



Recycling processes and quality of secondary materials: Food for thought for waste-management-oriented life cycle assessment studies

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1. Introduction

The challenges the waste industry is facing in order to address the ambitious targets set by the European Commission circular economy strategy are numerous, since at least 65% of municipal waste and 75% of packaging waste need to be recycled by the year 2030 (European Commission (EC), 2015). However, a “clean cycle” strategy to recycle as much as possible, while removing toxic or unwanted substances from the cycle should be established (Brunner, 2009; Velis and Brunner, 2014). The EU action plan for the circular economy also aims at promoting the use of recycled materials as a substitute of primary resources. However, the inherent properties of a material can be unfavourably affected by recycling processes (this is the so-called down-cycling phenomenon) and thus its marketability.

Life Cycle Assessment (LCA) is a widespread tool used in waste management in order to guide decision-makers towards optimal strategic choices. A key aspect of LCA studies on waste management is to account for the material and energy recovery and the related substitution effects, which substantially influence the study outcome (Laurent et al., 2014a).

In this paper the down-cycling phenomena for typical waste materials such as paper, plastics, wood and metals (aluminium and steel) are explained. Moreover, recommendations are given to enhance the modelling of the substitution of primary materials in waste-management-oriented LCA studies leading to improving the robustness of their conclusions and recommendations.

2. The down-cycling phenomenon

Technical properties of materials can be unfavourably affected by recycling processes (European Commission – Joint Research

Centre (EC-JRC), 2010 p. 359; Bartl, 2014; Geyer et al., 2015). This means, for example, that the secondary material (i.e. the material obtained from recycling) can replace the primary material only to a limited extent, i.e. in certain applications, after additional treatments, and/or for a limited time span. The qualitative degradation that certain materials undergo during the use and recycling stages may limit the number of cycles that they can afford. In other cases, a higher amount of the recycled material is necessary compared to virgin material to provide the same functionality. Furthermore, the secondary material might need to be mixed with primary material or with higher quality secondary material to meet the minimum technical specifications for its utilization (European Commission – Joint Research Centre (EC-JRC), 2010, p. 359). All these limitations imply that the quality of recycled materials is often lower compared to the corresponding primary materials. The following sub-sections describe the down-cycling phenomenon for some materials typically included in municipal waste management LCA studies, i.e. paper, plastics, metals (aluminium and steel) and wood, and for which we had knowledge at the time of writing. Indeed, the recommendations that are given at the end of the paper can be considered valid for any material, e.g. also glass and textile waste, even if not included in the following sub-sections.

2.1. Paper

There is a general agreement, based on laboratory studies, that fibres can be recycled 5–7 times on average (Pro Carton, 2016). This depends on the type of the original virgin fibre, its initial processing and its use in paper and cardboard products. In fact in the recycling process the fibres lose their resilience due to the progressive length reduction affecting the bonds between each other, and this implies the need of adding virgin fibres to guarantee the proper resistance of recycled paper and cardboard (Pro Carton, 2016). Also, according to Bajpai (2014), recycled fibres have lower strength and higher drainage resistance than virgin ones because of the loss of bonding capacity related to a reduced fibre swelling.

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Contamination can also contribute to the reduced strength of secondary fibre. In fact, the manufacturing of paper products requires the use of various chemicals either directly in pulp and paper production or in the following conversion processes (i.e. printing, gluing). With increasing recycling rates, this may imply accumulation or unintended spreading of chemical substances contained in paper products (Pivnenko et al., 2015a).

