Developing circularity, renewability and efficiency indicators for sustainable resource management: Propanol production as a showcase

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A B S T R A C T
Resource efficiency analysis is an important tool in the chemical sector to evaluate the performance of new process concepts. However, such analysis does not account for the renewability and circularity of resources. Therefore, a resource efficiency and use analysis, including those two aspects, is proposed in this paper. A renewability indicator and recovery indicator were calculated as a measure for renewability and circularity, respectively. In addition, the resource efficiency was determined at different levels. At the life cycle level, the cumulative exergy extraction of the natural environment method was applied and the cumulative degree of perfection, including waste-as-resources, was calculated. Exergy calculations were used to determine the exergetic efficiency at process chain and plant level and to identify inefficiencies. A new propanol production concept, using biogas (scenario BG), marginal gas (scenario MG) and associated gas (scenario AG), was selected as a case study. Exergetic efficiencies are high at the individual process level (between 90 and 100%). However, the preceding biogas production in scenario BG is inefficient (exergetic efficiency of 12%). The exergetic efficiency at the process chain level amounts to 45–68% due to the high exergy content of the recycling stream and the low conversion of methane into propanol per pass. Scenario AG has the highest cumulative degree of perfection (including waste-as-resources) compared to the other scenarios (28% against 6 and 14% in scenario BG and MG). In contrast, when looking at both renewability, circularity and efficiency, scenario BG is identified as the most promising scenario. Thus, this study shows that it is important to include those three aspects in resource efficiency analysis. Finally, implementing renewable electricity production and heat integration in the process concept may increase the resource efficiency.

1. Introduction

The chemical industry is facing various issues such as dependency of fossil resources, energy intensive production processes, climate change due to significant greenhouse gas emissions and biodiversity loss due to toxic emissions into the environment. Concrete actions must be taken in this respect to realise the necessary transition to a resource-efficient, climate neutral and environmentally sustainable economy. In 2019, the European Commission launched the European Green Deal with measures to guide this transition. Herein, environmental assessments of technologies, including resource efficiency analysis, are put forward as important tools to evaluate innovations in the chemical industry (Thormann et al., 2021). Several definitions were reported for the term resource efficiency. The European Commission (2022a) describes it as "using the Earth’s limited resources in a sustainable manner". Many studies applying a resource efficiency analysis in the chemical sector are available in the literature (Ghannadzadeh and Sadeqzadeh, 2016; Luis and Van der Bruggen, 2014; Lozano et al., 2018). However, different indicators are used to measure resource efficiency in industrial systems. Huysman et al. (2015) proposed a framework to structure these indicators and to enhance their use in environmental impact assessment methods. Resources are divided in three categories in this framework: natural resources, industrial resources and waste-as-resources.

A resource efficiency analysis is performed at various levels. At the...
flow undeniably has an exergy content. For instance, in the oil sector, the associated gas is seen as a waste fraction of the petroleum production related to petroleum production is not taken into account. However, the actual exergy content of associated gas should not be neglected when performing a resource efficiency analysis. Various circularity indicators, which can be integrated in the resource efficiency analysis, are being developed at different scopes (e.g., process level) (Gonçalves et al., 2021; Moraga et al., 2019). The system can be analysed from the input perspective (utilising waste as a resource) or from the end-of-life perspective (utilising its end-of-life products into another life cycle). Life cycle thinking is also applied in some cases (e.g., circularity performance indicator from Huysman et al. (2017)). A comprehensive overview of developed circularity indicators can be found in Moraga et al. (2019). In this paper, both renewability and circularity will be integrated into the resource efficiency analysis, in addition to the CEENE method. This new concept is called a resource efficiency and use (REU) analysis throughout this paper. The circularity of resources is viewed from the input perspective. Fig. 1 shows an overview of the REU analysis.

