Eco-efficient process improvement at early development stage: identifying environmental and economic process hotspots for synergetic improvement potential

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Just Accepted

ACS Publications

1 **Eco-efficient process improvement at early development stage: identifying environmen-**
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16
17 **Abstract**
18
19 We present here a new eco-efficiency process improvement method to highlight combined

20 environmental and costs hotspots of the production process of new material at a very early

21 development stage. Production specific and scaled-up results for life cycle assessment (LCA)

22 and production costs are combined in a new analysis to identify synergetic improvement po-

23 tentials and trade-offs, setting goals for the eco-design of new processes. The identified

24 hotspots and bottlenecks will help users to focus on the relevant steps for improvements from

25 an eco-efficiency perspective and potentially reduce their associated environmental impacts

26 and production costs. Our method is illustrated with a case study of nanocellulose. The results

27 indicate that the production route should start with carrot pomace, use heat and solvent recov-

28 ery and deactivate the enzymes with bleach instead of heat. To further improve the process,

29 the results show that focus should be laid on the carrier polymer, sodium alginate, and the

30 production of the *GripX* coating. Overall, the method shows that the underlying LCA scale-up

31 framework is valuable for purposes beyond conventional LCA studies and is applicable at a

32 very early stage to provide researchers with a better understanding of their production pro-

33 cess.

34

35

36 **Keywords**

39   **Introduction**
40

41   Life cycle assessment (LCA) has been established as an internationally accepted tool to meas-

42   ure the environmental impact of processes, products and services. As such, it is helpful in as-

43   sessing and informing the development of new materials and processes that aim to achieve a

44   more sustainable profile. During the early development stages of R&D, the degree of flexibil-

45   ity is still high and changes can be implemented at relatively low costs.[1] Throughout the vari-

46   ous phases from laboratory research to possibly mini- and then pilot-plant before finally

47   building a large-scale production plant, the incurring costs for altering the process increase

48   drastically.[2] As a consequence, it seems recommendable to define and find the right proce-

49   dures as early in the development process as possible.

50   The problems with early stage assessments are threefold: a) the lack of data;[3] b) particularly

51   for chemical processes, final large-scale production plants (machineries, reactors, pipes, etc.)

52   are not at all comparable to their respective early stage lab-scale processes. This makes it im-

53   practical to use extrapolation factors from the laboratory results to predict environmental im-

54   pacts; and c) the lab-scale process has not yet been optimized and lacks the economies of

55   scales of a production plant. An LCA of a new product under development does not therefore

56   sufficiently reflect its potential environmental impact.

57   In the literature, several studies and methods can be found that aim at integrating environmen-

58   tal impacts into process design. Sugyiama and co-workers presented a stage-gate decision

59   framework for chemical process design.[4] They rely on the assumption that energy loss (during

60   reaction and separation) is an indicator of potential environmental impacts and financial costs

61   and is composed of five weighted values, namely the presence of water, product concentra-

62   tion, min. boiling point difference, inherent waste amount and reaction energy.[5] Specifically

63   for the laboratory stage, a quick preliminary assessment of chemical processes was presented

64   based on the aforementioned stage-gate decision framework.[6] In this method, five weighted

65   economic and environmental parameters contribute to the final score. Other approaches pro-

66   posed to model lifecycle inventory data and certain LCA impact categories of chemicals by

67   only looking at the molecular structure of the target molecule.[7,8] In a review of early stage en-

68   vironmental assessment of bio-based chemicals, 33 methods were examined and categorized

69   into two groups; full assessments and early stage methods.[9] The latter was thereby further

70   broken-down into single- and multi-indicator methods. The authors concluded that the full as-

71   sessments have a broad coverage of the environmental assessment issues. However, since

72   those assessments are data intensive, it is difficult to apply them during R&D, especially at an

73   early stage. On the other hand, early stage methods offer a limited coverage but can be more

74   easily applied. It is stressed that the primary goal should be to identify critical issues as early

75   as possible and steer the R&D in the right direction.

76   None of the cited articles assessed potential environmental impacts through the modeling of a

77   scaled-up cradle-to-gate production plant of the specific lab process. Azapagic and co-

78   workers describe in their methodology how to include sustainability consideration into pro-

79   cess design during the various design stages.[10] Only in the detailed design stage is a cradle-to-

80   gate LCA of the process included. However, this takes place at a more advanced stage and no

81   indication on how the laboratory process can be translated into large-scale production is giv-

82   en. Having such a detailed LCA study already in the preliminary stage of the process design,

83   would therefore help in improving the process design.

84   Studies to determine scaling laws for LCA with empirical data of different energy equipment

85   have already been published.[11,12] The comparison of empirical data with theoretical engineer-

86   ing-based values helped to distinguish between learning and scaling effects.[13] For cases where

87   a pilot plant has been installed and an LCA study of it has been performed, a scale-up method

88   for chemical processes has been presented.[14,15] However, this requires an existing pilot plant

89   which is only built in an advanced developmental stage. In order to fill this gap and include a

90   scale-up during what would be the preliminary design stage, we developed an LCA scale-up

91   framework.[16] The framework provides an indication in the form of mathematical formulas, es-

92   timates and generic data on how a chemical laboratory process can be scaled-up for lifecycle

93   assessment studies of a large-scale production plant.

