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Addressing the complex challenge of understanding and quantifying substitutability for recycled plastics

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ABSTRACT

Plastics are highly versatile materials, which can substantially vary in their properties, within a single polymer type, to cope with the range of different applications. Sorted bales of end-of-life plastics typically still contain a mixture of various plastic types and grades, which significantly affects the technical properties of the recycled material made from these bales. The evaluation of the technical recycling quality is crucial to determine which applications can be targeted with the produced recyclates, and if actual substitution of virgin plastics is possible. This is relevant from a technical perspective, but also for sustainability assessments such as life cycle assessment. The functionality of a plastic material is determined by a range of mechanical characteristics and by its ease of processing. These aspects cannot be generalized, as they are dependant on the intended application. This work, therefore, proposes a concept for calculating the effective technical substitutability, in which application specific functions generate a score for processability and mechanical recycling quality. To illustrate this concept, the substitutability of three commercially available recycled plastics is determined for four different application categories. The results show the strong application dependency of the changes in technical quality for a given recycled plastic stream. The approach is a step forward in the correct determination of the substitutability and consequently the proper assessment of the potential of end-of-life plastics as secondary resource.

1. Introduction

The abundance of types of plastics implies a high versatility in their properties, which allows their application in many sectors. The packaging sector is by far the largest single market in this respect, with around 40% of the European plastic converters demand (PlasticsEurope, 2019a). Due to the relatively short lifetime of such packaging products, these plastics make up 60% of the total collected post-consumer plastics (PlasticsEurope, 2019b). The current estimate of effective recycling rate (the amount that is actually converted to new pellets) for such end-of-life plastics is, for a country recognized as frontrunner such as the Netherlands, 26% (Brouwer et al., 2018). By 2030, however, Europe's target is 55% for the recycling of plastic packaging (European Commission, 2018), implying there is still a very large gap to fill. In addition to the packaging sector, major steps still need to be undertaken in the proper recycling of plastics coming from the building & construction, automotive and electrical & electronic equipment sectors (Buekens and Yang, 2014; Cardamone et al., 2021; Miller et al., 2014; PlasticsEurope, 2019a). The challenge to increase plastic recycling has been taken up by initiatives like the Circular Plastics Alliance, who have pledged to work towards the uptake of 10 million tonnes of recycled plastics in new products by 2025 (Circular Plastics Alliance, 2020).

These ambitious targets of course require that the recycled materials are suitable for substitution of virgin plastics or other raw materials (e.g., wood, metals). The effectiveness of plastic recycling is not solely a quantitative matter; much depends on the purity of the recycled streams (da Silva Müller Teixeira et al., 2021; Roithner and Rechberger, 2020). The difficulty here resides primarily in the fact that little is known about

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Abbreviations

HDPE	high-density polyethylene
HDT	heat deflection temperature
IM	injection moulding
ISO	International Organization for Standardization
IV	intrinsic viscosity
LCA	life cycle assessment
LDPE	low-density polyethylene
LLDPE	linear low-density polyethylene
MFI	melt flow index
MPO	mixed polyolefin fraction
NIR	near-infrared spectroscopy
PA-6	polyamide 6
PE	polyethylene
PET	polyethylene terephthalate
PP	polypropylene
rPE	recycled polyethylene grade
RQ	recycling quality
TDS	technical datasheet
VST	Vicat softening temperature

Symbols

a_c	Charpy (un)notched impact strength
a_i	Izod (un)notched impact strength
dl	decilitre
E	elastic modulus
E_f	flexural modulus
E_t	tensile modulus
f()	scoring function
F^{rec}	flow property of the recycled material
F^{vir}	flow property of the virgin material
g	gram
J	joule
kJ	kilojoule
m	metre
m_f	dart drop impact
MPa	megapascal
Pa	pascal
P_i^{rec}	mechanical property of the recycled material
P_i^{vir}	mechanical property of the virgin material
RQ^{mech}	mechanical recycling quality factor
RQ^{proc}	processability recycling quality factor
s	second
U^{rec}	physical resource potential
w_i	weight (importance) of the mechanical property Pi
$\alpha^{rec:vir}$	substitutability
γ	substitution potential
ε_b	tensile strain at break
η	melt viscosity
η^{rec}	resource recovery efficiency
π^{virgin}	market response
σ_f	flexural strength
σ_m	tensile strength
σ_y	yield strength

the exact impact of the level of impurity on the performance or quality of the secondary resources. Displacement of a primary material with a secondary resource is a complex interplay of various mechanisms (Zink et al., 2016). One-to-one displacement is rare, and is greatly distorted when the recycled materials have inferior technical properties (Zink et al., 2016). The potential of recycled materials to substitute primary raw materials is in whether or not they are interchangeable, in other words whether they can provide the desired functions for a given application (Vadenbo et al., 2017). This is not only important from a product design perspective, but also in life cycle assessment (LCA) studies the substitution of secondary resources is often poorly estimated, or no proper explanation is provided for the used estimation of substitutability. According to Viau et al. (2020) some LCAs are undermined by a lack of knowledge about the loss of quality, the functions and the end use of the particular secondary resource. Some efforts have been done already on improving this issue. The recent study of Eriksen et al. (2019), for example, examined the substitution potential of plastic waste streams by focusing on the quality of the original materials. However, this study relied on a partially subjective classification of high and low quality mainly grounded on a legislative categorisation. Therefore, the substitution potential of the recycled plastics is most likely overestimated or underestimated. The work of Rigamonti et al. (2020) proposed to base substitutability of secondary resources on their technical functionality. Here, the calculation of the technical functionality is based on a single main technical property. The work of Huysman et al. (2017) aimed at finding an indicative quality factor based on the compatibility (i.e., interfacial energy) which exist between different plastic types. Nevertheless, this quality is strongly linked to the complex composition of the waste stream (Alassali et al., 2020; Brouwer et al., 2020, 2018; Faraca and Astrup, 2019; Hahladakis and Iacovidou, 2018; Iacovidou et al., 2019). Yet, to date there is no uniform method for determining this technical functionality for end-of-life plastic fractions.

The difficulty in properly assessing the technical functionality of recycled plastics is related to their complex composition. Whilst plastic products are typically made from a single virgin plastic type and grade, the occurrence of an end-of-life plastic stream composed of a pure mono plastic is rare. Due to mis-sorting and the presence of multilayer materials, recycled plastics often include small amounts of at least one other polymer (Alassali et al., 2020; Brouwer et al., 2020; Chen et al., 2021; da Silva Müller Teixeira et al., 2021; Eriksen and Astrup, 2019; Kaiser et al., 2018; Niaounakis, 2020; Robert Dvorak, 2011; Strobl et al., 2021; Walker et al., 2020). Besides contamination by other polymer types, non-polymeric impurities (metals, paper, …) will also affect the technical quality of the recycled material. In addition, the usability of recycled plastics is highly application specific (Dahlbo et al., 2018).

First of all, the recycled plastic must have the necessary mechanical properties for the final product to meet its specifications. Mechanical properties is a general term which covers a vast number of material characteristics such as stiffness, ductility, toughness, strength and hardness (Callister and Rethwisch, 2014). Whether the value should be high or low, as well as the extent of importance of each property, depends on the intended application. For example, a packaging film requires a low value for stiffness, while a pipe requires a high value. The fact that both high and low values for a parameter can be more favourable for the potential of a raw material, and that this highly depends on the intended application, is an important attribute that should be clear when studying the circular economy of plastics. Secondly, even when mechanical characteristics of the recycled plastic may match the product requirements, it must also be possible to effectively process the material into a new product. The suitability of a plastic to a certain processing technique (film blowing, injection moulding, etc.) is largely determined by its flow behaviour in the melt phase. Some techniques (like pipe extrusion) require a very viscous ('low') flow, while others (like injection moulding) require a very easy ('high') flow (Spalding and Chatterjee, 2016).