The production of 1-propanol is selected as a case study to conduct this REU analysis. Current propanol production is energy intensive and heavily relies on conventional fossil resources (Nanda et al., 2020; Walther and François, 2016). Therefore, a novel process concept, denoted as C123, was proposed, converting widely available, cheap and wasted methane sources such as biogas (scenario BG), marginal gas (scenario MG) and associated gas (scenario AG) into 1-propanol (European Commission, 2022b; Fonseca et al., 2021; Motte et al., 2022; Sintef, 2022). Biogas is often produced via anaerobic digestion and it is mainly used as fuel for combined heat and power (CHP) installations to produce electricity and heat. After removal of CO₂ and other impurities such as H₂S, biogas based methane can directly be injected in the gas grid (Welland, 2010). Marginal gas is not used, because these gas reservoirs are not exploited yet (Ministry of Petroleum Resources, 2013). Associated gas is often flared for economic and technical reasons (Soltaniez et al., 2016). When the latter feedstock is converted into propanol, flaring emissions can be avoided. Motte et al. (2022) showed that scenario AG was the best option when considering the global warming burden, water consumption and human health damage by carcinogenic. The dependency on fossil fuels was the lowest in scenario BG. These new pathways, starting from the selected feedstocks, could potentially increase the resource efficiency of the current propanol process level, process chain level and plant level, exergy calculations can be used for this purpose. Exergy is the maximum amount of useful work, which can be obtained from a resource or system when it is brought to equilibrium with a reference environment through reversible processes. Due to the irreversibility of actual processes, exergy is always lost. The exergy loss can be a useful tool to quantify the depletion of natural resources (Dewulf et al., 2008; Mahian et al., 2020). At the life cycle level, the Cumulative Exergy Extraction of the Natural Environment (CEENE) method, developed by Dewulf et al. (2007), is one of the methods that can be applied. The benefits of using this thermodynamic method are a full coverage of natural resources including land use, no need for using weighting factors and avoidance of double counting. However, this method also has some limitations.

First, CEENE does not take into account the renewability of the feedstocks used by the (industrial) system. Two approaches could be found in the literature to solve this issue. Huysveld et al. (2015) proposed to address the non-renewable character of fossil resources by including the ancient solar energy consumption during its formation (Dukes, 2003). In contrast, Dewulf et al. (2000) calculated a renewability parameter defined as the fraction of renewable exergy consumption divided by the total exergy consumption. Afterwards, the renewability parameter and the efficiency are shown in a graphic to find in the literature to solve this issue. Huysveld et al. (2015) proposed to address the non-renewable character of fossil resources by including the ancient solar energy consumption during its formation (Dukes, 2003). In contrast, Dewulf et al. (2000) calculated a renewability parameter defined as the fraction of renewable exergy consumption divided by the total exergy consumption. Afterwards, the renewability parameter and the efficiency are shown in a graphic to represent two key sustainable features with respect to resource efficiency and use: life cycle exergetic efficiency and renewability degree of primary resources. In most studies, a renewability factor was calculated using a similar approach as in Dewulf et al. (2000) (Liu, 2014; Liu et al., 2010; Ugliati and Brown, 1998; Zvolinschi et al., 2007). However, the exergy content of the inputs was replaced by their energy or emergy content. Pradhan et al. (2008) defined a renewability indicator specific to the biofuel sector as the fuel energy output divided by the non-renewable energy input.

Second, CEENE is not applicable to circular systems, because originally, it was only developed for linear economies, where primary inputs are extracted from nature. This impact assessment method is linked to LCI databases (e.g., Ecoinvent). When waste or recyclable material are used as a resource, the input is considered burden free in these databases (the preceding life cycle steps are not considered). In that case, no exergy value is assigned to this input flow, even though in reality the flow undeniably has an exergy content. For instance, in the oil sector, associated gas is seen as a waste fraction of the petroleum production and consequently, when used as an input, the environmental burden related to petroleum production is not taken into account. However, the actual exergy content of associated gas should not be neglected when performing a resource efficiency analysis. Various circularity indicators, which can be integrated in the resource efficiency analysis, are being developed at different scopes (e.g., process level) (Gonçalves et al., 2021; Moraga et al., 2019). The system can be analysed from the input perspective (utilising waste as a resource) or from the end-of-life perspective (utilising its end-of-life products into another life cycle). Life cycle thinking is also applied in some cases (e.g., circularity performance indicator from Huysman et al. (2017)). A comprehensive overview of developed circularity indicators can be found in Moraga et al. (2019). In this paper, both renewability and circularity will be integrated into the resource efficiency analysis, in addition to the CEENE method. This new concept is called a resource efficiency and use (REU) analysis throughout this paper. The circularity of resources is viewed from the input perspective. Fig. 1 shows an overview of the REU analysis.

Fig. 1. Illustration of REU analysis with the integration of renewability and circularity.

<table>
<thead>
<tr>
<th>Resource efficiency analysis</th>
<th>Renewability</th>
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<tbody>
<tr>
<td>Process level (a-level) – plant level (b-level)</td>
<td>Nature of resource taken into account (fossil versus renewable)</td>
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<tr>
<td>Exergetic efficiency</td>
<td>– Share of renewable resources?</td>
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<td>Life cycle level (y-level)</td>
<td>Circularity</td>
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<tr>
<td>Cumulative degree of perfection (CDP)</td>
<td>– Both for linear and circular economies</td>
</tr>
<tr>
<td>CDP*: including waste-as-resources</td>
<td>– Including waste-as-resources</td>
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production. In several studies, the exergetic efficiency of biogas production from various feedstocks, biogas purification and/or its use in a CHP and gas flaring reduction technologies was analysed (Barrera et al., 2016; De Meester et al., 2012; Mahian et al., 2020; Vilaridi et al., 2020; Xydis et al., 2013; Zaresharif et al., 2021). The conversion of biogas and associated gas into chemicals such as propane has not yet been examined.