2.2. Plastics

The properties of recycled plastics typically differ from those of virgin plastics due to thermal, chemical, mechanical, and biological degradation (Kazemi Najafi, 2013), which may hinder their utilization. Furthermore, plastics consist of macro molecules (i.e. polymers), that can be affected by elevated temperature and mechanical treatments. This means that properties of recycled plastics are not consistent, but inherent loss of properties occurs at each recycling step, which limits the number of recycling cycles (Rajendran et al., 2012; Bartl, 2014). For instance, when polypropylene is recycled and processed several times, its molecular weight decreases and crystallinity increases. These opposing effects significantly influence the tensile strength and elongation, whereas the tensile modulus is affected to the lesser degree (Rajendran et al., 2012). Elamri et al. (2015) compared two polyethylene terephthalate (PET) polymers obtained from mineral water bottle with a virgin PET polymer and they found out that virgin PET showed better rheological and viscosimetric properties than the recycled PET polymers. Other unfavourable aspects of recycling processes are e.g. greyish colour and worse processing properties of recycled polymers, which are caused by limited sorting specificity and remaining content of additives, fillers, polymer cross contamination, non-polymer impurities and degradation (European Commission – Joint Research Centre (EC-JRC), 2010, p. 359; Pivnenko et al., 2015b). For instance, Oblak et al. (2015) underlined the worsening of the processability of high density polyethylene (HDPE) through the first 30 reprocessing cycles due to changes in mechanical and structural properties of the material.

One of the factors primarily influencing the quality of plastics recycling is the presence of additives (Pivnenko et al., 2015b). For example, the content of chromium in 48 analysed waste plastic samples showed potential spreading and accumulation of chemicals ending up in the waste plastics (Pivnenko et al., 2015b). Moreover, toxic additives such as bromated flame retardants (BFRs) included in one type of plastic products may subsequently be introduced into plastics used for other applications (Pivnenko et al., 2017), hence in the long term contaminating the whole material cycle.

In the bottle-to-bottle recycling process of PET, small amount of contaminants remain in the polymer and result in the need for a layer of virgin PET to protect the product (Bartl, 2014) or in a maximum admissible content of recycled PET in the new product (Ministero della Salute, 2010). With sophisticated decontamination processes (Welle, 2011), however, higher purities can be obtained but additional energy needs to be invested.

2.3. Metals

Metals are claimed to be infinitely recyclable without the loss of quality. In order to guarantee certain properties, however, alloying elements are often added to the pure metal to tailor its characteristics for a specific application. Furthermore, during the preparation of scrap material for recycling in a subsequent system, contamination with unwanted elements may occur. The mix of different alloy types and the presence of contaminants may reduce the material spectrum substituted by secondary materials. The “Metal Wheel” (UNEP, 2013) visualized the destination of different

elements in base-metal minerals and highlights the different base-metals characteristics. For example, it shows which elements can be recovered in subsequent processing, it points out the elements ending up in alloys or compounds not detrimental to the carrier metal, and it identifies detrimental substances for metal recycling.

According to the concentration of the alloying elements, aluminium alloys belong either to the wrought alloy category (alloy content up to 10 wt%) or to the cast alloy category (alloy content up to 20 wt%) (Paraskevas et al., 2015). Due to the strict requirements on alloy composition, contamination by alloying elements may constitute a problem in the recycling of aluminium. Two alternative reprocessing operations are used: remelting or refining. Remelting produces wrought alloys for rolled and extruded products, meanwhile refining produces cast alloys for shape-cast products and deoxidation aluminium (Cullen and Allwood, 2013). Mixed scrap streams contain a high variety of alloying elements, which prevent their recycling into a wrought product, therefore most of the mixed aluminium scrap is nowadays used to produce cast alloys, which act as a sink in the so-called aluminium cascade recycling (Paraskevas et al., 2013). While wrought alloys can be recycled into cast alloys, i.e. down-cycled to lower quality alloys, the reverse is unlikely (Cullen and Allwood, 2013). On the basis of chemical thermodynamics, Nakajima et al. (2010) quantitatively demonstrated the limit to the removal of impurity elements during the aluminium remelting process. Most of the impurities occurred as difficult to remove, except for elements such as magnesium and zinc. Another strategy to adjust the concentrations of contaminants to the desired target alloy is to dilute the scrap with primary aluminium. The quality of secondary aluminium is also affected by its oxidation level, e.g. estimated between 11% and 23% for aluminium from bottom ash above 0.8 mm (Biganzoli and Grosso, 2013).