94   For a product to be successful on the market, the economic side is of decisive importance..

95   Hence, when assessing the sustainability of a material or product, the economic aspects

96   should never be neglected. To predict the costs of a product at a commercial scale while it is

97   still in the laboratory stage, similar difficulties occur as with the assessment of potential envi-

98   ronmental impact. In order to obtain a comprehensive cost estimate, a lot of detailed

99   knowledge about the production process is needed that is usually only available at an ad-

100   vanced stage. The literature offers a wide range of methods to estimate product costs. Niazi

101   and co-workers classify these into four techniques, namely the intuitive and analogical as well

102   as the parametric and analytical technique.[17] The first two are qualitative whereas the last two

103   are quantitative techniques. They propose a decision support model to decide which technique

104   should be chosen based on data quality and design stage. The qualitative techniques are used

105   in the early design stages and mostly based on past data of similar processes. For case-based

106   reasoning approaches – an intuitive technique that uses information on design, cost and time

107   of previous products – neural networks have been used to estimate the cost of new product

108   development.[18] In a recent publication, a model to simulate life cycle costs has been presented

109   that uses an algorithm that relies on similar products.[19] Other quantitative methods include

110   mathematical models for regression analysis.[20,21] The qualitative techniques on the other side,

111   are more suitable for more advanced stages as detailed information is needed. Such tech-

112   niques include methods such as operation- or activity-based approaches where a product is

113   decomposed into components.[22,23] Galli and co-workers performed an economic assessment

114   of a new lab-scale chemical production process to better understand the cost behavior with

115   differing operating conditions.[24]

116   Eco-efficiency analysis is an attempt to combine environmental and economic aspects and

117   was first introduced by Schaltegger and Sturm.[2525] The goal of this approach is to optimize a

118   process or product in both aspects, meaning that profits are maximized while the environmen-

119   tal impact minimized. Eco-efficiency analyses can be applied to technologies at the micro-

120   level or at the macro-level to explore its possible implications on wider society.[26] BASF has

121   been developing and using such an eco-efficiency approach on numerous projects by combin-

122   ing LCA with life cycle costing (LCC).[27,28] In this approach, BASF applied a normalization of

123   the LCA and LCC, which is then mostly used to inform alternative selection. For example, to

124   choose the best indigo dye production and dyeing process or to assess alternative curing and water

125   packaging systems.[29] Eco-efficiency analyses have also proven to be useful for procurement port-

126   folio optimization.[3030] Most of these examples, however, include the assessment of already exist-

127   ing processes. A more recent study applied a methodology to combine the environmental and

128   economic assessment of a new lab-scale process for eco-design purposes.[31] It helped to find the

129   optimal operative conditions while optimizing both, costs and the environmental impact. Their re-

130   sults show the importance of performing the two assessments combined as they conclude that ap-

131   plying the evaluations independently would result in different operative conditions.

132   To include a similar assessment at a very early laboratory development stage for the purpose of

133   highlighting eco-efficiency hotspots and bottlenecks for process design, the above-described eco-

134   efficiency analyses are limited. To assess potential environmental impacts at the industrial pro-

135   duction of a new material or process that is still in the laboratory research stage, a framework

136   to scale-up chemical processes for LCA studies has been developed by these authors.[16] The

137   results showed that the impact per output unit can be reduced considerably after scale-up.[32]

138   Even more importantly, it demonstrated that the contributions of the single production steps

139   can change drastically between laboratory and industrial scale. This leads to different hotspots

140   in the two LCAs. An LCA based on the lab results only could therefore lead to inefficient or

141   even wrong prioritization for process improvement. The same can be said about the costs and

142   thus the eco-efficiency analysis. This means that to improve a production process from an

143   eco-efficiency perspective at a very early stage, it is of pivotal importance to include scale-up

144   calculations.

145   In this manuscript, we present a specific method to improve the projected eco-efficiency of

146   the industrial production of a given production process even though it only exists at the labor-

147   atory scale. Our scale-up framework developed for LCA purposes is expanded to include var-

148   iable production costs. This new method actually consists of combining eco-efficiency analy-

149   sis and this expanded scale-up framework. Instead of focusing on alternative comparison and

150   selection, our new method mainly targets and recognizes synergetic improvement potentials

151   of a given process and helps therefore to set design goals by highlighting hotspots and bottle-

152   necks at a very early research stage. A case study of nanocellulose illustrates the method.

153

154   **Method for estimation of scaled-up production costs and eco-efficiency analysis**

155   A comprehensive cost estimation of a production plant includes all involved costs, which are

156   separated into capital and operating expenditures. Capital expenditure (or investment) com-

157   prises all costs that are incurred before production plus those incurred through maintaining the

158   production operable, while operating expenditure comprises all costs incurred through the

159   running of a production operation. Operating expenditures can further be separated into fixed

160   and variable costs, which are characterized by their independency from or ligation to the pro-

161   duction output, respectively. Variable costs thus comprise costs of the resources used for the

162   production, such as raw material, energy and electricity inputs. Labor can, depending on the

163   case, be part of the fixed or variable costs or even both.

