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ENVIRONMENTAL LIFE CYCLE STUDIES OF POLY(HYDROXYBUTYRATE)- AND POLYPROPYLENE-BASED COMPOSITES

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Abstract

A comparative life cycle assessment (LCA) has been performed in order to evaluate the possible environmental benefits of replacing conventional petrochemical plastics with poly(hydroxybutyrate) (PHB) based composites (filled with sugar cane bagasse (SCB) or organophilic montmorillonite, OMMT), or polypropylene (PP) based composites (filled with OMMT). The end products studied are packaging film (conventionally produced from PP), agricultural film (conventionally produced from low-density polyethylene, LDPE), a cathode ray tube (CRT) monitor housing (conventionally produced from high impact polystyrene, HIPS) and internal car panels (conventionally produced from glass fibre filled polypropylene, PP-GF). The environmental impact is evaluated on the basis of non-renewable energy use (NREU) and the global warming potential over a 100 years time horizon (GWP100). The results for the case studies show that the use of a SCB or OMMT filler alone does not necessarily decrease the environmental impacts. However, if the material properties of the filled polymer (higher Young modulus or tensile strength, lower density) are clearly better compared to the conventional material, this is likely to be the case. Environmental benefits can also be enabled by more environmentally friendly polymers used as matrix of the composite, as was the case for PHB. In the waste stage, free nanoparticles might be released (e.g. as a consequence of biodegradation), which could lead to adverse health effects. In conclusion, the few cases studied in this paper indicate that polymer nanocomposites can offer new opportunities for saving non-renewable energy and mitigating greenhouse gas emissions but further research is required to identify the most promising fields and to avoid negative side effects.

Key words: Life cycle assessment (LCA), composite, nanoclay, sugar cane bagasse.

1. Introduction

In the last two decades the preparation of composites has been one of the key topics in polymer science. More recently, two classes of composites have received a lot of attention, namely nanocomposites and natural fibre composites. Nanocomposites are very much appreciated because with a relatively small quantity of filler, commonly an organophilic modified clay, it is possible to improve several polymer properties, e.g., to reduce gas permeability, to improve thermal stability and to enhance mechanical properties [1,2]. But also composites filled with natural fibres (e.g. flax, hemp, wood flour) offer good mechanical properties combined with low density and cost. [3,4]

In applications such as structural elements and films, the improved mechanical properties, caused by the addition of fillers allow to reduce the amount of material required to fulfil the function. As a result of material savings, the environmental impact of composite products can be expected to be lower than that of products made out of conventional material, provided that the energy required to produce and to embed the fillers is not too high. In order to assess the possible environmental benefits of polymers filled with nanoclay or natural fibre, they need to be evaluated by means of a methodology that includes all of the steps of the life cycle: to this end, life cycle assessment (LCA) which is an internationally standardized method, is commonly applied.

This paper combines the insight gained by Pietrini et al. [5] and Roes et al. [6], who both studied the environmental effect of nanofillers in polymers. Pietrini et al. studied a biodegradable polymer (poly(3-hydroxybutyrate) (PHB), filled with nanoclay and natural fibre), while Roes et al. studied a petrochemical polymer (polypropylene (PP), filled with nanoclay). Earlier work on LCA of nanocomposites was done by Lloyd and Lave [7], who studied automotive panels made from nanocomposite. Lloyd and Lave used steel and aluminum car panels as a reference, while we focused on PP panels. Both studies use Ashby material indices [8] to estimate the material savings by using materials with better mechanical properties. However, the values we use for the Young modulus (required for Ashby's material indices), originate from experimental data, while Lloyd and Lave used an idealized model from Brune and Bicerano [9] that is subject to substantial uncertainty by assuming perfect adhesion between the clay platelets and the matrix. For our analysis, we used LCA databases (e.g., Ecoinvent) that are based on measured inputs and emissions of discrete industrial processes (e.g., per kg or MJ of product). Lloyd and Lave, in contrast, applied a hybrid method which derives environmental impacts from the economic output of the respective sector or subsector and is hence less accurate.

2. Methodology

2.1 LCA fundamentals

As indicated in the ISO-14040 series, the life cycle assessment methodology distinguishes four phases: goal and scope definition, inventory analysis, impact assessment, and interpretation [10].

Goal and scope definition: LCA application, type, reason and audience are stated, together with the geographical and temporal scope. The system boundaries and functional unit (FU) are also determined.

Inventory analysis (LCI): the process is modelled in a flowchart. The inputs (energy, materials) and the environmental releases of each process phase are determined, in relation to the functional unit.

Impact assessment (LCIA): on the base of LCI, the potential impacts related to the functional unit are determined. The results can be grouped and weighted.