Our present work aims to integrate circularity and renewability of resources into resource efficiency analysis, resulting in a REU analysis. To quantify the former aspect, the recovery indicator (RI) is proposed. For addressing renewability in the analysis, the approach builds upon Dewulf et al. (2000). The production of 1-propanol via the C123 process was performed, also taking into account circularity and renewability. The results for the different C123-scenarios were compared based on CDP*, renewable share of the used resources and RI to find the most promising scenario. Finally, improvements to the preliminary process concept were recommended based on the results of the REU analysis.

2. Methodology

2.1. Resource efficiency and use analysis

2.1.1. Calculation of exergetic efficiency and cumulative degree of perfection

A resource efficiency analysis was conducted from cradle-to-gate and at different levels, namely the life cycle level (γ-level), plant level (β-level), process chain level (α-level) and process level (Van der Vorst et al., 2009). At plant, process chain and process level, exergy calculations are performed to quantify the exergetic efficiency of a process concept and its individual production steps. The total exergy value of a flow consists of the chemical exergy, physical exergy, kinetic exergy and potential exergy. The last two terms are negligible (Dewulf et al., 2007). The chemical and physical exergy were calculated for gases via the following formulas (Szargut, 2005):

$$E_{x,\text{chemical}} = \sum x_i \times E_{x,i}$$

$$E_{x,\text{mix}} = \frac{R}{\text{MM}_{\text{mix}} \times 1000} \times (T_0 - \sum x_i \times \ln (\gamma_i \times x_i))$$

$$E_{x,\text{physical}} = \sum \frac{C_p,i \times (T - T_0) - T_0 \times C_v,i \times \ln \left(\frac{T}{T_0}\right)}{T_0} + R \times T_0 \times \ln \left(\frac{P}{P_0}\right)$$

With $E_{x,i}$ = exergy value for compound i in mixture in kJ/mol, $x_i$ = molar fraction for compound i, $E_{x,i}$ = standard exergy value for compound i in kJ/mol, $C_v,i$ = gas constant in J/mol/K, $T$ = temperature in K, $P$ = pressure of reference environment in atm, $P_0$ = total pressure of reference environment in atm, $n$ = number of compounds present in gas mixture, $E_{x,\text{physical}} = $ physical exergy in MJ/kg.

Next, all exergy values (in MJ/kg) were multiplied by the mass flow rates. In these calculations, all gas mixtures were considered as ideal gases. Therefore, the physical exergy was determined via the formula for an ideal gas and the activity coefficients in Eq. (2) are equal to 1. When the exergy value $E_x(i)$ for a chemical compound or a biological feedstock was not available, this was calculated via the group contribution method or the $\beta \times$ lower heating value (LHV) method, respectively (Szargut, 2005). In case of utilities, $\beta$-factors were used to convert energy values to exergy values. For example, a $\beta$-factor of 1 was used for electricity. The $\beta$-factor for heat was derived via the formula below (Dewulf et al., 2008).

$$\beta = 1 - \frac{T_0}{T}$$

With $\beta = \text{factor to convert energy to exergy}$, $T_0 = \text{temperature of reference environment in K}$, $T = \text{temperature in K}$.

Finally, the rational exergetic efficiency of each production step was calculated as follows (Romero and Linares, 2014):

$$\eta_i = \frac{\sum E_{x,\text{final outputs}}}{\sum E_{x,\text{inputs}}}$$

With $\eta_i = \text{rational exergetic efficiency of production step}$, $\sum E_{x,\text{final outputs}} = \text{sum of the exergy values for all useful outputs}$, $\sum E_{x,\text{inputs}} = \text{sum of the exergy values for all inputs}$.

Only products and by-products are taken into account in the calculation of the exergetic efficiency; waste and emissions are excluded.