164   Estimation of capital expenditure and fixed costs is difficult at such an early stage of devel-

165   opment. The information obtained from the scale-up is insufficient and these expenses are

166   highly case and site specific, i.e. they depend on the specific plant design, which normally

167   takes place after the process has been established at a more advanced developmental stage.

168   The same is true of the estimation of labor, which is why it is excluded from the variable cost

169   assessment in this paper. Including prices of capital expenditures (e.g. reactors) would also

170   mean that the longevity and production output of the plant over its entire lifespan would re-

171   quire inclusion, information that is difficult to obtain or estimate at such an early state and that

172   would go beyond the scope of the presented eco-efficiency analysis specifically designed for

173   early stage process design.

174   However, an estimation of the variable costs (without labor) is possible with the available in-

175   formation from the scale-up. Analogously to the case of the environmental impact, hotspots

176   can be identified and recommendations for improvements are possible.

177   By applying the LCA scale-up framework,[16] a lot of useful information about the process is

178   obtained beyond the fundamental purpose of LCA. Hence, the results offer the possibility to

179   include economic calculations by estimating the costs related to production. This is due to the

180   fact that the scale-up provides detailed quantitative data about each process step. With a sim-

181   ple conversion of the quantitative input and output data into their respective prices, a first es-

182   timation of the variable production costs is obtained. So far, the scale-up framework does not

183   include detailed estimation of the impact of all reactors and equipment but uses a consolidated

184   average value for the entire infrastructure. In addition to the above-mentioned reasons and in

185   order to be congruent with the environmental impact values and keep efforts of this eco-

186   efficiency simple, the capital expenditures are not added on the economic portion. If one

187   chooses to add such data, established methods can be used to estimate capital costs, such as

188   those that have been applied in other studies.[24]

189   The LCA results and such cost estimations for the production phase can then be combined to

190   an eco-efficiency analysis. We developed such an analysis that is used in combination with

191   the LCA scale-up framework and applied for process improvement at a very early laboratory

192   stage. By applying this new method at an early development stage, it helps to highlight and

193   identify hotspots for synergetic improvement potential and trade-offs and thus will influence

194   the eco-design of a new material, being the main goal of this analysis. The application of this

195   method offers a competitive advantage to yield a more sustainable product.

196

197   The relative contribution (in %) of a component (e.g. an entire process step or a specific in-

198   put) towards the overall results of the environmental impact (i.e. LCA results) is plotted

199   against its contribution (in %) to the costs. Such a plot graphically illustrates the environmen-

200   tal-to-costs-behavior of every single component. Such a component has one of three states: it

201   behaves proportionally (1), over-proportionally (2) or under-proportionally (3) in relation to

202   environmental impact towards financial costs. That behavior describes components with

203   equal, higher or lower relative LCA results compared to relative costs, respectively. The se-

204   cond group has a higher potential to reduce the environmental impact while the third group is

205   more likely suitable for cost reduction. The closer a value is to group 1, the higher is the po-

206   tential for a synergetic effect, reducing both at the same time. Therefore, components within

207   (or close to) group 1 seem to be the most interesting for improvement opportunities, as they

208   are attractive from an economic as well as an environmental perspective.

209   Focusing on proportionality alone, does not give any information about the relevance of a

210   component. Hence, a certain minimal contribution (e.g. 20 %) has to be defined for each case.

211   The minimal contribution is defined as the sum of the two contributions (i.e. LCA and costs).

212   If it exceeds that minimal contribution, it is regarded as relevant for consideration to improve

213   the process. The further a data point is located away from this relevance barrier, the higher it

214   should be prioritized for a more detailed analysis. This procedure automatically gives an equal

215   weighting factor to the potential environmental impact and financial costs. Since the aim is to

216   evaluate and improve a production process relative to its momentary state, we suggest that

217   this equal weighting for the process improvement should be used, allowing for an equal rela-

218   tive improvement of both impacts.

219   The minimal contribution has to be defined in each case according to the goals of the analysis.

220   As a default mode, we suggest to calculate it as 200 % (potential environmental impact and

221   financial costs each 100 %) divided by the number of steps/data points that are evaluated.

222   This procedure easily highlights which data points are above average.

223   The weighting of the relevance barrier can also be changed based on the preference of the as-

224   sessor who has to define the priorities. This is important to consider as there is no normaliza-

225   tion with an external reference value. A higher weighting of the environmental impact would

226   flatten the slope of the relevance barrier while a higher weighting of the costs would steepen

227   it. As a result, also the proportionality barrier would change as it is always perpendicular to

228   the relevance barrier. This also means that the distance to the proportionality barrier should be

229   considered relative to the weighting.

230   The results are best illustrated graphically (Figure 1). The proportionality barrier divides the

231   three groups, while the relevance barrier (here 20 %), running orthogonally to it, defines the

232   case specific minimal amount at which a component becomes relevant for improvement con-

233   siderations. Every component that lies on or above the relevance barrier is treated as relevant

234   whereas those that are below are regarded as not relevant in relation to the entire process. The

235   relevance barrier is therefore used to choose the components (e.g. process steps) that should

236   be prioritized when addressing improvement efforts since they bear a higher potential (due to

237   the larger contribution). Therefore, for process design aimed at improving environmental and

238   financial costs impacts simultaneously, the components towards the upper right corner (close

239   to proportionality barrier and far above the relevance barrier) allow for the greatest potential.