Interpretation: the data obtained in previous steps of LCA are discussed and conclusions are drawn.

2.2 Goal and scope definition

The purpose of the LCA studies discussed in this paper was to investigate whether filling polymers with nanoclay or natural fibre has environmental benefits compared to unfilled, or conventionally filled polymers. To this end, four products have been studied: packaging film, agricultural film, automotive panels, and a cathode ray tube (CRT) monitor housing. Table 1 lists the materials that were assumed for the different products. Among the various nanoclays, we chose organophilic montmorillonite (OMMT). As natural fibre, we chose sugar cane bagasse (SCB).

Table 1: Materials used for the different product alternatives.

Product	Conventional material	Alternative material
Packaging film	PP	PP-OMMT composite
Agricultural film	LDPE	PP-OMMT composite
CRT monitor housing	HIPS	PHB-OMMT composite PHB-SCB composite
Automotive panels	PP-glass fibre composite	PP-OMMT composite PHB-OMMT composite PHB-SCB composite

(PP = polypropylene, OMMT = Organophilic montmorillonite, LDPE = Low-density polyethylene, HIPS = High-impact polystyrene, PHB = Poly(3-hydroxybutyrate), SCB = Sugar cane bagasse)

In full-scale LCA studies numerous impact categories are taken into account, among them eutrophication, acidification, ozone layer depletion, ecotoxicity and human toxicity [11]. In contrast, only two environmental indicators are considered in the present study, namely 'non-renewable energy use (NREU)' and 'global warming potential in the next 100 years (GWP100)'. This choice was made due to a general lack of data on the PHB production process, which is in an early stage of development. Large-scale facilities do not yet exist and as a result, data related to other impacts were not available for our analysis. Next to NREU and GWP100, Roes et al. [6] considered also the categories abiotic depletion, ozone layer depletion, photochemical oxidant formation, eutrophication and acidification for the PP-OMMT products. These results are not discussed in this paper to keep the results uniform. While our environmental analysis is therefore bound to be incomplete, it can provide a useful first approximation of the environmental impacts. Given the findings of

Huijbregts et al. [12], the use of NREU can be generally considered a good indicator of the environmental performance of products and processes, unless the release of toxic compounds is meaningful. Regarding the technology status and the temporal and geographical scope, the assumed polymer production and processing technologies are representative for today's industrial facilities in Europe.

2.3 Functional units

We estimate the material requirements and the attendant non-renewable energy use for the following functional units (FU):

- *Packaging film:* the amount of packaging film needed for 1000 bags of 200 g "Fruitfante" candies produced by Schuttelaar B.V. (Waddinxsveen, The Netherlands). The function of these bags is to provide sufficient physical protection and to act as a barrier in order to preserve the candies. Pure PP is the conventional material used to produce such a bag. The weight of the conventional packaging material required for the functional unit is 3.66 kg.
- *Agricultural film:* the amount of plastic film needed to cover a standard tomato greenhouse with a volume of approximately 650 m^3. The purpose of this film is to provide thermal insulation and UV stability, combined with the necessary mechanical strength. We considered low-density polyethylene (LDPE) to be the conventional material, with a total surface of 431 m^2 and a weight of 2.38 t.
- *CRT monitor housing:* one housing of an average 17" CRT monitor for a desktop computer; the conventional housing is made of HIPS, with a weight of 2.2 kg [13].
- *Internal car panels:* the total of all internal panels of one average car with a lifetime of 10 years and a total distance travelled of 150,000 km; according to the Association of Plastics Manufacturers (PlasticsEurope) the weight of the conventional internal panels is 20 kg for an average car with a total weight of 1130 kg [14]; the composition of the panels is assumed to be 63 wt-% PP, 30 wt-% glass fibres (GF) and 7 wt-% maleic anhydride grafted polypropylene (MAPP) as compatibilizer between the PP matrix and GF [15].

2.4 Weight saving by using nanoclay- and SCB composites

As said in the introduction, improved material properties can be expected to allow a reduction in material use (unless there are other overriding limitations, for example concerning handling of the products). Ashby [8] developed a generic approach in order to determine the performance of a material in a given function. The performance of the material is expressed by Ashby's material index (MI) which is a function of material properties like the Young modulus (E), tensile strength (σ_f) and density (ρ). Ashby developed equations for MI for different functional requirements, such as strength limitation or stiffness limitation. The larger the value for MI is, the better the mechanical properties of the product under consideration are and the less material is hence required to fulfil a given function. The material indices developed by Ashby are based on design that is 'stiffness-limited', 'strength-limited', 'vibration-limited', 'damage-tolerant', 'electro-mechanical' or 'thermal and thermo-mechanical'. Within each category, the appropriate function and constraints are chosen (functions could be e.g. 'tie', 'shaft', 'beam', 'column', 'panel' or 'plate'; constraints could be e.g. required specification of dimensions).