At the life cycle level, the CEENE method was applied. This method takes into account the exergy extracted from the natural environment for industrial processes and consumption. Eight impact categories were determined for cumulative exergy consumption quantification (in MJ), namely abiotic renewable resources, minerals, fossil fuels, nuclear energy, metal ores, water resources, land and biotic resources and atmospheric resources. The reference environment of Szargut was chosen for these calculations with a temperature of 298 K and a total pressure of 1 atm (Dewulf et al., 2007). A distinction must be made between natural and human-made systems to avoid double counting when accounting for resource use. For example, small-scale forestry is seen as a natural system (without human intervention), while cultivation of crops such as maize is considered as a human-made system (with human intervention) due to land occupation deprived for other human purposes (Alvarenga et al., 2013). Inputs were also divided into three different categories to distinguish them based on their origin, more specifically natural resources (e.g., water), industrial resources (e.g., purified natural gas) and waste-as-resources (Huysman et al., 2015). Waste-as-resources refer to waste fractions and recyclable materials (by-products from a preceding process/system), which are both not allocatable in LCA databases. The European Directive 2008/98/EC was used to distinguish between waste fractions and by-products (Eur-Lex, 2018). The efficiency of a system at this level, also called cumulative degree of perfection (CDP), was obtained via Eq. (7) (Szargut and Morris, 1987).

$$CDP = \frac{E_{x,\text{products}} + E_{x,\text{by-products}}}{CEENE}$$

With $E_{x,\text{products}} = \text{exergy value of the desired product in MJ}$, $E_{x,\text{by-products}} = \text{exergy of the produced by-products of this system in MJ}$ and $CEENE = \text{cumulative exergy extraction of the natural environment for the whole system in MJ}$. Eq. (7) holds for a linear economy and this expresses how efficient resources are used in the C123 process concept. In Section 3, the results of the REU analysis are represented via a Grassmann diagram (Grassmann, 1959).

2.1.2. Addressing waste-as-resources in calculation of CDP

Waste-as-resources such as manure and associated gas are considered burden free in LCI databases linked to the CEENE method. Consequently, their corresponding exergy input is not accounted for and the
exergy content of these waste-as-resources (WAR) needs to be added to the CEENE value in Eq. (7) as an additional term (see Eq. (8)). Only the exergy content of the waste-as-resources themselves should be included, not the extraction of natural resources related to the generation of waste-as-resources. For example, associated gas is a waste fraction of the petroleum production. The natural resources extracted for this process step are entirely attributed to petroleum production (cut-off approach).

$$CDP^* = \frac{E_{\text{primary}} + E_{\text{by-products}}}{\text{CEENE}} + E_{\text{WAR}}$$

(8)

With $CDP^*$ = the cumulative degree of perfection including waste-as-resources, $E_{\text{primary}}$ = the exergy value of the desired product in MJ, $E_{\text{by-products}} = \text{exergy of the produced by-products in this system in MJ}$, CEENE = the cumulative exergy extraction from the natural environment for the whole system in MJ, $E_{\text{WAR}} = \text{exergy content of waste-as-resources such as associated gas in MJ}$. This last parameter is not cumulative unlike the CEENE-value. The sum of CEENE and $E_{\text{WAR}}$ is called the total extraction of resources (TER) throughout this paper.

2.1.3. Addressing renewability of resources in resource efficiency analysis

When calculating only the CDP, the origin of primary resources (renewable or fossil) is not an important factor in the resource efficiency analysis of a process concept. However, using renewable resources could decrease the dependency of chemical production processes on finite fossil reserves. To take into account renewability in the resource efficiency analysis, a renewability factor (RF) was calculated as suggested by Dewulf et al. (2000) and this is coupled with the CEENE method.

$$RF = \frac{\text{CEENE}_{\text{renewable}}}{\text{CEENE}}$$

(9)

With $RF$ = renewability factor, $\text{CEENE}_{\text{renewable}} = \text{the renewable share of the CEENE in MJ and CEENE} = \text{the cumulative exergy extraction from the natural environment for the whole system in MJ (both renewable and non-renewable).}$ The following categories from the CEENE method were included as renewables: abiotic renewable resources, water resources, land and biotic resources and atmospheric resources. For waste-as-resources, the cut-off approach was used.

2.1.4. Addressing circularity of resources in resource efficiency analysis

Another factor, which is not covered in resource efficiency analysis, is the circular performance of the system. For instance, associated gas is a waste-as-resource that can be used to produce propanol, creating a circular economy and replacing the virgin material (V), namely naphtha. The RI is proposed in this work, as no suitable circularity indicator related to the chemical sector was available according to the consulted literature. This indicator can be calculated via Eq. (10):

$$RI = \frac{E_{\text{WAR}}}{E_{\text{WAR}} + E_V}$$

(10)

With $RI = \text{recovery indicator}$, $E_{\text{WAR}} = \text{the exergy content of waste-as-resource}$ and $E_V = \text{the exergy content of the virgin material (e.g., naphtha)}$. Fig. 2 shows a general illustration of this method. The RI was calculated at the process level and open loop recycling was assumed. For waste-as-resources, a cut-off approach was again used, but its exergy content ($E_{\text{WAR}}$) was still taken into account.