Much research in determining the quality of recycled plastics is focused on investigating the various contaminants present (Alassali et al., 2020; Camacho and Karlsson, 2000; Karlsson, 2004; Stangenberg

Table 1
Technical characteristics of plastics, including their physical quantities, units and ISO standards.

Technical characteristic	Description	Property [unit]	Standards
Mechanical			
Strength	The material's ability to withstand an applied load without failure (tensile strength) or plastic deformation (yield stress).	Tensile strength σ_m [MPa]	ISO 527
		Flexural strength σ_f [MPa]	ISO 178
		Yield strength σ_y [MPa]	ISO 527
Stiffness	The material's resistance to elastic deformation.	Elastic modulus E [MPa]	
		Tensile modulus E_t [MPa]	ISO 527
		Flexural modulus E_f [MPa]	ISO 178
Toughness	The total energy a material can absorb/dissipate before failure.	Surface under tensile curve [J/m^3]	ISO 527
Ductility	The total plastic deformation a material can withstand under loading before failure.	Tensile strain at break ε_b [%]	ISO 527
Impact strength	The material's ability to withstand a sudden load at a high deformation rate without failure	Charpy (un) notched impact strength a_c [kJ/m^2]	ISO 179
		Izod un(notched) impact strength a_i [kJ/m^2]	ISO 180
		Dart drop impact m_f [g]	ISO 7765
Processability			
Ease of flow	A polymer's melt viscosity is defined as the measure of the rate at which polymer chains can move relative to each other, at a certain temperature and shear rate (Gilbert, 2017).	Melt viscosity η [Pa•s]	ISO 3219
	The polymer's mass extruded under a fixed load over a specified time through a die of specified length and diameter at a fixed temperature.	Melt -Flow Index MFI [g/10 min]	ISO 1133
	Polymer specific (i.e., PET). The intrinsic viscosity is a measure of duration of a dissolved polymer to dilute through a specified capillary, relative to the pure solvent.	Intrinsic Viscosity IV [dl/g]	ISO 1628

et al., 2004; Strobl et al., 2021; Vilaplana and Karlsson, 2008; Wagner et al., 2020). However, it is of paramount importance to be able to make a proper assessment of the loss in technical functionality of recycled plastics in order to determine the best possible circular route for this important material.

In this manuscript we focus in more detail on the complex determination of substitutability of recycled plastics, based on their technical functionality. For this purpose, we compiled literature values for reprocessed end-of-life plastics for the properties elastic modulus, tensile strength, elongation at break and flow during processing. An explanation on the diversity in required technical performance is given. Furthermore, this work provides a concept for calculating the substitutability of a certain recycled plastic. For this purpose, two recycling quality (RQ) factors are used, one taking into account the processability and the other factor focusing on the mechanical requirements of the intended application.

2. Technical performance of end-of-life plastics

2.1. Technical properties of plastic materials

As a consequence of the wide range of applications for plastics, plastics are available in diverse sets of technical properties. Polymers are versatile materials, whose properties depend not just on their characteristic chemical formula, but also on their macromolecular architecture. During the polymerisation process, chains are produced with different lengths and varying degrees of molecular branching (Ashby and Jones, 2006; Spalding and Chatterjee, 2016), resulting in many different subtypes of a single polymer, commonly called 'grades'. The selection of the plastic grade with the proper set of mechanical and processing related properties relies mainly on the technical datasheets (TDS) provided per virgin grade. The technical quality of plastics is a result of mostly mechanical properties, typically complemented with a property that describes the flow behaviour of the melt phase. This is important regarding the choice of the appropriate processing technique and ease of processing. Table 1 summarises the principal mechanical characteristics as well as their related physical quantities, which are measured in standardized tests. Strength, stiffness, and impact strength are considered the predominant mechanical characteristics to describe overall material behaviour (Van Belle et al., 2020). Strength and stiffness are both measured in the tensile (or flexural) test, which plots the required stress levels to obtain a certain strain at relatively low deformation speeds. Strength is a material's ability to withstand stress without failure and is most often measured in tension. The material's strength can be described by different quantities. Tensile strength is the highest possible stress a material can handle without breaking, whereas yield strength describes the elastic limit. Applied stresses higher than yield stress will cause plastic deformation in the material, which is irreversible. Imagine tightening a bottle cap, but the force applied deforms the bottle, making it unusable. Stiffness is an expression of rigidity; it is the resistance to elastic deformation. As this describes the initial material response (from unloaded to stressed), it is an extremely important property to describe the behaviour of materials and products in their use phase. Stiffness on the other hand is expressed by the elastic modulus E, which is measured in either tension or under flexure. Some products such as bottles must have a degree of rigidity (high modulus value), while other applications (such as film products) must be flexible (low modulus value). Accordingly, a sufficiently high modulus will prevent a plastic chair from sagging under the weight.

Other properties taken from the tensile (or flexural) test are ductility and toughness. Ductility indicates the ability of a material to undergo plastic deformation when subjected to a load. For example, most plastic films will plastically deform upon being stretched, without instantly breaking. Ductility is approximated by the strain at break, which in fact includes not just the plastic strain but also the elastic strain. Materials with low ductility are called brittle. Toughness is a material's ability to convert energy into (any type of) deformation without breaking. It is determined by the surface area under a stress-strain curve.

In practice, however, it is often relevant to know as well the material behaviour at high deformation speeds, which is equivalent to sudden loads, like when a product is dropped. This property is the impact strength, which is measured in either an Izod or Charpy test. Impact strength and toughness typically follow the same trend, which often causes their terminology to be confused (Ragaert et al., 2020). Impact strength measurements require a sample with several millimetres thickness, which is why the method is unsuitable for films. Film materials will typically make use of the so-called dart drop test instead. This involves dropping a dart from a standard height and examining the weight of the dart at which a stretched film fails. All these properties are determined through standardised testing methods. The last column in Table 1 shows the specific standards according to ISO.

Besides these main mechanical characteristics, other properties include hardness, tear resistance, thermal and electrical properties. The