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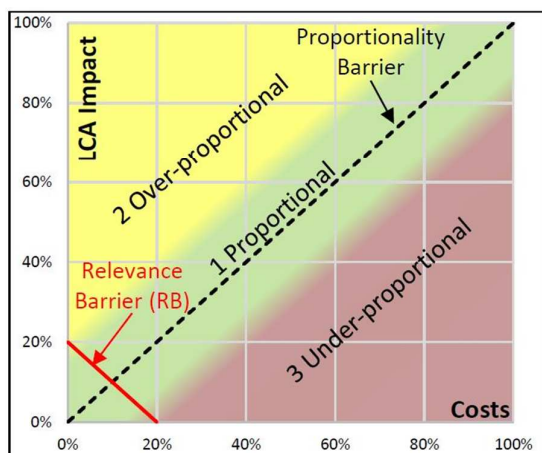
Figure 1. *Developed eco-efficiency analysis based on the scale-up results.*

Once a component has been identified, measures should be applied to improve it. As a result, the entire process should become more favorable from an eco-efficiency perspective. However, this has to be performed and monitored systematically to ensure that the targeted improvement measures do indeed lead to a more eco-efficient process. Figure 2 shows an example of the position of a process step that has been identified as a hotspot. After improvement measures have been undertaken, the point will most likely shift in this graph. This can include six different scenarios. In the ideal case, both impacts have been lowered, meaning that a clear improvement (++) has been achieved and the measures were successful. The opposite scenario would yield higher values for both impacts and thus a worsened process (--). In between these extreme scenarios, there is the possibility that one impact (e.g. LCA) has been lowered while the other (e.g. costs) increased. In such a case, it is not as clear whether an overall improvement has been achieved, as it requires the comparison of two factors that are not comparable *per se*. To be comparable, a monetary value must be given to the potential environmental impact (or vice versa), which is subject to a separate field of research. This evaluation is therefore in many cases subjective depending on the importance that an assessor gives to each potential impact. Our suggestion is to use the relevance barrier, which already includes a weighting. Based on this weighting, the indifference line which runs parallel to the relevance barrier and through the identified step should be used as threshold. Everything that lies below

262   this line (+) is therefore considered as an overall improvement of the examined step while the

263   contrary is the case above the line (-).

264   While the previous point holds true in most cases, an improved eco-efficiency of an identified

265   step does not guarantee that the process has been improved overall. Changes to that step

266   might affect (positively or negatively) other steps in the process. However, as the step has

267   been identified as a hotspot with large contribution, it is unlikely that the production process

268   as a whole is affected negatively. To assure that this case is excluded, an alternative assess-

269   ment of the entire process is performed as a last step. The difference with an alternative as-

270   sessment is that two different processes are compared on an absolute scale (i.e. points and Eu-

271   ros (EUR)). Here, a relevance barrier does not seem useful. Therefore, it is only distinguished

272   between a clear process improvement (++) or impairment (--). No conclusive recommenda-

273   tion is given for the areas in between, leaving it in the competence of the assessor.

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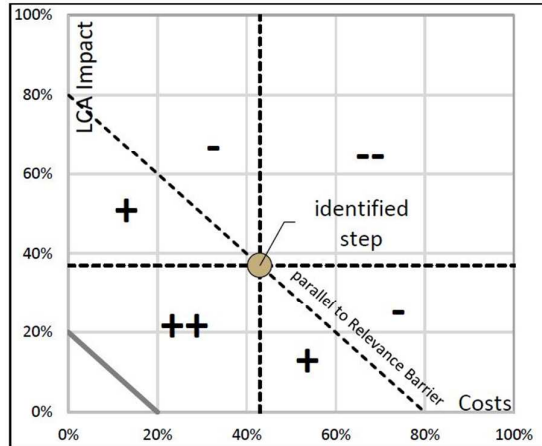
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276   **Figure 2.** *Graphical illustration of areas of improvement as well as impairment.*

277

278

279   **Case study**

280

281   The case study considers a specific nanocellulose production process that uses food waste as a

282   starting material and is still in the laboratory development stage. The process has already been

283   scaled up by the authors, using the LCA scale-up framework to perform a cradle-to-gate LCA

284   study.[32] Those data are, on the one hand, used for the assessment of potential environmental

285   impact through the here-described eco-efficiency analysis and, on the other side, serve as the

286   basis to calculate the production costs.

287   Only two production routes are compared from an eco-efficiency perspective to choose the

288   more promising in the first step. Then, this selected alternative is used to illustrate the applica-

289   tion of the eco-efficiency method. As a result, targeted recommendations are given to improve

290   the production process. After implementation of the improvements, the results are recalculat-

291   ed for comparison and validation of the measures.