2.1.5. Graphical selection of best scenario

$CDP^*$, RF and the RI were plotted in a three-dimensional figure for different scenarios. In the ideal situation, a product is entirely obtained from renewable resources and the $CDP^*$ and RI are equal to one. It was assumed that all indicators had an equal weight. Finally, the norm between the scenarios and the origin was calculated. The scenario furthest from the origin was considered the best option in terms of REU.

2.2. Case study: C123-scenarios for 1-propanol production

2.2.1. Description of C123 process concept

In Figs. 3–5, the preliminary process concepts for all three C123-scenarios are shown. These concepts have been discussed in detail in our previous work (Motte et al., 2022). However, some additional production steps (e.g., purification of feedstocks) have now been included and are discussed in this section. Scenario BG, MG and AG are situated in Germany, Russia (Angara-Lena Terrace) and Russia (Khanty-Mansi), respectively. In scenario BG, the gas is purified via a biological desulphurisation (Weiland, 2010). Afterwards, the CO$_2$ content is drastically reduced before the oxidative conversion of methane (OCoM) (Fonseca et al., 2021). In scenario MG, the liquid redox process is used to remove H$_2$S from the gases (Gendel et al., 2009). In scenario AG, a desulphurisation is not required. However, natural gas liquids such as propane and butane must be separated via a de-ethaniser to avoid the formation of by-products (butanol, pentanol, oxygenates, etc.). In fact, the associated gas contains 15% more higher hydrocarbons (e.g., propane, butane, etc.) on a weight basis than marginal gas (Snytnikov et al., 2018). Next, in all scenarios, the pretreated gas is converted into propanol via three reactions: OCoM, hydroformylation (HF) and hydrogenation. Gas processing is needed after each production step, called post-treatment throughout this work. Electricity is supplied via the German electricity grid in scenario BG. For scenarios MG and AG, a gas turbine is considered to be installed at the production site. More details about the preliminary process concept, the reasoning for the choice of the location,

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**Fig. 2.** General illustration for the determination of the recovery indicator (RI). Depending on the nature of the virgin material and the waste-as-resource, the production process may be slightly different, (e.g., additional pre-treatment step).
Fig. 3. Illustration of production steps for scenario BG that is located in Germany. The red and the green dotted box represent the process chain (α-level) and the plant-level (β-level), respectively. Products indicated in blue are useful outputs in this process concept.

Fig. 4. Illustration of production steps for scenario MG that is located in Russia (Angara-Lena Terrace). The red and the green dotted box represent the process chain (α-level) and the plant-level (β-level), respectively. Products indicated in blue are useful outputs in this process concept.
the scale and the assumptions can be found in our previous work (Motte et al., 2022).

2.2.2. Resource efficiency and use analysis applied to C123-scenarios

For the REU analysis, Eqs. (11)–(15) were used to calculate the CDP* and RI for the different C123-scenarios. The numerator specifies the useful outputs obtained in each scenario.

Scenario BG:

\[
CDP^* = \frac{Ex_{\text{propanol}} + Ex_{\text{digestate}} + Ex_{\text{sulfur}}}{CEENE} + Ex_{\text{manure}}
\]

(11)

\[
RI = \frac{Ex_{\text{manure}}}{Ex_{\text{manure}} + Ex_{\text{naphtha}}}
\]

(12)

Scenario MG:

\[
CDP^* = CDP = \frac{Ex_{\text{propanol}} + Ex_{\text{sulfur}}}{CEENE}
\]

(13)

Scenario AG:

\[
CDP^* = \frac{Ex_{\text{propanol}} + Ex_{\text{natural gas liquids}}}{CEENE} + Ex_{\text{associated gas}}
\]

(14)

\[
RI = \frac{Ex_{\text{associated gas}}}{Ex_{\text{associated gas}} + Ex_{\text{naphtha}}}
\]

(15)

In scenario MG, the exergy content of the feedstock was already considered in the CEENE. Moreover, RI was not calculated in this scenario as marginal gas is not a wasted feedstock. In all scenarios, RF is calculated via Eq. (16):

\[
RF = \frac{CEENE_{\text{renewables}}}{CEENE}
\]

(16)

The CDP* values were compared to the CDP* for the current propanol production methods. Both the bio-based and fossil-based production from propanol were selected as reference scenarios. Sugar beet and wheat (Ref P1) and naphtha (Ref P2) are the feedstocks, respectively.