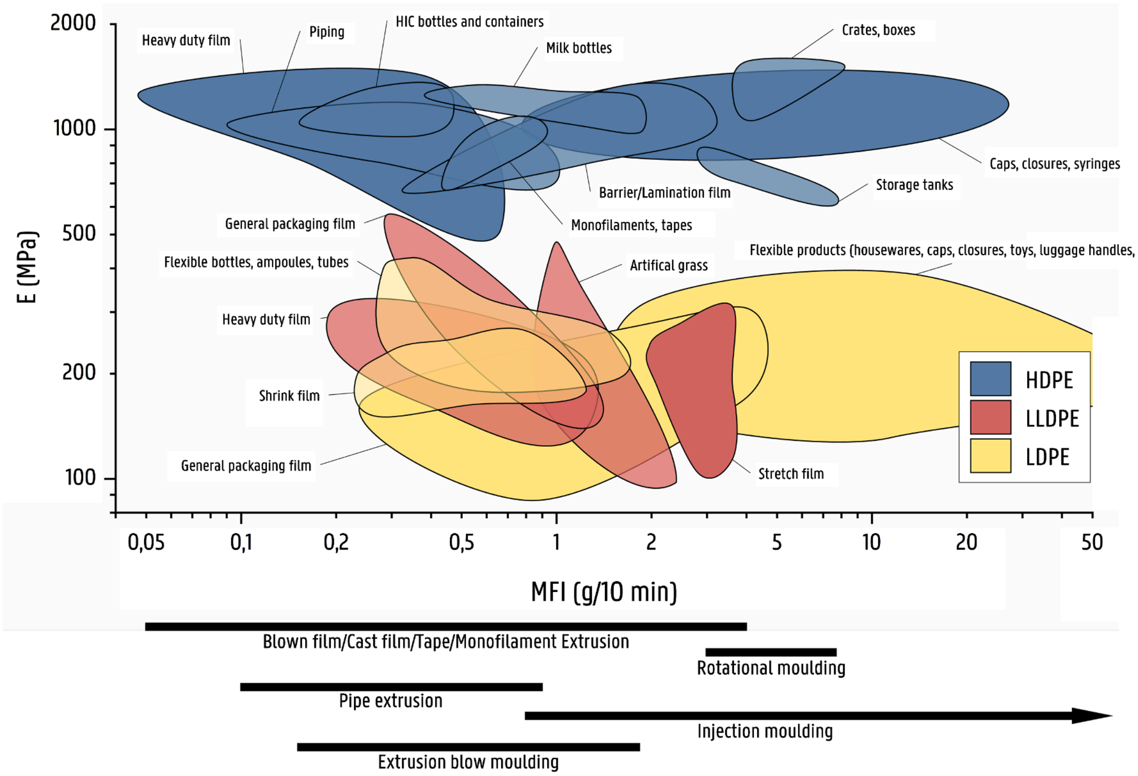

Fig. 1. Mapping of the elastic modulus and MFI for commercially available PE grades for a selection of typical applications. More information on the data and sources used for this figure can be found in Supplementary information (Table S 1).

mechanical durability of a plastic material is also largely determined by the environment in which the intended application is to be used. Furthermore, the time and temperature dependence of the mechanical functions must be considered. Certain applications demand that plastics fulfil minimal values for very specific properties. For instance, polymer grades suitable for piping must meet the requirements for creep or long-term loadings. In some cases (e.g., housings of electric equipment), plastics may not exhibit deformation at high temperatures, and consequently adequate values for Heat Deflection Temperature (HDT) or Vicat softening temperature (VST) must be met (Van Krevelen and te Nijenhuis, 2009). In other applications, plastics must also be able to endure cold temperatures, e.g., plastic boxes intended for freezer storage must be sufficiently impact resistant at these low temperatures. The long-term storage of chemical products requires that detergent bottles must comply with certain values for their resistance to Environmental Stress-Cracking (Hayes et al., 2015; Robeson, 2013).

In addition to the mechanical properties, the TDS provides information about the processability of the respective polymer grade by including the flow behaviour. Typically, this is summarized by the melt flow index (MFI), which describes how much mass of the molten material will flow through a capillary opening in 10 min, when a certain load is applied at a certain temperature. A few polymers, PET foremost amongst them, use the measurement of intrinsic viscosity (IV) instead, which is a solvent-based technique. In general, high values mean a 'good' ('high') flow and low values indicate a very viscous melt phase with 'poor' ('low') flow. The intended application, and more specifically the polymer processing technique used, will determine whether a high or low flow is required.

To highlight the relevance of the mentioned technical parameters, we give the example how important and different they can be, even within one polymer type. Fig. 1 gives an overview of different PE grades in use and their applicability along with typical values for the related key mechanical property modulus and the key processing property MFI. PE is the world's most-used polymer of which the predominant types are low-density polyethylene (LDPE), linear low-density polyethylene (LLDPE) and high-density polyethylene (HDPE) (Statista, 2017). Fig. 1 shows the enormous range in technical properties and the association with their molecular architecture and applied mechanical processing. For example, it is clear that film and bottle grades require a low MFI, and in addition this MFI value must lie within a narrow range. This is in contrast to the large MFI window for injection moulded products, such as caps, closures, … Depending on the desired heavy-duty film, HDPE or LLDPE grades can be applied, and the modulus can vary from 130 to 300 MPa (LLDPE) and 500–1350 MPa (HDPE). HDPE milk bottles, on the other hand, require a modulus value in the region of 1200 MPa.

In fact the diversity in commercially available grades per polymer type is many times higher than what can be detailed in Fig. 1 due to (i) continuous technological evolution in reactor design, (ii) varying reaction conditions (temperature, pressure, time) (iii) use of different types of catalysts and (iv) introduction of other monomers (Karger-Kocsis and Bárány, 2006; Spalding and Chatterjee, 2016). Each of these distinct plastic grades is characterised by their molecular architecture and related structure-property relationships (Spalding and Chatterjee, 2016).

2.2. *The complexity of recycled plastics technical quality*

Waste streams do not come with a standard TDS per recycled polymer type. The generally complex composition of the recycled material renders an estimation of the different technical properties difficult. In recycling, the different grades of a single polymer type usually end up in the same material stream as current separation techniques (either sensor based techniques such as NIR or density separation) do not really differentiate between the various grades of the same polymer type (Chen et al., 2021; Vyncke et al., 2020). In addition, the presence of hard to separate products (e.g., multilayer materials) and sorting errors cause the inclusion of other types of plastics and non-polymeric impurities (Berwald et al., 2021; Horodytska et al., 2018; Kaiser et al., 2018;

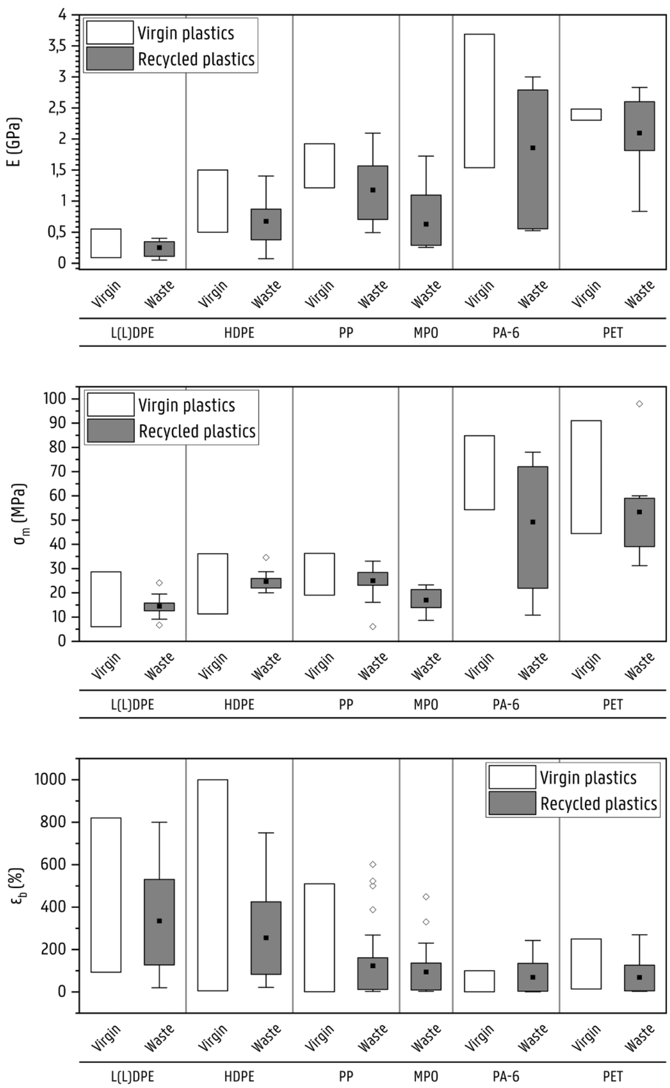

Fig. 2. Mechanical properties (E modulus, Tensile strength, and Elongation at break) of virgin and recycled plastics (references in Supplementary Information (Table S 2)). The mean values for the recycled plastics are indicated (■).

Kleinhans et al., 2021). The uncertainty about the technical functionality of recycled plastics causes a considerable hurdle for polymer processors to use secondary plastics in their products. The lack of systematic insight in the technical quality of recycled plastics prevents knowing their potential to substitute their virgin counterparts.

Fig. 2 and Fig. 3 respectively summarize an inventory of the available data on average key mechanical properties (tensile properties; elastic modulus, tensile strength, and strain-at-break) and processing related MFI/IV values of both the virgin plastics and the corresponding recycled end-of-life plastics. Values for virgin polymers are averaged from existing materials databases (MatWeb, 2021; SpecialChem, 2021), while values for the recycled polymers are averaged from what can be found in literature, as no databases are available for recycled plastics.