For packaging film and agricultural film, we chose the formula:

$$\mathrm{MI}_{\mathrm{tie}} = \sigma_f / \rho \tag{1}$$

thereby assuming strength-limited design (the material should not tear) and that the function is represented by the Ashby category 'tie'. 'Tie' represents a function where the force works parallel to the surface of the film, causing it to tear apart.

For CRT monitor housing and automotive panels, we chose the formula:

$$MI_{panel} = E^{1/3} / \rho \tag{2}$$

thereby assuming that the design is stiffness-limited and that it is represented by the Ashby category ‘panel’ (when manufacturing monitor housings and automotive panels, the design will determine the material use to a large extent. However, assuming equal design (shape), the stiffness of the material will determine whether the material will not bend during use, hence keeping the product suitable for its function. Therefore, we estimate material use with a material index representing ‘stiffness-limited design’. Since bending comes before breaking, we choose as limitation ‘stiffness’ and not ‘strength’).

By applying the MI to two materials with different properties (e.g. a conventional versus a new, innovative material) it is possible to calculate the weight difference between the two end products, which have the same functionality, as shown in Eq. 3:

$$\text{Change of weight (\%)} = \left(\frac{MI_{conv}}{MI_{new}} - 1\right) \bullet 100 \qquad (3)$$

With Ashby’s method, materials are compared exclusively on the basis of their mechanical properties. With respect to packaging film, however, this is not the only aspect that must be taken into account. The barrier properties of this material are of major importance because the food that it envelops must be preserved. In particular, the oxygen transmission rate (OTR), representing an indicator for the oxygen permeability, should be minimized. We estimated the material reduction due to the use of nanocomposite film by assuming that the thickness of the film is proportional to the OTR (the higher the OTR, the thicker the film must be). We found a weight reduction that was much higher than the weight reduction based on mechanical properties. This means that the mechanical properties are the limiting factor and therefore the OTR is of no relevance.

Table 2 shows the mechanical properties and the material indices of the various materials used in this study. As shown in Table 2, all PHB based composites show a lower Young modulus compared to conventional materials. The modulus of the PP-GF composite is 3 to 4 times larger than the modulus of the PHB composites. In addition, PHB has a high density, which is also to its disadvantage. Therefore, despite the relatively high density of PP-GF (1.40 t/m^3 as compared to 1.22-1.10 t/m^3 for the PHB composites), the MI$_{panel}$ of PP-GF (1.28 GPa$^{1/3}$ m^{3}t^{-1}) is still the highest one among all the polymers considered. The PP-5OMMT shows a lower modulus than PP-GF as well. However, here the reduced density nevertheless yields a higher MI$_{panel}$. The changes in weight, resulting from the new materials, are shown in Table 3. The lower MI$_{panel}$ of the PHB composites results in an increase in weight of the final product. In contrast, the use of PP-3OMMT for packaging film and agricultural film and PP-5OMMT

Table 2: Mechanical properties and material indices (MI) of the samples.

	ρ (t/m^3)	E (GPa)	σ_f (MPa)	MI$_{panel}$ (GPa$^{1/3}$m^{3}t^{-1})	MI$_{tie}$ (MPam^{3}t^{-1})
PP	0.91		33.5 [16]		36.8
LDPE	0.92		24.0 [18]		26.1
HIPS	1.05	2.00 [17]		1.20	
PP-GF	1.40	5.75 [17]		1.28	
PP-3OMMT	0.93		37.4 [16]		40.2
PP-5OMMT	0.94	1.80 [16,17]		1.29	
PHB-5OMMT	1.22	1.48 [19]		0.93	
PHB-10OMMT	1.23	1.71 [19]		0.97	
PHB-10SCB	1.15	1.69 [19]		1.04	
PHB-20SCB	1.10	1.73 [19]		1.09	

Table 3: Change of weight as a results of using the new materials (σ_f and OTR behind PP-3OMMT indicate on which parameter the calculation is based).

	Packaging film (%)	Agricultural film (%)	CRT monitor housing (%)	Internal car panels (%)
PP-3OMMT (σ_f)	-9	-36.5		
PP-3OMMT (OTR)	-17.5			
PP-5OMMT				-1.25
PHB-5OMMT			+27.9	+36.3
PHB-10OMMT			+23.4	+31.5
PHB-10SCB			+15.9	+23.5
PHB-20SCB			+9.9	+17.1

for automotive panels allows a reduction in weight according to the Ashby method. This is thanks to a higher MI_{tie}, which means lower material requirements.