2.2.3. Data collection

Flow charts with all production steps for the different C123-scenarios were constructed in Aspen Plus version V11. Data such as the weight fractions, temperatures and pressures of different process flows was collected in function of 1 kg propanol (scale of 10 kt per y) via simulations. Production information for the biological desulphurisation of biogas and the liquid redox process was obtained from industry (confidential information). Heat capacities for different compounds in the gas mixtures were found in the NIST database (NIST, 2022). For the reference scenarios, data, such as energy requirements, amount of chemicals used, etc., was collected from the Ecoinvent database version 3.6, Belboom and Leonard (2016), Environmental Energy Agency (2019), Kang et al. (2014), Lioussse et al. (2019) and Mohsenzadeh et al. (2017).

To calculate the exergetic efficiency of each production step, all chemical and thermo-physical exergy values (in kJ/mol) were retrieved from Szargut et al. (1988). For the exergetic efficiency of the feedstock, additional chemicals and utilities production, data was obtained from the literature (Boulamanti et al., 2013; Collet et al., 2017; Dufour et al., 2012; Skone et al., 2016). Next, the Phyllis database was consulted to estimate the exergy content of manure and maize silage via the β × LHV method (Phyllis2, 2022). For the calculation of the CDP*, CEENE values were obtained by linking the Ecoinvent database version 3.6 to the CEENE method in the Simapro software version 9.2. In Appendix A, an overview can be found with all inputs, outputs and the corresponding exergy content for each production step.

3. Results and discussion

3.1. General results and discussion case study

Fig. 6 shows the Grassmann diagram for scenario BG. This scheme represents the results of the exergy calculations at the process and plant.
level. All red process blocks correspond to the C123-production steps (forming together the process chain level or alpha level). The green process blocks correspond to the supporting processes (forming together the plant level or beta level). The width of a coloured line is proportional to the exergy content of this flow. Finally, the rational exergetic efficiency of each production step is shown in the right bottom corner of each process block. In scenario BG, the rational exergetic efficiency of all C123-production steps is between 90 and 100% at the alpha level. The overall exergetic efficiency at the alpha level amounts to 45%. However, the preceding biogas production and chemicals production are much less efficient (with \( \eta_r \) amounting to 12.8% and 9.0, respectively). It results in an overall exergetic efficiency of 8.4% at the beta-level.

Table 1 shows the results for the REU analysis at the life cycle level. The TER for each scenario, the TER values for the separate impact categories (in percentages) and the CDP* are included in this overview. The impact categories land and biotic resources, water resources and fossil fuels have the highest contribution to the TER value (respectively, 72.2, 12.5 and 9.7%). Scenario BG has a TER value of 724 MJE\(_{\text{ex}}\) per kg propanol and a CDP* of 6%. When waste-as-resources are included, the CDP from the resource efficiency analysis decreased with 30%, so the rational exergetic efficiency of this process step. Changing the feedstocks (e.g., manure) for biogas production will only yield marginal gains. Moreover, manure also contains micronutrients, which are essential for a high methane production by the bacteria in anaerobic digestion (Xu et al., 2018). Wang et al. (2021) suggested a direct interspecies electron transfer to increase the methane production. As the weight fraction of methane in the biogas will increase, the exergy output and exergetic efficiency will also be enhanced. However, more research is needed to

The Grassmann diagram for scenario AG is represented in Fig. 8. Also in this scenario, the exergetic efficiency of all indicated production steps is higher than 90%. At the alpha level and the beta level, an overall efficiency of 68 and 40%, respectively, is achieved. Moreover, the CDP* amounts to 28% (see Table 1). Finally, fossil fuels contribute more than 76.2% to the TER value and 65% of the exergy input for fossil fuels (excluding the feedstock associated gas) is caused by the production of electricity and heat. Thus, the production of these utilities from renewable resources can drastically increase the CDP* in this case. RF and RI amount to 0.34 and 0.64, respectively, assuming that 1.74 kg associated gas (with an exergy content of 79.78 MJE\(_{\text{ex}}\)) is converted into 1 kg propanol.