292

293   **Eco-efficiency analysis and improvement potential.** All data about the scale-up and the

294   LCA impact using the ReCiPe endpoint indicator[33] are obtained from our LCA scale-up

295   study.[32] As a background database, the *ecoinvent* v3.1 with the cut-off system model was

296   used[34] and the live cycle impact assessment was calculated with the *OpenLCA* v1.4.1 soft-

297   ware. As a cradle-to-gate study, the functional unit is 1 kg of produced nanocellulose yarn

298   starting from the carrot waste source. The system boundaries for the LCA include all the steps

299   including the solvent recycling and waste treatment steps whereas the obtained by-products

300   from the enzamatic step are cut-off. A flow chart with the system boundaries can be found in

301   the supporting information. In order to estimate the costs, we used the life cycle inventory

302   (LCI) of the scaled-up nanocellucose production process. Every input was translated into its

303   purchase price using average prices. However, the outputs, such as waste, were excluded. De-

304   tails about prices and sources for chemicals, electricity and other inputs, are specified in the

305   supporting information. Two alternatives are compared that only differ in the starting materi-

306   als used as cellulosic sources, i.e. whole carrot waste and carrot pomace. The used LCA im-

307   pact data excludes the infrastructure to be in line with the costs. Furthermore, besides the ex-

308   clusion of the waste treatment, the costs have the system boundaries as the environmental

309   impact for the functional unit of 1 kg of produced nanocellulose yarn.

310   According to the above-described method, we first choose the more eco-efficient of the two

311   alternatives. Hereby, the eco-efficiency analysis clearly shows that the pomace is the prefera-

312   ble material given that it has a lower impact from both perspectives, environmental and costs

313   (Figure 3). Hence, the pomace case is chosen as the more eco-efficient alternative and will be

314   used in the following steps to improve the process itself.

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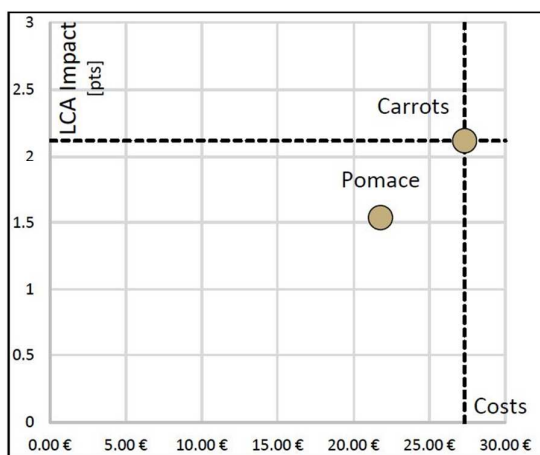
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317   **Figure 3.** *Eco-efficiency per kg of produced nanocellulose yarn comparison of alternative starting materials.*

318

319   Figure 4 displays environmental-cost-behavior of the pomace case, divided into process steps.

320   The relevance barrier was defined at 20 % with an equal weighting.

321   The analysis of the process steps reveals that only two steps are located above the relevance

322   barrier, the *GripX* production – a coating copolymer that is synthesized separately and used to

323   functionalize the cellulose – and the sodium alginate – the carrier polymer for the cellulose

324   yarn spinning. Of these, the former can be found much further from the relevance barrier and,

325   at the same time, displays an almost perfect proportionality. This makes it an ideal candidate

326   for synergetic process improvement. Sodium alginate, on the other hand, is characterized by

327   an under-proportional behavior. Therefore, the highest improvement potential can be obtained

328   through focusing on the *GripX* (5) first and then the sodium alginate (6) which is why, for the

329   next step, we focus on the improvement of the *GripX* step.
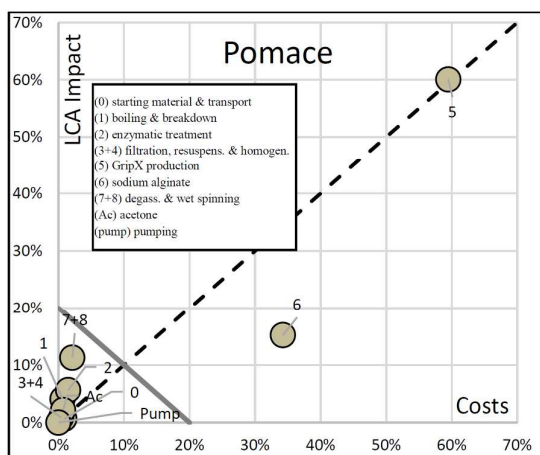
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331
332
333 **Figure 4.** *Eco-efficiency analysis of the NanoCelluComp technology divided into process step.*
334

335

336 In order to improve the *GripX* production step, an eco-efficiency analysis of this step split into

337 inputs sources is performed (Figure 5). Here the picture is clear that the materials used in the

338 process contribute almost exclusively to the entire impact of both. When looking more closely

339 into the production process this becomes more evident as vast amounts of solvents, especially

340 ethanol with 1000 l/batch and dimethyl sulfoxide (DMSO) with 500 l/batch, are processed

341 and disposed of afterwards. As a logical improvement step, the amount of these materials

342 must be reduced. As these solvents are not part of the product but only used as processing ma-

343 terials, the implementation of measures to regain and reuse them appears to be feasible.

344 Therefore, a distillation of the solvent waste seems to be a good option, although this will re-

345 quire additional heating energy.