Table 3 shows that using OTR as basis for weight-saving calculation for packaging film, results in a weight change of -17.5%. However, because its mechanical properties (σ_f) do not allow a weight saving of this magnitude, the maximum weight saving for packaging film is -9%. This value was used in further calculations of packaging film.

3. Inventory analysis

Fig. 1 shows in a simplified form the lifecycles of the products, made from PHB composites and PP nanocomposites. For the production of PP-nanocomposite, maleic anhydride-grafted polypropylene (MAPP) is used. This is a blend of maleic anhydride, styrene and peroxides and it improves the adhesion between PP and fillers (in this case montmorillonite). This blend is also used for PP-glass fibre composite. Detailed descriptions of all processes and an overview of all energy and material inputs in all the life cycle stages of the products can be found in Pietrini et al. [5] and Roes et al. [6]. Only in case of automotive panels, the use phase substantially influences the final results; part of the impacts, due to fuel combustion by a car, is assigned to the car panels. This is done by allocating to the panels a part of the impacts caused by fuel combustion throughout the entire life of a car (150.000 km). The allocation is performed on a weight basis, taking into account that fuel consumption varies with the weight of the car. The assumed weight of the car in this study is 1242 kg (including 1.6 passengers, which is an average). The weight of the conventional, PP-GF panels is 20 kg. Using the Sedan equation[1], we estimate the fraction of impacts allocated to the panels to be 0.012. We use a similar approach for the OMMT- and SCB-filled panels. For the CRT monitor housing, packaging film and agricultural film, there are no such considerations. For the production of PHB, two datasets were available: PHB1 and PHB2, both produced by fermentation. PHB1 concerns production, in which the sugar to feed the microorganisms is extracted from sugarcane. In case of PHB2, the sugar originates from cornstarch. [20].

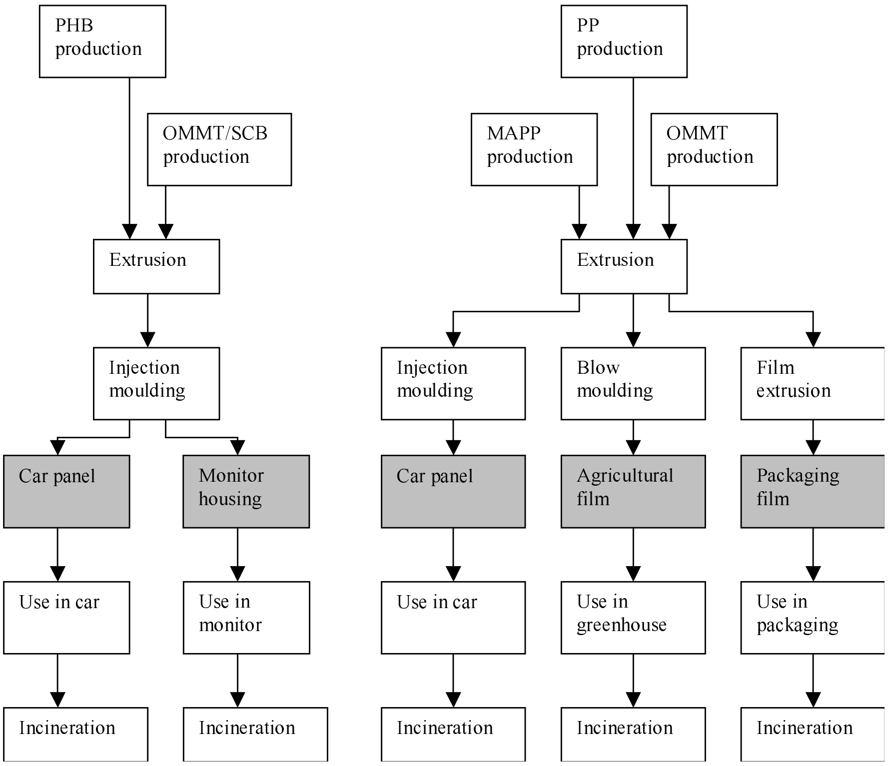

Fig. 1: Simplified flowcharts for the lifecycles of the products made from OMMT- and SCB-reinforced polymers (MAPP = maleic anhydride-grafted polypropylene).

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[1] $f_2 = f_1 \cdot \left(\frac{W_1}{W_2}\right)^{0.72}$, where f = car efficiency (km/kg fuel) and W = weight of the car.