### 3.2. Comparison C123-scenarios based on rational exergetic efficiency

The resource efficiency analysis at the process level shows that the biogas production has a very low exergetic efficiency (12.8%). De Meester et al. (2012) reported a rational exergetic efficiency between 30.6 and 66.6% excluding the combination with a CHP. Note that an exergetic efficiency of 50% is considered for a CHP. However, different feedstocks were selected in both studies, which complicates the comparison. The inefficiency of the anaerobic digestion can be explained by the high feedstock amount (33.19 kg manure and 10.93 kg maize silage for 3.57 kg biogas), and consequently a high exergy input, required for the biogas production. In addition, the methane production via anaerobic digestion is limited as the electron transfer between syntrophic bacteria and methanogens, which enable this microbial biomass conversion, is inefficient. No immediate solutions are available to increase the exergetic efficiency of this process step. Changing the feedstocks (e.g., manure) for biogas production would only yield marginal gains. Moreover, manure also contains micronutrients, which are essential for a high methane production by the bacteria in anaerobic digestion (Xu et al., 2018). Wang et al. (2021) suggested a direct interspecies electron transfer to increase the methane production. As the weight fraction of methane in the biogas will increase, the exergy output and exergetic efficiency will also be enhanced. However, more research is needed to
integrate this concept into industry.

Looking at all C123-production steps, the most pronounced exergy destruction in the preliminary process concept is situated in the CO$_2$ reduction stage (92.0%) in scenario BG and in the post-treatment stage of the HF in scenarios MG and AG (90.5 and 90.1%, respectively). In the post-treatment stage of the HF, both the pump for the recirculation of unconverted propanal and the compressor for the pressurisation of the added hydrogen gas consume a lot of electricity (4.21 and 3.74 MJ/kg propanol, respectively). The CO$_2$ reduction also needs 8.75 MJ of heat per kg propanol, for solvent regeneration, and 1.29 MJ of electricity per kg propanol. No major exergy losses occur in the pretreatment stages (desulphurisation in scenarios BG, MG and AG and natural gas liquids recovery in scenario AG). Next, the exergetic efficiency of all chemical reactions is high. For the oxidative conversion of methane, it amounts to at least 94.2% in all scenarios. The high efficiency of the HF (98.9%) can be explained by the high and selective conversion of ethylene to propanal. Consequently, not much progress needs to be made anymore at these production steps.

The production of additional chemicals such as oxygen and hydrogen requires a lot of electricity (18.3 MJ/kg propanol in total), resulting in supporting processes with a low exergetic efficiency. However, the exergy content of these flows is rather low. Oxygen has a very low exergy value on a molar basis and hydrogen is produced in very small amounts. Therefore, these processes do not affect the results to a major extent. The detailed results for all other production steps can be consulted in Appendix B.

Finally, the overall exergetic efficiency at the alpha level is rather low (45–48%) in all scenarios except scenario AG (68%). This can be explained by the high exergy content of the recycling stream. Exergy is lost each time the conversion steps from methane into propanol are passed. The methane conversion per pass must be increased to obtain more propanol. Approximately 0.95 kg methane is converted into propanol in one pass for all scenarios, considering a functional unit of 1 kg propanol. A higher efficiency is reported for scenario AG thanks to the natural gas liquids that also could be sold on the market.

### 3.3. Comparison C123-scenarios based on cumulative degree of perfection (CDP$^*$), renewability and circularity

At the life cycle level, a low CDP$^*$ (6%) is found for scenario BG due to the inefficient conversion of maize silage and manure into biogas. Scenarios MG and AG have a higher CDP$^*$ (14 and 28%, respectively).
due to a lower TER value (see Table 1). Moreover, the relative share for land and biotic resources is higher in scenario BG than in the other scenarios due to manure and maize silage production (72.2% in scenario BG versus 0.3–0.4% in the other scenarios). The opposite is true for the category fossil fuels. Scenario AG looks the most promising scenario based on the CDP*. The additional recovery of natural gas liquids in scenario AG explains again the higher CDP* compared to scenario MG.

The highest RF and RI are obtained in scenario BG. RF in scenarios MG and AG is much lower than in scenario BG as the utilities, such as electricity, must be produced from fossil fuels in the remote locations that were selected for these scenarios. The feedstocks are also non-renewable. In scenarios MG and AG, water resources contribute to the RF for almost 100%, while in scenario BG, the share of water resources, land and biotic resources and abiotic renewable resources amounts to 31, 31 and 8.5%, respectively. The high contribution of land and biotic resources can be explained by the use of maize silage as a feedstock for biogas production and the partial renewable electricity production from biomass in Germany. RI is the highest in scenario BG due to the large amount of manure used with a relatively high exergy content. However, the RI value in the case of associated gas is also relatively high as the associated gas has a higher exergy content than the equivalent amount of naphtha.