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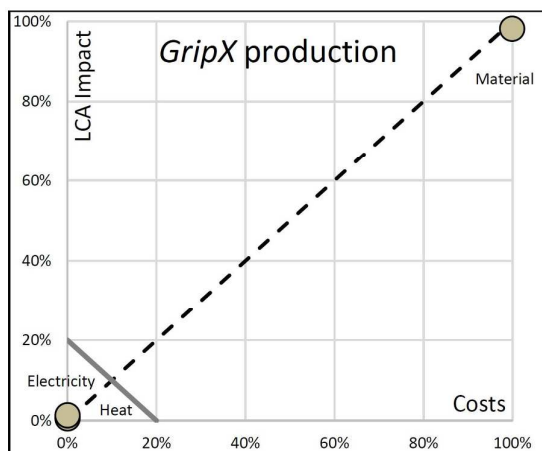
Figure 5. *Eco-efficiency analysis of the GripX production (5) step divided into input sources.*

The distillation has been included and the results recalculated (see supporting information for further specifications). Figure 6 shows the shift of step 5 after the improvement measure. The values are left relative to the overall impacts of the process without distillation for comparability reasons. Including the distillation has the effect that the *GripX* production comes closer to the barrier and switches from a proportional to clearly over-proportional behavior. It is also visible that, based on the assumptions, all the other steps are not influenced by this measure. Sodium alginate is now the component with the largest distance to the relevance barrier. This distance is mainly due to the large cost contribution, which is evidenced by the high degree of under-proportionality. Since step 5 is clearly improved from both perspectives and none of the other steps are affected, it is obvious that the overall impact of the entire process has been improved from an eco-efficiency perspective. This would make the last step, the alternative comparison before and after the measure, unnecessary. However, for the sake of completeness and illustration purposes, an alternative comparison is performed here to verify whether an eco-efficiency improvement has occurred. As anticipated, Figure 7 shows that after the inclusion of the distillation the eco-efficiency of the entire process has been improved.
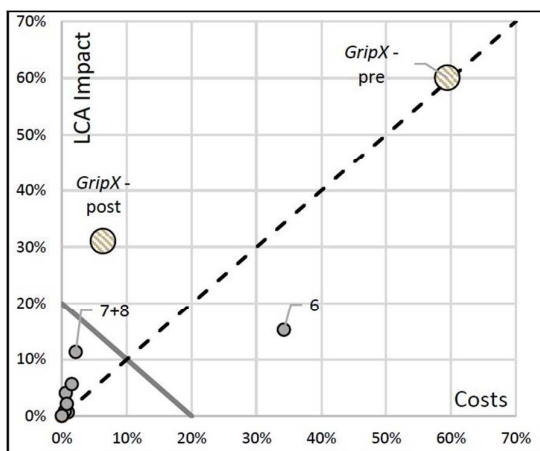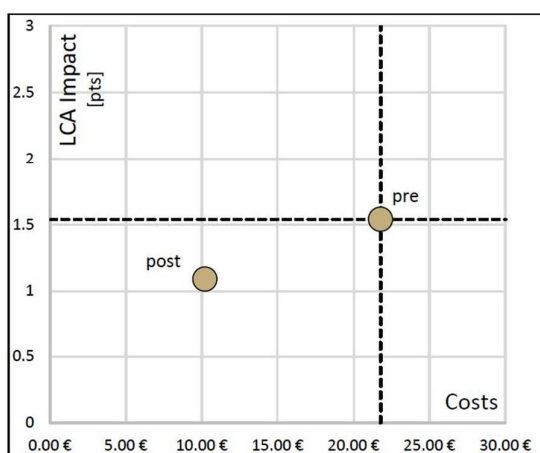
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366
367 **Figure 6.** *Eco-efficiency shift of the GripX production step after (post) inclusion of distillation.*

368

369 **Figure 7.** *Eco-efficiency per kg of produced nanocellulose yarn comparison before (pre) and after (post) inclu-*
370 *sion of distillation.*
371

372

373   The presented procedure could now potentially be applied again for further process improve-

374   ment with the sodium alginate (6) and the *GripX* (5) steps being the most likely to be ad-

375   dressed.

376   With solvent recovery, sodium alginate becomes a main cost factor in the yarn production.

377   Given that the costs for the microfibrillated cellulose (MFC) production (steps 1-5) are signif-

378   icantly lower, spinning a yarn with a higher MFC/alginate ratio would result in lower costs.

379   However, this is restricted by the technical feasibility of altering that ratio. Also, a higher ratio

380   would mean a larger amount of *GripX* needed, as its quantity is coupled to the MFC, ultimate-

381   ly resulting in a greater potential environmental impact.

382

383   While the above-described case was used as an illustrative example using two of the calculat-

384   ed scenarios (i.e. 1P and 3P), the scaled-up nanocellulose production process has already been

385   simulated and investigated from an LCA perspective using various additional production

386   route scenarios.[32] Therefore, we applied the eco-efficiency analysis to compare all the scenar-

387   ios and select the most promising one, of which, briefly, recommendations about the im-

388   provement potential are given. Details about the translation into costs of the different scenari-

389   os (Table 1) can be found in the supporting information.