4. Life cycle impact assessment

In Tables 4 and 5 and Figures 2-5 the results of the comparative LCA's of packaging film, agricultural film, CRT monitor housing and internal car panels are shown. In Figures 2-5, the results are displayed as indexed results, relative to the conventional material, because GWP100 and NREU have different units.

Fig. 2 shows clearly that there is hardly any difference in environmental impacts between pure PP packaging film and PP nanocomposite packaging film. For agricultural film (Fig. 3), however, the nanocomposite product has significantly lower impacts. In the case of CRT monitor housing (Fig. 4) all the PHB based composites score better compared to their conventional counterpart made of HIPS. Both NREU and GWP100 are substantially lower both for PHB produced from sugar cane (PHB1) and PHB derived from cornstarch (PHB2).

In particular, the PHB1 case shows very low values, as a consequence of the very low environmental impacts for the production process of this polymer. Best savings are observed for PHB1-10SCB composite. This sample shows a reduction for both indicators by about 99% compared to conventional HIPS housing. If compared with PHB-OMMT samples, PHB-SCB composites show better savings.

A very different picture is found for the use of PHB for internal car panels (Fig. 5). Most of the samples show clearly higher impacts both for NREU and GWP100. Only PHB1-20SCB seems to have little lower impacts for both NREU and GWP100 than the conventional product. The use of PP-5OMMT for the production of car panels leads to very similar environmental impacts compared to PP-GF.

5. Life Cycle Interpretation

5.1 Life cycle interpretation: discussion

The main reasons for differences in life cycle impacts between conventional products, SCB-composites and nanocomposites are 1) the production of the nanoparticles in the composites, 2) the different environmental impacts per kg of polymer matrix production, or 3) the changed weight of the SCB- or nanocomposite products.

The production of the nanoclay particles used in nanocomposite products contributes only very little to the overall environmental impacts, especially for packaging film and agricultural film, due to the very low clay load of the nanocomposites. The fact that

Table 4: Impact results for packaging film and agricultural film, cradle-to-grave.

	Packaging film		Agricultural film	
	NREU (GJ/FU)	**GWP100 (kg CO$_2$-eq/FU)**	**NREU (GJ/FU)**	**GWP100 (kg CO$_2$-eq/FU)**
PP	0.283	15.9	-	-
LDPE	-	-	155.9	9242
PP-3OMMT	0.284	15.7	107.1	5642

Table 5: Impact results for CRT monitor housing and internal car panels, cradle-to-grave.[2]

	CRT monitor housing		Internal car panels	
	NREU (GJ/FU)	**GWP100 (kg CO$_2$-eq/FU)**	**NREU (GJ/FU)**	**GWP100 (kg CO$_2$-eq/FU)**
HIPS	0.200	15.15	-	-
PP-GF- Pietrini	-	-	8.16	569.9
PP-GF- Roes	-	-	8.23	569.3
PHB1-5OMMT	0.009	0.55	9.01	642.5
PHB1-10OMMT	0.023	1.27	8.84	627.2
PHB1-10SCB	0.001	0.15	8.10	578.8
PHB1-20SCB	0.008	0.48	7.74	552.1
PHB2-5OMMT	0.171	8.74	10.60	722.8
PHB2-10OMMT	0.171	8.77	10.29	700.6
PHB2-10SCB	0.140	7.19	9.46	647.7
PHB2-20SCB	0.125	6.41	8.89	610.2
PP-5OMMT			8.21	570.4

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[2] NREU and GWP100 of PP-GF are different in Roes et al. [6] and Pietrini et al. [5]. For NREU, Roes et al. used older, but more detailed data. For GWP100, Pietrini et al. estimated emissions based on carbon content of the polymer, while Roes et al. used PlasticsEurope data [21]. Pietrini et al. used an estimate for conventional and new material, because no inventory data on GWP100 were available for PHB.

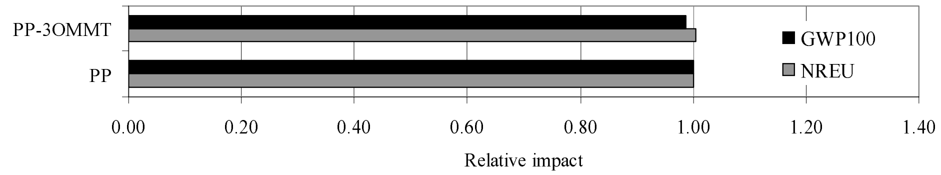

Fig. 2: Relative NREU and GWP100 of packaging film, cradle-to-grave.