The number of references used to calculate the average values for these end-of-life plastics are indicated between brackets: L(L)DPE (14), HDPE (18), PP (33), MPO (11), PET (20) and PA-6 (7). An overview of the data and reference list can be found in Supplementary information (Table S 2). LDPE and LLDPE were grouped together in Fig. 2 and Fig. 3, as papers mentioning the properties of end-of-life plastics make little or no distinction between these two PE types. LDPE and LLDPE are commonly blended during processing (Spalding and Chatterjee, 2016; Utracki and Wilkie, 2014), but also in waste streams these plastics are sorted to the same fraction (Kleinhans et al., 2021). The mixed polyolefin fraction (MPO) is a widespread recycled fraction due to the mixed collection and difficult separation of L(L)DPE, HDPE, and PP. In broad terms, MPO can be divided into flexible MPO and rigid MPO, as, for example, in the work of Luijsterburg and Goossens (Luijsterburg and Goossens, 2014) and Hubo et al. (Hubo et al., 2016).

Looking at the mean values, the mechanical properties of the recycled plastics appear to be reasonably close to that of the virgin plastic counterpart, mostly being a bit lower. The mean values for the elastic moduli for the recycled plastics HDPE, PP and PET are respectively 14, 3 and 9% lower compared to the minimal value of the virgin counterparts. A reduced stiffness has also been observed for the MPO materials compared to the minimum values for elastic modulus of virgin HDPE (20%) and PP (48%) homopolymer grades. However, the presented Fig. 2 does not distinguish between flexible and rigid MPO fractions. If L(L)DPE plastics are present in the MPO stream, this will result in a decrease of the E moduli (Kolařík, 1998; Strapasson et al., 2005; Yin et al., 2015). The minimum values found in the literature for E modulus are for all the considered recycled plastics lower than those for the virgin counterparts: L(L)DPE (81%), HDPE (91%), PP (59%), MPO (HDPE \67%; PP\79%), PA-6 (66%) and PET (64%). This indicates that these recycled plastics are undergoing a significant reduction in stiffness. However, higher maximum values in comparison with the maximum values of the virgin polymers can also be found for PP and PET. This increase in modulus may be due to the presence of contaminants (e.g., metal and wood particles) with a higher stiffness. Besides these contaminants, degradation of the polymer chains can increase stiffness values (Aurrekoetxea et al., 2001; Del Mar Castro López et al., 2014; Jmal et al., 2018; Mantia and Vinci, 1994).

The mean tensile strength values for the recycled polyolefins L(L)DPE, HDPE and PP are well between the limits defined for the virgin homopolymers. The lowest literature values found for the tensile strength of the recycled plastics PP, PA-6 and PET are clearly lower than the minimum values for the virgin counterparts, respectively 15, 80 and 30%. The minimum values for MPO fractions are also significantly lower compared to HDPE (57%) and PP (55%) homopolymers. This may be attributed to the incompatibility that arises between PP and different types of PE during reprocessing, resulting in poor adhesion between PP and PE (Chaffin et al., 2000; Jordan et al., 2018; Lo et al., 2004; Poon et al., 2004; Song et al., 2012; Van Belle et al., 2020). Once again, this difference strongly depends on the composition of the MPO stream.

The mean values for the elongation at break of the recycled plastics are well within the defined range of the virgin analogues. It should be noted, however, that the majority of the reported literature data tends towards lower values of elongation at break. The presence of impurities in the recycled plastics can reduce the elongation at break. Also, for the elongation at break values of MPO mixtures, a negative impact is noticed. This can again be ascribed to the poor adhesion existing between PP and PE.

As already demonstrated by Fig. 1, preferred values for key mechanical properties are strongly related to the type of application. Unavoidably, this results in a very large spread on the different mechanical properties of the several plastic types, as can be clearly seen in Fig. 2. It is therefore impossible to make a generalised comparison between the virgin plastics and their recycled counterparts in terms of minimal requirements for a whole plastic type.

Similarly, a certain plastic's MFI is strongly application dependant, as already shown in Fig. 1. Injection moulding applications require high MFI values, whereas extrusion processes demand much lower values. Fig. 3 therefore shows wide spreads for the MFI parameter of the virgin materials. For the intrinsic viscosity (IV) values shown for recycled PET, it is obvious that these are substantially lower than values for virgin PET material. This reduction in IV is due to the chain degradation of the PET polymer (Badía et al., 2009). However, in bottle-to-bottle recycling, polycondensation steps are usually implemented in the recycling process to again increase the IV, as such compensating the degradation (Rastin

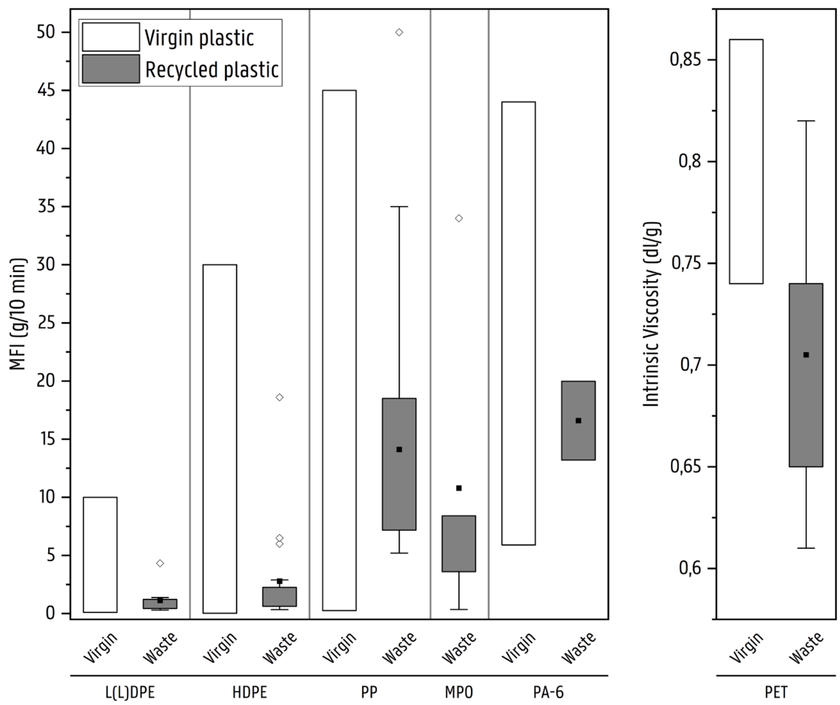

Fig. 3. MFI and IV values of virgin and recycled plastics (references in Supplementary Information (Table S 2)). The mean values for recycled plastics are indicated (■).

et al., 2016; Thoden van Velzen et al., 2016; Welle, 2011).

3. Scoring the recycling quality of end-of-life plastics

The wide variety of grades per plastic type makes it difficult to define one single reference value for each technical property per polymer type. A simple ‘the higher, the better’, is too limited as an approach and must be redefined to an appreciation of the degree to which the recycled plastic can truly substitute the full technical functionality of a relevant virgin plastic. A comparison of properties between recyclates and virgin plastics is very application-specific and more about matching than about high values.