390

391   **Table 1.** *Explanation of the different nanocellulose production systems. Adapted from Piccinno, F.; Hischier, R.;*
392   *Seeger, S.; Som, C. Predicting the environmental impact of a future nanocellulose production at industrial scale:*
393   *Application of the life cycle assessment scale-up framework. Journal of Cleaner Production 2018, 174, 283–*
394   *295).[32] Copyright 2018 Elsevier.*

Name	Starting Material	Enzyme Deactivation	Distillation*	Heat recov. from steps	Acetone scenario
1 C	Waste of whole carrots	heat	-	-	Emission to air
2 C	Waste of whole carrots	ClO_2	-	-	Emission to air
3 C	Waste of whole carrots	heat	68/95 %	-	Emission to air
4 C	Waste of whole carrots	heat	68/95 %	1 & 2	Emission to air
5 C	Waste of whole carrots	ClO_2	68/95 %	1	Emission to air
1 P	Carrot pomace waste	heat	-	-	Emission to air
2 P	Carrot pomace waste	ClO_2	-	-	Emission to air
3 P	Carrot pomace waste	heat	68/95 %	-	Emission to air
4 P	Carrot pomace waste	heat	68/95 %	1 & 2	Emission to air
5 P	Carrot pomace waste	ClO_2	68/95 %	1	Emission to air
6 P	Carrot pomace waste	ClO_2	68/95 %	1	No acetone used
7 P	Carrot pomace waste	ClO_2	68/95 %	1	Combustion of acetone vapor

*recycling rates of solvents: H_2O and AcOH 68 %; DMSO and EtOH 95 %

395

396   A comparison of the various scenarios shows that the pomace cases with the solvent recovery

397   (3P–7P) are clearly favorable (Figure 8) from an eco-efficiency perspective as both impacts

398   are clearly improved. The inclusion of heat recovery (4P–7P) and deactivation of the enzymes

399   with ClO_2 (5P–7P) instead of heat further improves the process. The difference between the

400   three most promising scenarios (5P–7P) lies only in the handling of the acetone for solvent

401   exchange while spinning the yarn. However, although it is the least favorable of those three,

402   the system where the acetone vapor is burned (7P) seems to be the most realistic for imple-

403   mentation. Also the heat recovery from the acetone burning step is not considered, which

404   would improve the results slightly. Therefore, the eco-efficiency analysis for the process im-

405   provement recommendations is performed with system 7P.

406
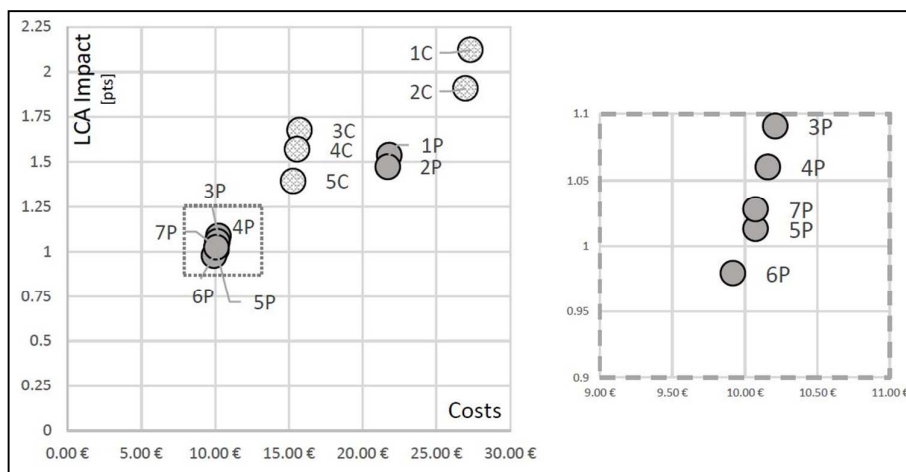
407

408   **Figure 8.** *Eco-efficiency alternative comparison of nanocellulose production process routes per kg nanocellu-*
409   *lose yarn.*
410

411   The separation by source clearly shows that the material accounts for the greatest contribution

412   (Figure 9). It displays an under-proportional behavior, hence its contribution to the potential

413   environmental impact is lower in comparison to financial costs. Reducing the material con-

414   sumption through various measures (e.g. recycling, higher yield, etc.) could have a large ef-

415   fect from a cost perspective, while still having a meaningful reduction of the potential envi-

416   ronmental impact. However, if cost reduction should be targeted through the exchange of a

417   material with an alternative of a cheaper unit price, this might cause a higher potential envi-

418   ronmental impact (depending on the material), changing the results of this eco-efficiency

419   analysis. This is the reason why a before-after comparison is always recommendable. Heat

420   and electricity are the other two impact sources regarded as relevant. Both behave over-

421   proportionally, especially in the case of heat. The use of less heating energy would thus main-

422   ly result in a relevant reduction of the potential environmental impact. The impact of the elec-

423   tricity consumption only surpasses the relevant state by a small amount.