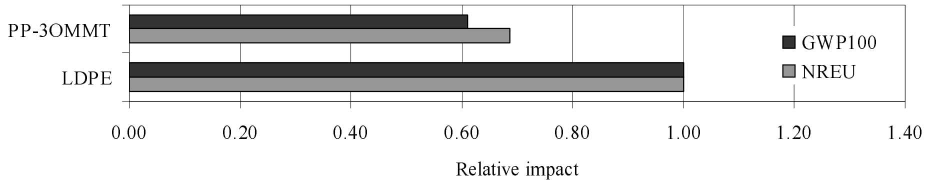

Fig. 3: Relative NREU and GWP100 of agricultural film, cradle-to-grave.

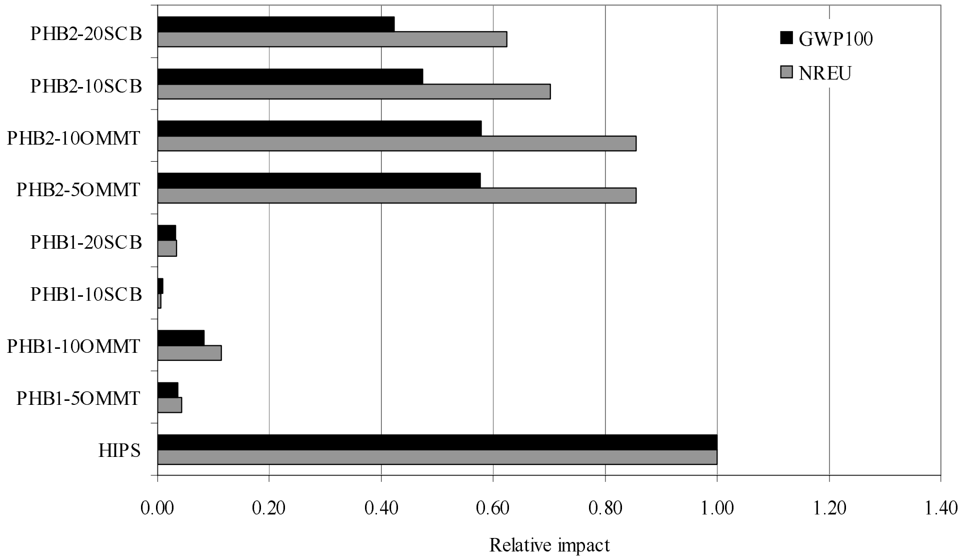

Fig. 4: Relative NREU and GWP100 of CRT monitor housings, cradle-to-grave.

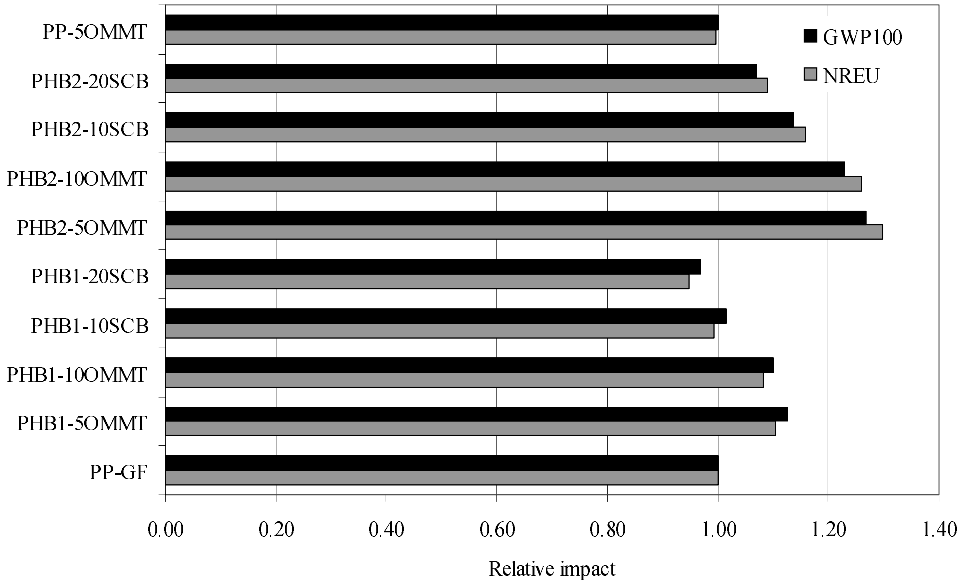

Fig. 5: Relative NREU and GWP100 of internal car panels, cradle-to-grave.[3]

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[3] Both Pietrini et al. [5] and Roes et al [6] calculated NREU and GWP_{100} of PP-GF. Due to different assumptions, they found different values. PP-5OMMT is relative to PP-GF from Roes et al. [6], while all the others are relative to PP-GF from Pietrini et al. [5] See table 5. The reason is that PP-5OMMT is calculated with the same assumptions as PP-GF from Roes et al., as opposed to all the values for PHB, which are calculated with the assumptions from Pietrini et al.

