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## 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 (Europen 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

https://doi.org/10.1016/j.wasman.2018.03.001 0956-053X/© 2018 Elsevier Ltd. All rights reserved. Centre (EC-IRC), 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 (Mnistero 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 basemetals 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

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