In order to set up such a concept, our paper starts from the work of Vadenbo et al. (2017), in which a broader framework for substitution is presented. In this paper, the substitution potential γ, is based on four determining factors (Eq. (1)): (1) the physical resource potential U^{rec}, which is the percentage of the target plastic in a given waste stream, (2) the resource recovery efficiency η^{rec}, which is the yield of the applied waste management system (collection, sorting and pre-treatment), (3) the substitutability $\alpha^{rec:vir}$, which represents the functional performance of a recycled plastic compared to that of a virgin grade for a specific application, and (4) the market response π^{virgin}, which is the expected displacement of the virgin plastic by the recycled material. Note that an essential distinction has been made between the overall substitution potential γ and the more technical substitutability $\alpha^{rec:vir}$, i.e., the ability of a recycled plastic to match the functional properties of the virgin material.

$$\gamma = U^{rec} \cdot \eta^{rec} \cdot \alpha^{rec:vir} \cdot \pi^{vir} \tag{1}$$

Generally, U^{rec} being the plastic composition, and the resource recovery efficiency η^{rec} are given facts for converters, after the waste has undergone the first steps of a recycling chain, such as sorting. Recent research has focused on describing in detail the compositional analysis of complex plastic waste streams (Alassali et al., 2020; Brouwer et al., 2018; Eriksen et al., 2019; Eriksen and Astrup, 2019; Faraca and Astrup, 2019; Roosen et al., 2020; Strobl et al., 2021). In addition, studies have also been carried out regarding the efficiency of Material Recovery Facilities (Kleinhans et al., 2021; Mastellone et al., 2017) and even the modelling of their outputs (Kleinhans et al., 2021). This type of studies gives an insight in polymer types and non-polymer contaminants present.

The factor that is less well elaborated is the technical substitutability factor $\alpha^{rec:vir}$. Vadenbo et al. (2017) proposed to determine $\alpha^{rec:vir}$ as the ratio of the ‘functionality’ of the recycled resource to that of the displaced resource. Rigamonti et al. (2020) introduced further guidelines and based their calculation of substitutability on the technical functionality of the materials. Their approach relies on the identification of a single main technical property that characterizes or is required to fulfil the key-function of the specific application.

Currently, however, most proposals ignore or oversimplify the functionality of an intended application. As apart from product properties itself, also the required production technique is important, which in turn dictates the required processing property MFI (or IV). Furthermore, as discussed above, not all (mechanical) properties are always desired to be ‘high’. Especially for the properties modulus (resistance to elastic deformation) and MFI (inverse resistance to flow), it is needed that the recycled plastic behaves similar to the virgin plastic it would displace. As such, our concept is based on the degree the property is ‘different to’ rather than ‘less than’. Finally, strain at break is a property which can vary several orders of magnitude between a virgin and recycled plastic, which should not be allowed to have a disproportionate effect on the overall result.

We therefore propose an advanced calculation method of $\alpha^{rec:vir}$ which (i) takes into account the intended product application via the processability and (ii) adapts the appreciation of different mechanical properties to the specific nature of said properties and the target application. To this end, we introduce the processability recycling quality factor RQ^{proc} and the mechanical recycling quality factor RQ^{mech}. Each of these factors appreciates part of the technical substitutability regarding ease of processing and mechanical properties, respectively. In order to cope with the application dependency of the functionality, scoring functions must be defined for each of the relevant

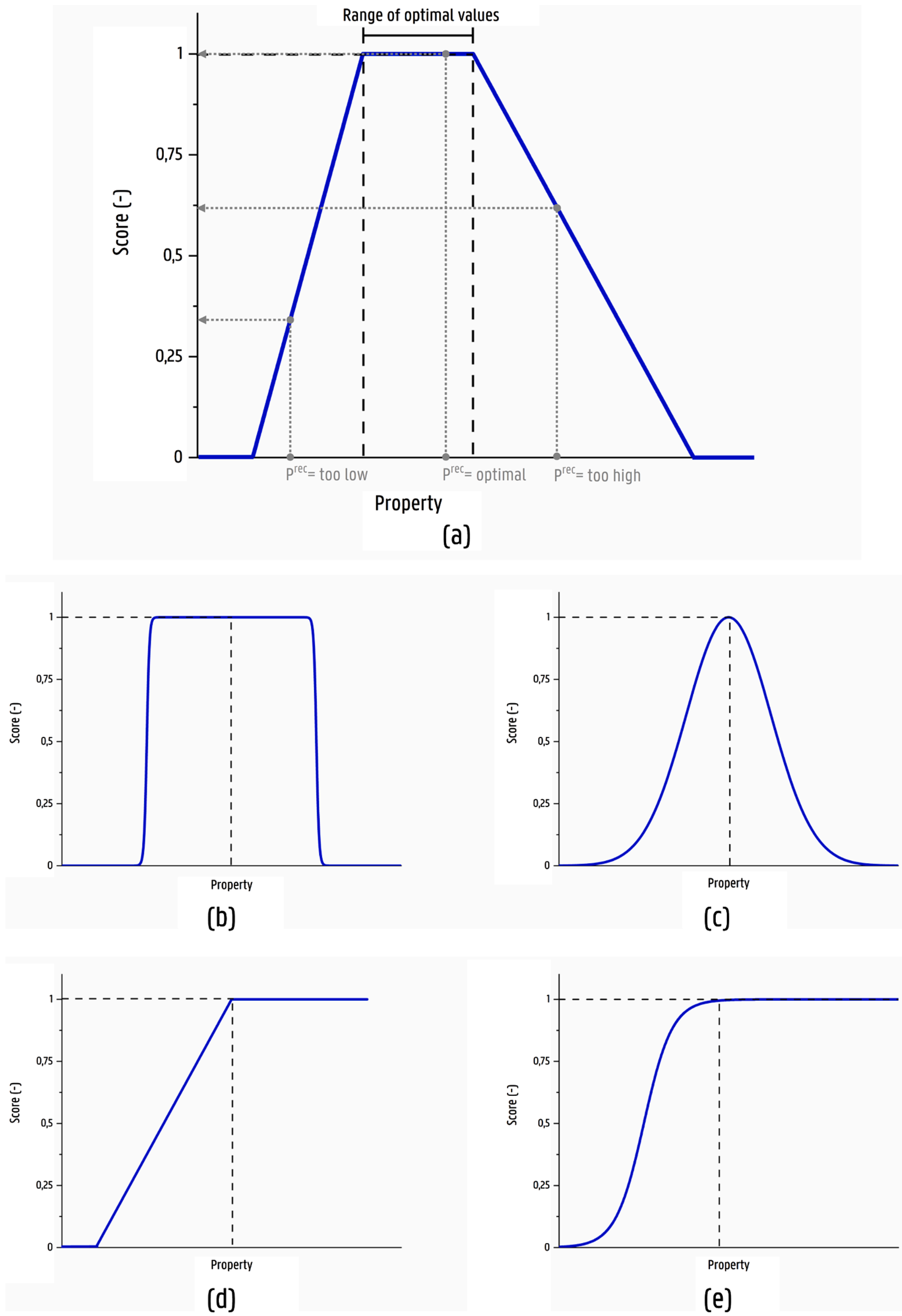

Fig. 4. Types of scoring functions (a) Trapezoidal (double) (b) Bell-shaped (c) Gaussian (d) Trapezoidal (single) (e) Sigmoidal.

properties per application or group of comparable applications (e.g., bottle, film, …). Depending on the severity of deviation of the recycled plastic's property to that of the application's typical virgin grades, each scoring function then returns a value between 0 and 1. Here, RQ^{proc} is defined by a scoring function f based on the relevant flow property F^{vir} (MFI or IV) of the virgin material as shown in Eq. (2).

$$RQ^{proc} = f\left(F^{vir},\ F^{rec}\right) \tag{2}$$

This scoring function should therefore generate a value close to 1 for recyclates whose MFI (or IV) value (F^{rec}) falls within the optimal processing window of the specific application. Conversely, recycled materials that are difficult to process for this application should be scored low, resulting in a low RQ^{proc}. Those impossible to process with the desired technology should be scored zero.