Steel is produced in two different processes depending on the raw material used. While the blast furnace basic oxygen furnace route (BF-BOF) is used to produce primary steel from pig iron (with only a small scrap input, mostly from internal recycling), the electric arc furnace (EAF) uses 100% steel scrap. Post-consumer scrap is collected in different quality grades and with different content of tramp elements and mineral materials (Eurofer, 2008). Decisive for steel quality and therefore for the field of application is the concentration of tramp elements, such as copper and tin. As these elements are not volatile and nobler than iron, they cannot be separated from the liquid steel and are, therefore, critical for the recycling process (Reck and Graedel, 2012; von Gleich et al., 2004). Carbon steel quality in an EAF, i.e. produced from scrap, is therefore strongly influenced by the quality of the scrap input which, in turn, depends on the alloying elements and on the degree of material separation (Haupt et al., 2017a). The scrap mix used as an input in an EAF is mixed focusing on the targeted output quality. The mixing allows diluting the tramp elements from lower quality scrap grades with higher quality scrap such that the level of contamination with tramp elements reaches an acceptable level. This procedure leads to cascading for higher quality scrap grades and dilution losses in case of recycling of lower quality scrap. Once the targeted output quality is reached, 100% secondary material is used in the production of goods. Due to the accumulation of tramp elements, however, high quality products such as sheet metal for the automotive industry are often produced from primary steel (Nakamura et al., 2014).

2.4. Wood

The recycling of wood poses a number of challenges. Firstly, the structure of wood cannot be recreated once wood is mechanically or chemically processed (Werner et al., 2006), with wood fibres also being shortened during crushing or milling. Secondly, wood

degrades over time because of biological reasons (e.g. rot, mould). Thirdly, waste wood is often contaminated. The contamination can be mechanical (e.g. concrete, stones, nails), or chemical, such as in chemically preserved lumber (e.g. copper) (Bolin and Smith, 2011). Copper chrome arsenate (CCA) –treated wood has been banned from the European and American market at the beginning of the millennia, but due to its longer life span it still poses a health risk (Chen and Olsen, 2016), particularly when recycled. As illustrated by the challenges above, down-cycling of wood as a material is practically unavoidable.

A typical material recycling application is the production of particleboards. Particleboards utilising both recycled waste wood and virgin wood can be produced, and according to Rivela et al. (2006) a recycled wood content of up to 30% does not impact the material properties of the boards.

Waste wood can, however, also be recycled into new materials. According to Rautkoski et al. (2016) recycling of construction and demolition (C&D) wood waste into higher value added products is possible via pulping. Examples of products based on waste wood pulp are textile fibre yarns or foam formed panels. The authors found no major constraints in using contaminated C&D wood waste as a feedstock for pulping and concluded that from the point of view of the mechanical properties, such feedstock can substitute virgin woodchips in both chemical and mechanical pulping. Moreover, the quality of foam formed panels was not affected, but the textile fibre yarns were coarser than when virgin feedstock is used.

As presented in Sommerhuber et al. (2016), even cascading use of wood waste can eventually lead to high value added recyclable products, such as wood-polymer composites produced from particleboards previously made of C&D wood waste. The use of recycled wood might nevertheless negatively affect the strength of the composite. Moreover, the presence of metals (e.g. copper, chromium), which could be linked to an insufficient separation of wood waste for particleboard production, was identified. Höglmeier et al. (2013) argue that around one fourth of C&D wood waste can be re-used and another fourth can be recycled in other high-value applications via cascading recycling. However, the chemical contamination is also mentioned as a constraining issue.

3. Current modelling of recycling in LCA studies

Recycling is methodologically a case of multi-functionality in LCA, with the product to be recycled having two functions: firstly the function(s) the product was primarily made for, and secondly the function of providing secondary resources for use in subsequent life cycles/systems (European Commission – Joint Research Centre (EC-JRC), 2010, p. 343).

As reported in the review of 222 LCA studies of solid waste management systems by Laurent et al. (2014b), to address multi-functional processes, system expansion has mostly been applied (ca. 75%), while allocation has been exclusively used in about 4% of the reviewed LCA studies. The most used approach is called “substitution by system expansion” (European Commission – Joint Research Centre (EC-JRC), 2010, p. 76). It is also known as “avoided burden method” (Finnveden et al., 2009). One of the main challenges in this approach is to identify the replaced products. When avoiding primary materials, the differences in quality between primary and secondary materials should be taken into account in the modelling. Despite such concerns, the vast majority of waste management LCA studies have so far assumed a 1:1 substitution ratio of recycled to virgin materials (Gala et al., 2015).