uncertainties in life cycle inventory (LCI) data of nanoclay production are high (they are based on estimates and pilot plant laboratory data) does therefore not impair the robustness of the results. The real impacts of nanoclay production are probably even lower than estimated in this study, because the energy use in the pilot plant (from which the data were derived) is most likely higher than in commercial-scale industrial plants. For the production of CRT monitor housing, the clay load is higher in the PHB-OMMT composites (5% and 10%). In combination with relatively lower impacts of the other steps in the process chain (compared to packaging film and agricultural film), nanoclay impacts are more pronounced here, as is shown in Fig. 6.

As mentioned above, the sugar used for the PHB fermentation process is produced from sugar cane (PHB1 case) or from corn starch (PHB2 case) [20]. In both cases, surplus biomass can be combusted for the production of electricity. This replaces the use of fossil fuels (e.g. coal and natural gas), leading to potentially substantial environmental credits. In the case of sugar cane, large amounts of biomass (the so-called bagasse) can be used for power generation. It is assumed that electricity is generated from the bagasse with an efficiency of 35%. This results in large credits and therefore in negative values calculated for the final impacts of PHB production in PHB1 case (for NREU -22.7 GJ/t (Fig. 6) and for GWP100 -3100 kgCO_2eq/t), making the total impacts very low. The lower quantity of surplus biomass available in corn starch offers relatively small savings, making this process less favourable from an environmental point of view.

Although the weight increases when using PHB composites in CRT monitor housing, their use still offers lower environmental impacts, thanks to the very low impacts of the PHB production process. For their use as automotive panels, this can, however, not compensate for the high fuel consumption and greenhouse gas emissions in the use phase that are highest for the product with the highest weight. Fig. 7 shows that the use phase represents the main part of the automotive panel's impacts. There are some uncertainties regarding the initial weight of the car and the panels, but the main message is that weight saving or weight increase of the panels has very significant influence on the environmental impacts, due to the high contribution of the use phase. If the use phase were not included, the PHB1 based internal car panels would have a NREU of only 1% (PHB1-10SGB) to 14% (PHB1-10OMMT) of the PP-GF

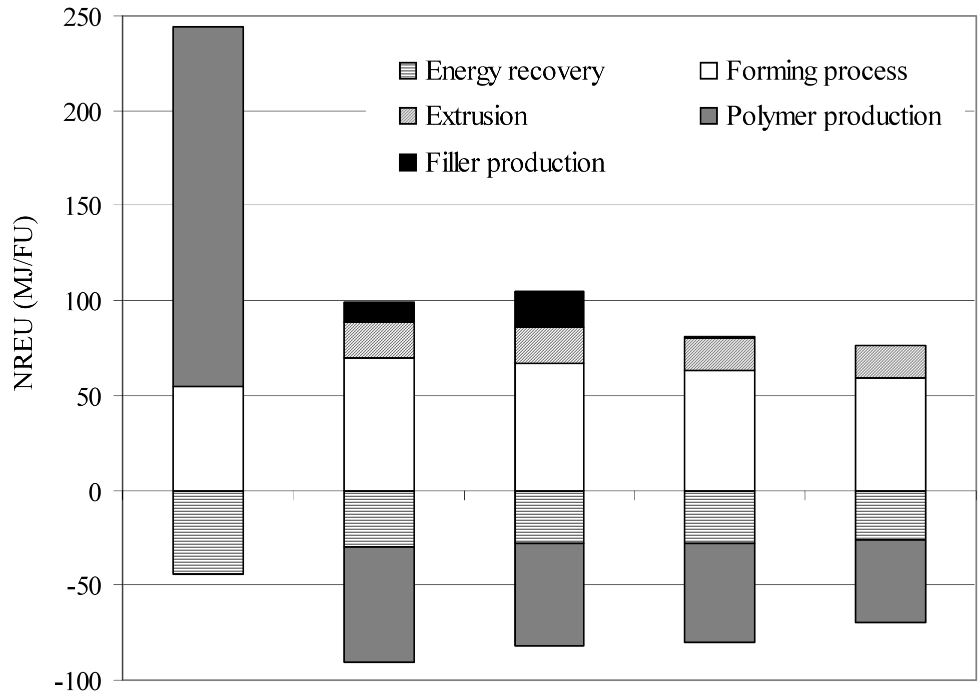

Fig. 6: Contribution of each production stage to NREU of CRT monitor housing for the PHB1 case.