In turn, RQ^{mech} is calculated as a weighted summation of the values obtained via scoring functions f of the essential mechanical properties of the virgin material P_i^{vir} (Eq. (3)). Here, both the scoring functions f for each mechanical property P_i and the weights w_i have to be adapted accordingly to the application. As such, the scoring functions should reflect the degree in which a decrease (or increase) in a certain mechanical property is undesirable for a given application. Such a function will vary per property/application combination. For example, the application group 'rigid injection moulding' is quite tolerant towards a high modulus value, while 'flexible injection moulding' is not. In turn, the chosen weights w_i should indicate the importance of each mechanical property in the functionality of that particular application.

$$RQ^{mech} = \sum_{i=1}^{n} w_i \cdot f\left(P_i^{vir}, P_i^{rec}\right) \quad (3)$$

With $\sum_{i=1}^{n} w_i = 1$

Here, we choose to compose RQ^{mech} with the four mechanical properties elastic modulus (E), yield strength (σ_y), impact strength (a_c) and strain at break (ε_b), allowing Eq. (3) to be rewritten as:

$$RQ^{mech} = w_E \cdot f\left(E^{vir}, E^{rec}\right) + w_\sigma \cdot f\left(\sigma_y^{vir}, \sigma_y^{rec}\right) + w_{a_c} \cdot f\left(a_c^{vir}, a_c^{rec}\right) + w_{\varepsilon_b} \cdot f\left(\varepsilon_b^{vir}, \varepsilon_b^{rec}\right) \quad (4)$$

Ultimately, per application a value for $\alpha^{rec:vir}$ can then be defined as the minimum of the two resulting RQ factors (Eq. (5)). In this way, $\alpha^{rec:vir}$ reflects the necessity of complying with the application demands in terms of both processing and mechanical properties. For example, a recyclate can score high for RQ^{mech}, but the required processing technique cannot be applied because the flow of the recycled plastic does not allow proper processing. Thus, this should result in an overall low value for $\alpha^{rec:vir}$.

$$\alpha^{rec:vir} = min\left[RQ^{mech}, RQ^{proc}\right] \quad (5)$$

Within this concept, defining the scoring functions is the crucial step in determining the RQ factors. Defining the functions is comprised of several aspects, which are to be considered for each of the property/application combinations:

- What is the range of optimal values for this property?
- In what sense should the property match that of the virgin plastic? Is it a threshold value, meaning that higher/lower than is always good or is deviation problematic in both directions?
- How tolerant is this application towards a deviation from the 'perfect' property value? Some applications are more robust than others: while a slightly too high MFI may be catastrophic for film blowing, it can be quite acceptable for injection moulding.

A proper scoring function of a technical property must be able to return a score of 1 if the value of the recycled plastic is in line with the optimal value(s) for a given application. A score less than 1 should reflect the proportional loss of technical quality caused by the deviation of the recycled plastic's property from the optimal value(s).

These functions are thus hard to grasp with a simple equation. To quantify the functions, technical data from TDSs of virgin plastic grades and expert opinions should be consulted. The shape of the function will reflect how (un)tolerant an application is to a deviation from the optimal value, translating to a corresponding numerical level of resulting quality loss. This can result in different shapes. Fig. 4a shows a trapezoidal function in which a recycled material's property within an optimal range is appreciated with a score of 1. A too high or low property of the recycled plastic is scored by a linear decrease, the slope of which is tuned according to the criticality of the deviation from the optimal value(s). This slope may be steeper on one side than on the other. For example, injection moulding is quite forgiving towards higher MFI values, but less so towards low values. Examples of other functions that may be appropriate for valuing a recycled plastic's property are shown in Fig. 4b–e. A bell-shaped type of function (Fig. 4b) is useful for properties that must lie strictly within an optimal range and moving outside this range has a dramatic effect on the functionality of the intended application. When the slope of a bell-shape function flattens, or when a Gaussian function is used instead (Fig. 4c), there is a transition from a range of optimal values to a single optimal value. This is interesting to score properties from the converter's point of view, as a specific converter often requires one ideal value for its processing line or target product. Properties like MFI and E are typically constricted by both lower and upper threshold values as being acceptable, hence they can be appreciated by scoring functions such as Fig. 4a, b and c. The mechanical properties σ, a_c and ε_b generally only have a lower threshold value to fulfil the functionality of the product. Thus, scoring functions as shown in Fig. 4d and e are more suitable. A sigmoidal function (Fig. 4e) gives a more gradual decline in score compared to the linear decrease of a single-sided trapezoidal function (Fig. 4d). The steepness of the function can be adjusted to express the severity of quality loss due to deviating from the optimal value.

Table 2
The recycled plastics within the case study and the mean values for their corresponding properties.

Property	Recycled plastic grades rPE_{bottle}	rPE_{film}	$rPE_{IMrigid}$
MFI (g/10 min)	0.42	0.54	4.5
E (MPa)	912	218	900
σ_y (MPa)	21	6.7	24
a_c (kJ/m^2)	15	91	5
ε_b (%)	115	147	400

Table 3
Optimal values for the four considered application categories based on values from TDSs of virgin grades of PE.

Property	Application group For film		For bottle		For IM rigid		For IM flexible	
	Min	Max	Min	Max	Min	Max	Min	Max
MFI (g/10min)	0.25	4	0.4	1.8	0.8	20	1.5	70
E (MPa)	89.6	300	1000	1300	850	1450	130	340
σ_y (MPa)	10		25		23		9	
a_c (kJ/m^2)	40*		6,5		4		40*	
ε_b (%)	300		300		200		300	

* Impact strength value of 40 kJ/m^2 is chosen from literature reference (Graft Polymer, n.d.).

4. Case study

To support the proposed concept, a case study is elaborated, in which the RQ factors and $\alpha^{rec:vir}$ values are calculated for three commercially available recycled PEs (rPE) for their use in four different product applications. The chosen application categories are film, bottle, injection moulding for rigid products (IM rigid) and injection moulding for flexible products (IM flexible). The three selected rPEs are sourced from waste bottles (rPE_{bottle}), film (rPE_{film}) or injection moulded rigids ($rPE_{IMrigid}$). These commercial grades are advertised as high quality for the respective applications. Trade names are kept confidential. Their properties are shown in Table 2. Mean values for rPE_{bottle} and $rPE_{IMrigid}$ were adopted from the available TDSs, while mean values of rPE_{film} were determined in-house on injection moulded ISO bars of the respective recyclate. This case study will investigate for each considered rPE whether it has the technical quality to be applied in each of the four considered product applications. A spreadsheet file with a complete example of the calculation of the RQ factors for the considered rPEs for one application is available as Supplementary material.

Based on a search of virgin grades' TDSs for the considered applications the minimal required value (for σ_y, a_c, and ε_b) or range of optimal values (for MFI and E) was determined for the four application categories considered in this work. These values can be found in Table 3. Virgin PE typically has a very high value for ε_b, as the molecular architecture of the material allows for high levels of shear yielding (Van Belle et al., 2020). The levels of ε_b that such PEs can reach is often in the