Indeed, some researchers and LCA practitioners have introduced a coefficient in order to take into account the effects of down-cycling at the Point of Substitution (PoS), i.e. the exact point along the recycling chain where the material derived from waste

can be used in substitution of the corresponding primary material. In some cases, the coefficient is calculated starting from the quantitative evaluation of a technical property (Rigamonti et al., 2009, 2010), in some other cases the values chosen are just arbitrary values (Gentil et al., 2009; Di Maria et al., 2015), whereas in other cases it is based on the market-price ratio of the secondary material to the superseded primary material (European Commission – Joint Research Centre (EC-JRC), 2010, p. 360; Schrijvers et al., 2016a; Rigamonti et al., 2009; Allegrini et al., 2015; Koffler and Florin, 2013). This last approach was also discussed, in a more complex way, e.g. by Schrijvers et al. (2016b) and Zink et al. (2015). In general, the coefficient calculated considering the market-price ratio is based on economic considerations but at the same time it implicitly includes information about the quality of the waste-derived material. In fact, if the quality is very low, the market-price ratio will be close to zero. The challenge is that it can vary over time and geographical context and therefore the LCA has a limited temporal and geographical scope. Finally, to support the determination of substitution potentials related to resource recovery, Vadenbo et al. (2016) provide a structure for the systematic reporting of information and assumptions expected to contribute to the substitution potential in order to make substitution modelling and the results thereof more transparent and interpretable.

4. Recommendations

No matter the approach chosen to calculate the coefficient which quantifies the amount of primary material that can be replaced by one unit of waste-derived material, two relevant aspects need to be considered:

- The first recommendation is to perform the calculation at the PoS, i.e. the exact point along the recycling chain where the material derived from waste can be used in substitution of the corresponding primary material. To accurately consider the recycling yield and the losses in the collection and recycling processes before the PoS is reached, a detailed material flow analysis (MFA) is recommended (e.g. Haupt et al., 2017b; Niero and Olsen, 2016). When relevant, e.g. in the case of end-of-life durable goods the relevance of including temporal aspects in the MFA, should be considered, as shown in the case of the regionally-linked, dynamic material flow modelling tool for rolled, extruded and cast aluminium products developed by Bertram et al. (2017) and the mass flow model incorporating the dynamics in copper impurities in steel and the life-time of products and future steel demand presented by Daehn et al. (2017). For paper, the substitution is at the level of pulp, i.e. pulp from waste paper replaces virgin pulp. For plastics, the PoS is at the end of the recycling chain. In the case of PET, for example, the LCA analyst should model all the recycling chain, inclusive of sorting into the different polymers, grinding, washing, drying and the possible extrusion process (as performed by Rigamonti et al., 2010). For aluminium, while comparing the features of the primary and the secondary material, Koffler and Florin (2013) pointed out the close correspondence between the recycling process of re-melting and alloying, and the production of primary alloys. In both processes additional alloy input(s) are added to an aluminium-rich raw material input to create a material output of desired specification. The PoS is therefore identified at the Al scrap level, since the aluminium scrap substitutes aluminium containing raw materials. On the contrary, since primary and secondary carbon steel are produced in two different processes, the output of the production, i.e. the liquid steel, offers the best PoS. Due to the large difference in input materials for the primary and the secondary

production of steel, setting the PoS at the steel scrap level is not meaningful. In addition, Haupt et al. (2017a) showed that the electricity demand in the recycling process at the electric arc furnace (EAF) depends on the scrap quality. This highlights the importance to consider the recycling process scrap-specifically and, therefore, the need to include the modelling of the recycling process in the examined system. Identifying the PoS for wood is challenging since wood can be used as feed-stock in multiple applications, including pulping, refining or sawing. In both pulping and refining the inherent properties of wood are altered in such a way that recycling of wood as material is not possible after these steps. However, final pulp products (e.g. paper, viscose fibres) can be recycled. On the other hand, waste-derived wood originating from the value chain of timber (i.e. sawing) can be used to substitute virgin wood in case it has not been chemically preserved, or otherwise contaminated. High quality wood waste can enter the production of wood products (e.g. particle boards). If of small dimensions (sawdust, fibres) it can be utilised in pulp production, subsequently producing paper or even textile products, when economically advantageous. Even though from the material recovery point of view the recycling of high quality wood waste into new timber products is a preferred route. Low quality wood waste is generally not suitable for recycling, but can be utilised in energy production.