In Fig. 9, an overview is given with the results for all indicators (CDP*, RF and RI). Based on these results, a three-dimensional figure was composed enabling the calculation of the distance between the performance of the scenarios and the origin. For scenarios BG, MG and AG, a value of 1.09, 0.40 and 0.78 is obtained. Thus, scenario BG is most remote from the origin and can be considered more promising than scenario MG and AG. This analysis shows that taking renewability and circularity into account in resource efficiency analysis can lead to very different conclusions. Scenario AG is the best scenario when only the CDP* is included, while according to the REU analysis, scenario BG seems the most interesting. In the next section, the CDP* and RF values for the C123-scenarios will be compared to the reference scenarios.

3.4. Comparison C123-scenarios with reference scenarios

First, care must be taken when comparing C123-scenarios with the reference scenarios, because they do not have the same TRL. The C123-scenarios have a TRL of 4, while the references are at TRL 9. For example, no heat integration is applied yet in the preliminary process concept. Further research is needed to accomplish this. However, at first sight, scenarios BG and MG have a lower CDP* than the reference scenarios (6 and 14% versus 19 and 29%). In contrast, scenario AG has a higher CDP* than Ref P1 and a similar CDP* than Ref P2. Moreover, Ref P1 and Ref P2 have an RF value of 0.66 and 0.29, respectively. This means that scenario BG uses more renewable resources than the references.

4. Conclusions and perspectives

In this work, renewability and circularity were integrated into resource efficiency analysis, resulting in REU analysis. The C123 process concept for 1-propanol production was selected as a case study to demonstrate this new approach. In the first step, the CEEENE method was applied at the life cycle level; at the process, process chain and plant level, exergy calculations were used to determine the exergetic efficiency of each C123-production step. Second, the REU analysis was carried out by calculating the RF- and RI-indicator to include renewability and circularity, respectively. Finally, a three-dimensional graphic was made to detect the most promising C123-scenario. The results show that all C123-production steps (at the process level) have a high exergetic efficiency, namely between 90 and 100%. However, in scenario BG, the biogas production has a very low exergetic efficiency due to the inefficient conversion of the feedstocks, such as maize silage and manure, into biogas. This biological process requires a significant exergy input. The exergetic efficiency at the process chain level is rather low (only 45–48%), except for scenario AG, as a result of the low methane conversion into propanol per pass. At the life cycle level, the largest CDP* was found for scenario AG (28%). Scenario BG has the lowest CDP* (6%) due to the inefficient biogas production. However, scenario BG emerged as the best option from the REU analysis, where renewability and circularity were also included. Thus, this study shows that it is important to include those two aspects in resource efficiency analysis.

In addition to efficiency, renewability and circularity, indicators for e.g., resource scarcity could also be included in sustainable resource management. However, this was beyond the scope of this research. Based on the REU analysis, improvements can also be recommended for the C123 process concept. Applying heat integration and renewable electricity production would increase the CDP* and RF in all scenarios. The RI will rise when the conversion of the feedstuffs into propanol per pass is higher. Furthermore, the following actions could be considered to further increase the CDP*: replacing the mono-ethanol absorption for CO₂ reduction by membrane technology and eliminating post-treatment steps in the preliminary design as much as possible. Finally, the simulation results need to be validated when the C123 process is applied at pilot or industrial scale. This is not possible at this moment, as the C123 process is at TRL 4.

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**Table 1.**

<table>
<thead>
<tr>
<th>Scenario</th>
<th>CDP*</th>
<th>RF</th>
<th>RI</th>
<th>Distance from origin</th>
</tr>
</thead>
<tbody>
<tr>
<td>BG</td>
<td>0.06</td>
<td>0.71</td>
<td>0.83</td>
<td>1.09</td>
</tr>
<tr>
<td>MG</td>
<td>0.14</td>
<td>0.38</td>
<td>0</td>
<td>0.40</td>
</tr>
<tr>
<td>AG</td>
<td>0.28</td>
<td>0.34</td>
<td>0.64</td>
<td>0.78</td>
</tr>
</tbody>
</table>

**Fig. 9.** Three-dimensional illustration of RF and RI in function of the CDP* for the calculation of the distances between the scenarios and the origin (indicated in red). In the table, all corresponding values for the CDP*, RF, RI and the distances between the scenarios and the origin are mentioned.
**CRediT authorship contribution statement**

**Jordy Motte:** Data curation, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Pieter Nachtegaele:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Mohamed Mahmoud:** Conceptualization, Data curation, Investigation, Writing – review & editing. **Hank Vleeming:** Conceptualization, Supervision, Writing – review & editing. **Joris W. Thymbart:** Funding acquisition, Supervision, Writing – review & editing. **Jeroen Poissonnier:** Supervision, Writing – review & editing. **Jo Dewulf:** Conceptualization, Methodology, Funding acquisition, 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.

**Data availability**

The data used for the exergy calculations can be consulted in the appendix.

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**Supplementary data**

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