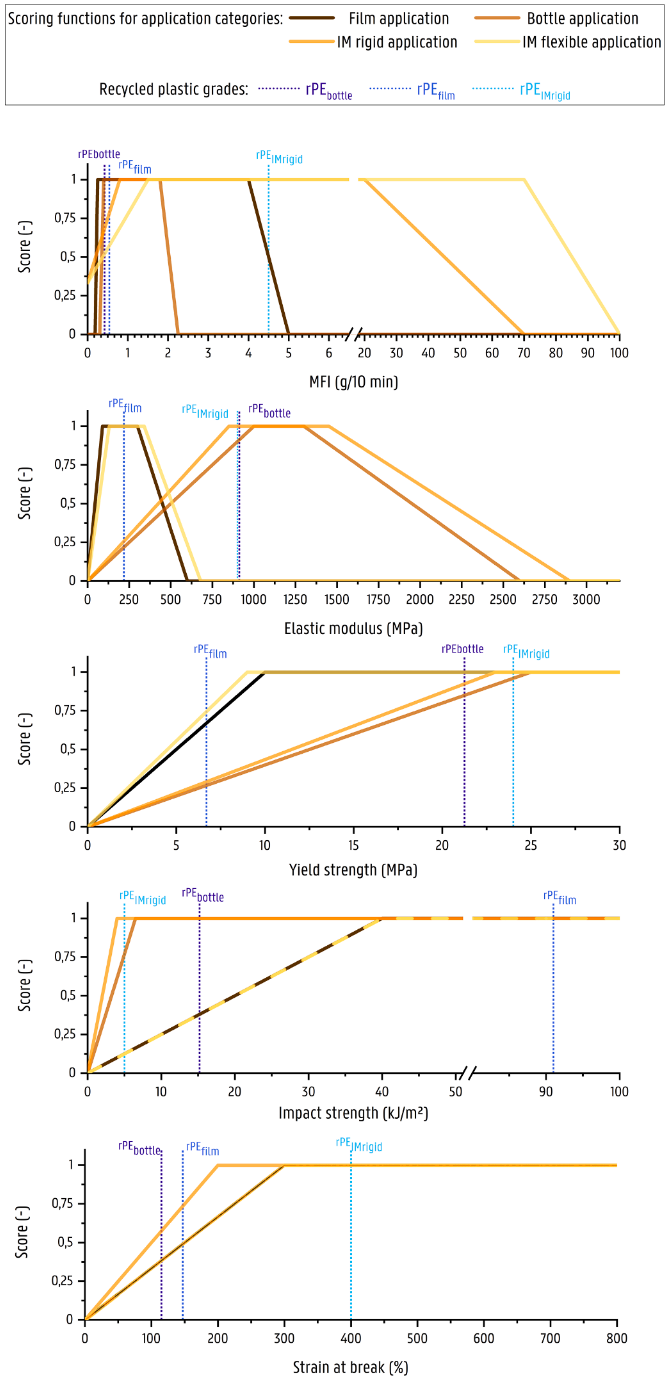

Fig. 5. The trapezoidal functions used for scoring the different properties per application group (Film, Bottle, IM rigid and IM flexible). The values for the properties of the three rPEs to be scored are indicated with dotted lines. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

range of 600–800 or even over 1000%, while this is rarely required for the product application. Therefore, to allow for a fair and functional assessment of the rPEs' ductility, the lower threshold for ε_b for the film, bottle and IM flexible category is set at 300%, which is considered as sufficiently high that all these common applications can be covered. IM rigid products are less demanding for ε_b, and thus the threshold for this category is set at 200%.To illustrate the principle of the proposed concept, the individual scoring functions are here approximated by trapezoidal functions

Table 4
The weighting factors used to calculate the RQ^{mech} for the considered application groups.

RQ^{mech} weight factor w_i					
		Application groups			
Case		Film	Bottle	IM rigid	IM flexible
1	E	0,25	0,25	0,25	0,25
	σ_y	0,25	0,25	0,25	0,25
	a_c	0,25	0,25	0,25	0,25
	ε_b	0,25	0,25	0,25	0,25
2	E	0,2	0,2	0,4	0,2
	σ_y	0,2	0,4	0,2	0,2
	a_c	0,2	0,2	0,2	0,4
	ε_b	0,4	0,2	0,2	0,2
3	E	0,3	0,3	0,5	0,3
	σ_y	0,3	0,3	0,2	0,2
	a_c	0	0,3	0,3	0,4
	ε_b	0,4	0,1	0	0,1

Fig. 5. Trapezoidal functions allow easy implementation of the optimal range obtained from the TDS search. In addition, the (in)tolerances of the different application categories for each property can conveniently be modelled by altering the slopes of the trapezoidal functions. The scoring functions for MFI were formulated taking into account the nature of the respective processing technique. Thus, the score for the bottle and film application category linearly decreases to 0 for a 0 to 25% deviation from the upper and lower limits of the optimal range. The slopes of the MFI scoring functions of the injection moulding techniques, on the other hand, are determined by stipulating that for a 75% deviation from the optimal values, a score of 0.5 is still returned. In this way, it is expressed that the processing techniques for film and bottle are more restrictive for deviations from the optimal range in comparison to the injection moulding techniques. Concerning the trapezoidal functions of the mechanical properties, the slopes are set in such a way that a recycled plastic whose respective property deviates by 50% from the desired value(s) is assigned a score of 0.5.

All values of the considered properties of the rPEs (Table 2) are indicated in Fig. 5 with a dotted line, from which the corresponding scores for each property can then be derived per application category.

In this case study, three sets of weighting factors are applied for the calculation of RQ^{mech}. A first set considers each property as equally important ($w_i = 0.25$). The second set gives a higher weight to one mechanical property (for film $w_{\varepsilon b} = 0.4$, for bottle $w_{\sigma y}$, for IM rigid w_E and for IM flexible $w_{ac} = 0.4$), while the other properties are weighted as equal ($w_i = 0.2$). Explanations on the choice of the most relevant mechanical property per application can be found in Supplementary material (Table S5). A third case assigns a more nuanced weighting, in which the different properties are valued according to their relative importance for the functionality of the specific application. As shown in Fig. 1, this can be remarkable different per product type. Table 4 provides a summary of the three cases considered in this work.

Using the scores from the trapezoidal functions, RQ^{proc} and RQ^{mech} were then calculated for each rPE and this for each application category. Finally, $\alpha^{rec:vir}$ can be determined as the minimum of these RQ factors. The scores for each property, RQ factors and $\alpha^{rec:vir}$ values are presented in Fig. 6. For each application, the proposed concept allows one to verify what the technical substitutability of the rPE is for this application. Furthermore, it allows comparing different recycled plastics within one application group. Fig. 6 also provides an arbitrary scale: excellent (>0.9), good (>0.7), suitable (>0.5) and unsuitable (<0.5). The proposed RQ and $\alpha^{rec:vir}$ factors are aimed at indicating the technical (un) suitability of a recycled plastic for a certain application.

Two main conclusions can be drawn from the results in Fig. 6. First, high values for $\alpha^{rec:vir}$ are only obtained if both RQ^{proc} and RQ^{mech} are high. This is clear from the fact that $\alpha^{rec:vir}$ are good ($\geq$ 0.70) to excellent ($\geq$ 0.90) for the application categories for which the rPEs are actually intended, namely rPE_{bottle} into bottle, rPE_{film} into film and $rPE_{IMrigid}$ into