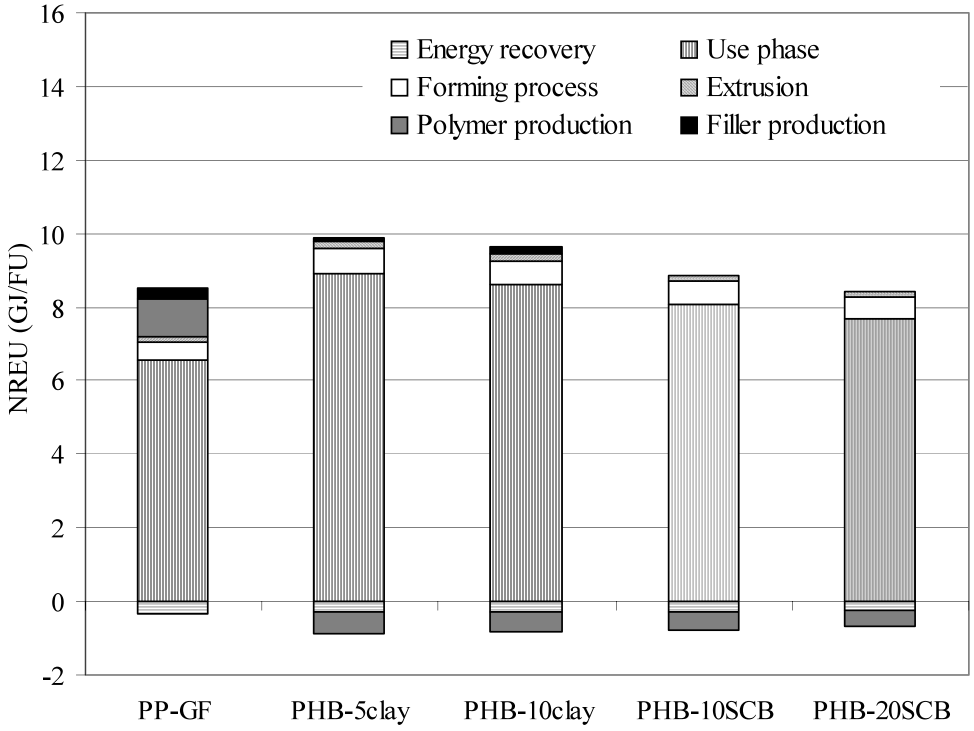

Fig. 7: Contribution of each production stage to NREU of internal car panels for the PHB1 case.

value.

In the case of agricultural film, the weight reduction of the nanocomposite, estimated from an improved tensile strength, is the primary reason for the observed lower NREU and GWP100.

When interpreting these results, it should, however, be considered that weight reduction has been estimated on the basis of material properties using Ashby's material indices and that these findings could differ from those obtained when performing a thorough study of real products. A further limitation is the exclusive analysis of non-renewable energy use and the greenhouse gas effect, while other aspects, such as ecotoxic effects of tallow-treated montmorillonite are not taken into account.

5.2 Life cycle interpretation: conclusions

A comparative environmental assessment has been performed in order to evaluate the possible environmental benefits of replacing conventional petrochemical plastics with PHB based composites, filled with SCB or OMMT, or PP based composites, filled with OMMT. The relatively low Young modulus and high density of PHB based composites, compared to conventional plastics, represent disadvantages for their environmental performance, but substantial environmental benefits can anyway be obtained, thanks to the very low NREU and GHG emissions from PHB production.

On a cradle-to-factory gate base, all PHB composites are superior in terms of NREU and GHG emissions compared to the conventional polymers for the two applications studied. When the analysis is extended to the system cradle-to-grave (including the use phase and post-consumer waste incineration with energy recovery), PHB composites score better only for CRT monitor housing. For the automotive application the weight of the functional unit becomes overriding, and only PHB-20SCB seems advantageous for both NREU and GWP100.

Regardless of the system boundaries, PHB-SCB composites show lower impacts than PHB-OMMT, and natural fibres seem the most promising filler in order to improve more the environmental performances of PHB composites.

The use of OMMT as filler in PP results in benefits for NREU and GHG emissions only in case of agricultural film. The primary reason is that LDPE, which is material for the conventional case, has a substantially lower Young modulus than the PP nanocomposite. As a result, substantial weight savings can be realized with PP nanocomposite when replacing LDPE in film applications.

Overall, we can conclude, that filling a polymer with nanoparticles *can* have environmental benefits, if it enables meaningful material savings compared to the conventional material that it replaces. This was in particular found to be the case for the agricultural film. The benefits observed for the PHB composites, however, are a result of the much more environmentally friendly biobased polymer, not a result of the use of a nanofiller.

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