- The second recommendation is to take into account for the quality of the waste derived material, i.e. to use a *quality factor* that considers to what extent the inherent properties of the material change during recycling. In the case of paper, as reported in Rigamonti et al. (2009), the quality factor can be calculated from the possible number of recycling cycles, as suggested in ISO/TR 14049 (ISO, 2012). Assuming that paper can be recycled five times, 1 kg of secondary pulp replaces (1–1/6) kg of primary pulp, i.e. a quality factor of 0.833. In the case of PET, the quality factor can be calculated considering the intrinsic viscosity of recycled and virgin polymers. For example, Valentino (2017) calculated a quality factor equal to 0.9 when recycled PET flakes comes from bottles reprocessing. For metals, dilution losses need to be taken into account, as well as the possible replacement rate based on the metal quality at the PoS. During the production of secondary steel, for example, high tin and copper concentrations of low quality scraps are diluted with high quality scraps (Haupt et al. 2017a). The quality factor can be assumed equal to 1 if the PoS is liquid steel and as long as secondary liquid steel is used to produce the same product category that we can produce from primary materials. As soon as secondary liquid steel needs to be excluded from the product cycle because there is no use for it (due for example to exceeding copper or tin concentrations (Daehn et al. 2017)), the quality factor needs to be smaller than 1. For aluminium, dilution and quality losses need to be considered (Paraskevas et al., 2015), in analogy with the case of steel. In the case of aluminium cans, Niero and Olsen (2016) proved the benefits of including the actual alloy composition of the body and lid while modelling multiple life cycles. An ideal closed loop system over 30 loops was modelled by means of a MFA of Al and alloying elements. The results of the variation of the mass fraction of the four main alloying elements in the can body (i.e. Mn, Fe, Si, Cu) showed that using pure Used Beverage Can (UBC) scrap to produce new can body (i.e. 3004 alloy) would be possible only for a few loops. If only UBC scraps were used for an ideal can-to-can recycling indeed after 3 recycling loops the minimum Mn threshold would not be guaranteed anymore. When using different types of Al scrap, e.g. mixed aluminium packaging scrap, not only alloying elements need to be reintegrated, but also primary Al has to be added (Niero and Olsen, 2016). Therefore,

in both cases, the quality factor should be lower than 1. In the case of wood, the quality of waste wood greatly depends on its source. Wood is prone over time to biological degradation (Alakoski et al., 2016), making it often unsuitable for material recycling. However, in cases where waste wood can replace particulates of virgin wood, i.e. wood chips (pulp, particle board production), the quality coefficient could be calculated based on the level of its degradation, contamination and mechanical properties. For example Rigamonti et al. (2010) calculated the quality factor based on the different physical and mechanical properties (modulus of elasticity and longitudinal bending strength) of secondary particle board compared to virgin plywood, which resulted 0.6.

Acknowledgement

The work of Melanie Haupt was supported by the National Research Programme “Energy Turnaround” (NRP 70) of the Swiss National Science Foundation (SNSF). Further information on the National Research Programme can be found at www.nrp70.ch.

The work of Jáchym Judl was supported by the research program Material value chains – ARVI, funded by the Finnish Funding Agency for Innovation, Tekes, and by the research project “Sustainable, climate-neutral and resource-efficient forest-based bioeconomy”, funded by the Academy of Finland, Strategic Research Council (decision no 293380).

The work of Monia Niero was supported by the Carlsberg Foundation through the project “Absolute Circular Economy” (ACE) toolkit to support companies in the implementation of Circular Economy strategies from an Absolute environmental sustainability perspective”.

We acknowledge Luca Zampori and Kaisa Manninen for their help in the preparation of the manuscript.

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