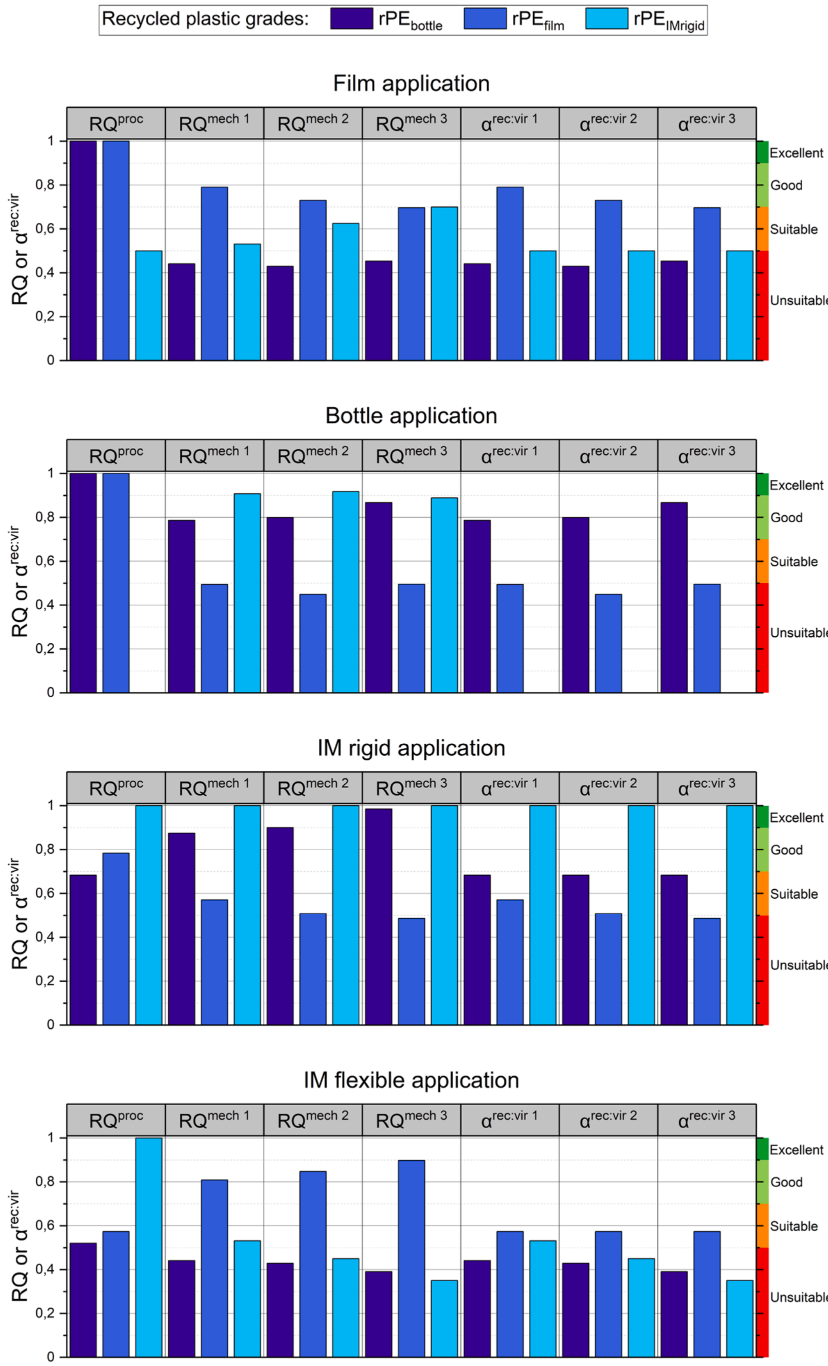

Fig. 6. Calculated values for RQ^{mech}, RQ^{proc} and $\alpha^{rec:vir}$ for the commercially available recycled grades rPE$_{bottle}$, rPE$_{film}$ and rPE$_{IMrigid}$ for each of the considered application groups. 1, 2 and 3 in superscript denote the three weighting sets applied, of which the separate weighting factors can be found in Table 4. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

IM rigid applications. rPE$_{bottle}$ scores high for RQ^{proc} for film application, but due to the mismatch in mechanical properties resulting in a low RQ^{mech}, it is attributed a low $\alpha^{rec:vir}$. For the bottle application, low $\alpha^{rec:vir}$ values for rPE$_{film}$ are similarly due to low RQ^{mech} values despite the excellent value for RQ^{proc}. In the bottle application category, rPE$_{IMrigid}$ is excellent in terms of mechanical properties (high RQ^{mech}), but its relatively high MFI predicts an impossible processing (RQ^{proc} = 0) into a bottle application. This causes $\alpha^{rec:vir}$ to be zero. Second, the weighting factors for the calculation of RQ^{mech} must be carefully chosen to ensure that the mechanical functionality of a particular application is well represented. Thus, when scoring the IM flexible application category, the highest RQ^{mech} is calculated for rPE$_{film}$, as a result of its matching E and a_c. By assigning higher and thus more relevant weights (weighting set 3) for these essential properties for an IM flexible product, this is better reflected in the RQ^{mech} factors of the rPEs. This is also clear for the bottle category: the first two sets of weighting factors attribute too much importance to the low ε_b value of rPE$_{bottle}$. The third and more application-specific set balances this out more accurately, resulting in a higher RQ^{mech} for rPE$_{bottle}$. By assigning higher weights to E and a_c in the IM rigid category, the difference between the RQ^{mech} factors of rPE$_{bottle}$ and rPE$_{IMrigid}$ is cancelled out. This suggests that the mechanical quality of rPE$_{bottle}$ is adequate for an IM rigid product.

Also noteworthy is the fact that none of these rPEs are suitable for the production of flexible products via injection moulding. rPE$_{film}$ scores the highest $\alpha^{rec:vir}$, but is limited by its low flow property (low RQ^{proc}). Here, rPE_{bottle} and $rPE_{IMrigid}$ do not match in terms of mechanical functionality (low RQ^{mech}). Note that rPE_{film} and rPE_{bottle} could be processed via injection moulding, but a possible reduced output/impact on the processability of these materials must be taken into account.

5. Conclusions and outlook

A proper assessment of the substitution potential of a recycled plastic is essential in the transition to a circular economy of these valuable materials. The displacement of a virgin plastic by a recycled one is largely determined by the compliance of their technical functionalities. However, this work shows that this technical functionality cannot be based on one single property. Recycled plastics must have the appropriate processing characteristics and mechanical properties to be reprocessed into the desired application. In addition, the abundance of available plastic types and grades further complicates a straightforward calculation of the technical substitutability of a recycled plastic.

The concept proposed in this paper allows a quantification of the substitutability for a recycled plastic intended for substitution in a specific application (group), taking into account both the processability and mechanical properties. For each key material property of a specific application (group) the deviation is quantified of the actual value (of the recycled plastic) from the acceptable range of values (determined by relevant virgin plastics), which yields a score between 0 and 1. Based on the corresponding scores for each property of a recycled plastic, the mechanical and processability recycling factors can be obtained. The proposed mechanical recycling factor is a weighted sum of the scores of the mechanical properties, in which the weighting factors also need to be tailored to the application. Ultimately, in this paper, the substitutability is determined by the minimum of both recycling factors.

To elaborate on this concept, the recycling quality factors and resulting substitutability values were calculated for three commercially available PE recycled materials, considering substitution into four application categories (film, bottle, IM rigid and IM flexible). In this paper, we approximated per application the different scoring functions for each relevant property using trapezoidal functions. The substitutability values obtained in this case study give, for each application, a proper indication of the technical quality of the recyclates considered. The considered recyclates score good (> 0.70) to excellent (> 0.90) for their intended application. In contrast, the generally low substitutability values of recyclates for use in other application categories indicate a strongly reduced functionality or even unsuitability.

From this work and the elaborated case study, it appears that there is a lack of a publicly accessible data that links the required properties to the respective application (group). Such a property-application matrix should make it possible to extend the proposed scoring system to establish a general technical quality assessment of recycled plastics. By considering the relevant application-specific properties, a proper estimation of the functionality loss of recycled plastics can be provided towards decision making of the different recycled plastics regarding their potential to be used in a particular application. This should help plastic processors to make a reliable comparison and selection between virgin and recycled grades on the one hand, and between different recycled plastics on the other hand. In addition, an extended scoring system can also indicate which additional pre-treatment and sorting steps are necessary to obtain a recyclate with the desired properties.

The proposed factors reflect the loss of technical functionality. However, involving more macromolecular knowledge in future research will help to indicate which are the detrimental contaminants. Further research could also extend the assessment of the technical functionality of recycled plastics to other properties (e.g., melt strength, resistance to environmental stress cracking, …) and quality limiting criteria such as odour and colour. Finally, it should be noted that substitution depends on more than technical substitutability alone. Aspects related to end-user attractiveness and market aversion must be carefully studied and taken into account. Within the circular economy, the importance of knowledge about the substitutability of recovered resources is high with a view to obtaining secure resource supplies.

CRediT authorship contribution statement

Ruben Demets: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. **Karen Van Kets:** Writing – review & editing. **Sophie Huysveld:** Writing – review & editing. **Jo Dewulf:** Writing – review & editing. **Steven De Meester:** Conceptualization, Resources, Supervision, Writing – review & editing. **Kim Ragaert:** Conceptualization, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.105826.

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