supplying AGRIMAL-HD. Tetra Pak Italiana S.p.A. is gratefully acknowledged for supplying MC.

References

- Ambrose, C. A.; Hooper, R.; Potter, A. K.; Singh, M. M. *Resour Conserv Recycling* 2002, 36, 309.
- Da, S.; Paula, M. M.; Medeiros Rodrigues, F. B. B.; Bernardin, A. M.; Fiori, M. A.; Angioletto, E. *Mater Sci Eng A* 2005, 403, 37.
- Morris, J. *J Hazard Mater* 1996, 47, 277.
- Mourad, A. L.; Garcia, E. E. C.; Braz Vilela, G.; Von Zauben, F. *Resource Conserv Recycling* 2008, 52, 678.
- Lopes, C. M. A.; Felisberti, M. I. *J Appl Polym Sci* 2006, 101, 3183.
- Avella, M.; Casale, L.; Dell'erba, R.; Focher, B.; Martuscelli, E.; Marzetti, A. *J Appl Polym Sci* 1997, 68, 1077.
- Karnani, R.; Krishnan, M.; Narayan, R. *Polym Eng Sci* 1997, 37, 476.
- Ren, S.; Hon, D. N. S. *J Reinforced Plast Compos* 1993, 12, 1311.
- Baroulaki, I.; Mergos, J. A.; Pappa, G.; Tarantili, P. A.; Economides, D.; Magoulas, K.; Dervos, C. T. *Polym Adv Technol* 2006, 17, 954.
- Fahramand, F.; Mehrabzadeh, M. *J Appl Polym Sci* 2001, 80, 2573.
- Sclavons, M.; Carlier, V.; De Roover, B.; Franquinet, P.; Devaux, J.; Legras, R. *J Appl Polym Sci* 1996, 62, 1205.
- Wunderlich, B. *Macromolecular Physics*; Academic Press: New York, 1980; Vol. 3.
- Avella, M.; Bogoeva-Gaceva, G.; Bužarovska, A.; Errico, M. E.; Gentile, G.; Grozdanov, A. *Polym Eng Sci* 2007, 47, 745.
- Stark, N. M.; Matuana, L. M. *Polym Degrad Stab* 2004, 86, 1.
- Colom, X.; Canavate, J.; Pages, P.; Saurina, J.; Carrasco, F. *J Reinforced Plast Compos* 2000, 19, 818.
- Xie, X. L.; Li, R. K. Y.; Tjong, S. C.; Mai, Y.-W. *Polym Compos* 2002, 23, 319.
- Dányádi, L.; Renner, K.; Móczó, J.; Pukánszky, B. *Polym Eng Sci* 2007, 47, 1246.
- Turcsányi, B.; Pukánszky, B.; Tüdős, F. *J Material Sci Lett* 1998, 7, 160.
- Hepworth, D. G.; Bruce, D. M. *Compos A* 2000, 31, 283.
- Brandrup, J.; Immergut, E. H., Eds. *Polymer Handbook*, 3rd ed.; John Wiley & Sons Inc, Interscience Publishers: New York, 1989.
- Pukánszky, B. *New Polym Mater* 1992, 3, 205.
- Avella, M.; Bogoeva-Gaceva, G.; Bužarovska, A.; Errico, M. E.; Gentile, G.; Grozdanov, A. *J Appl Polym Sci* 2007, 104, 3192.
- Avella, M.; Bogoeva-Gaceva, G.; Bužarovska, A.; Errico, M. E.; Gentile, G.; Grozdanov, A. *J Appl Polym Sci* 2008, 108, 3542.
- Karger-Kocsis, J. *Polypropylene: Structure, Blends and Composites*; Chapman & Hall: London, 1995; Vol. 3.
- Clemons, C. *Woodfiber-Plastic Composites: Virgin and Recycled Wood Fiber and Polymers for Composites Conference Proceedings*; Forest Products Society: Madison, WI, 1995.