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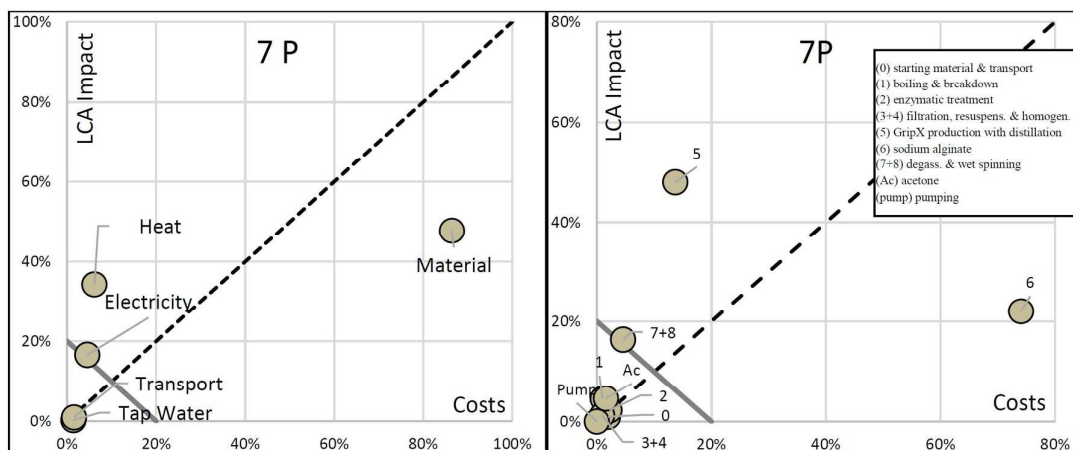
Figure 9. *Eco-efficiency analysis of the NanoCelluComp technology divided into source (left) and process steps*
428 *(right).*

429

430 The analysis of the process steps reveals that sodium alginate (6), as well as the *GripX* pro-

431 duction (5), are the two components with the highest relevance. However, the respective pro-

432 portionalities are not ideal, which limits the synergetic optimization potential of the single

433 process steps. The combination of both steps results in a very high synergetic potential given

434 that one behaves clearly under-proportional, while for the other the opposite is the case. The

435 alginate's distance to the relevance barrier is mainly due to the large cost contribution deriv-

436 ing from its high purchase price, evidenced by the high degree of under-proportionality. The

437 opposite is the case for the over-proportional *GripX* where the cost contribution has been low-

438 ered considerably thanks to the distillation. However, its contribution to the potential envi-

439 ronmental impact stays relatively high due to the heating involved. The third relevant process

440 step, that just surpasses the relevance barrier, is the spinning and degassing step (7+8). The

441 over-proportional behavior is explained by the high electricity use within this step.

442 Overall, the highest improvement potential of system 7P based on the analysis can therefore

443 be obtained through first focusing on sodium alginate (6) and *GripX* (5) together and to a

444 lesser extent, the degassing and spinning step (7+8).

445   Possible measures that should be investigated to improve the impact of the alginate include

446   replacing it with a different carrier polymer, improving the yield, buying in larger amounts to

447   reduce the purchase price and, as mentioned above, spinning at higher MFC/alginate ratio.

448   The impact of the *GripX* production might be reduced by replacing the solvents with other

449   more environmentally-benign alternatives, regaining the heat from the distillation (not yet in-

450   cluded in the calculations) and improving the yield. In contrast to the sodium alginate meas-

451   ure, a lower MFC/alginate spinning ratio would improve the impact caused by the *GripX* it-

452   self. Therefore, if technically feasible, the state after the improvement has to be compared as a

453   whole to see whether changing the ratio in any direction makes sense at all.

454   Lastly, the spinning and degassing step might be improved through higher yields and re-

455   placement of the wet spinning with a different spinning technique.

456

457   **Discussion**

458   The case study demonstrates that the here presented new method for eco-efficiency analysis at

459   very early development stages can provide highly valuable information to inform the prioriti-

460   zation of possible approaches for process improvement. The scale-up framework behind it is

461   crucial in predicting the potential environmental impact and financial costs of the production

462   process at a large scale. One of its advantages lies in the fact that new hotspots might be iden-

463   tified that would otherwise, by only looking at the lab-scale process, been missed. However,

464   the scale-up framework for a theoretically scaled-up and predicted production plant has an in-

465   fluence on the quality of the data and leads to some degree of uncertainty. Hence, the results

466   of this method should rather be regarded as indicators than as exact values, meaning that if

467   two data points lie close to one another then they should be regarded as equal for the here de-

468   scribed procedure.

469   While the method does include certain aspects of BASF's method, such as the two-

470   dimensional plotting of costs versus LCA impact, by graphically illustrating the results, it is

471   clear how our analysis differs. Our method focuses on the improvement of the production

472   process, i.e. from cradle to gate only, meaning that no use phase or end-of-life consideration is

473   included for the assessment of potential environmental impact or financial costs. This differs

474   from other methods as they used life cycle costs or purchase prices as well as the full LCA

475   where possible.[29] Furthermore, our method includes a procedure on how to obtain the required

476   environmental and costs data but does not contain a normalization factor for these results.

477   The eco-efficiency analysis is specifically designed to be used with the scale-up data at an

478   early development stage. Although it could also be applied without a scale-up on an existing

479   process, we do not see its advantage over other eco-efficiency analyses in this regard. Hence,

480   we strongly suggest to use it with the scale-up as that is where it differentiates itself from oth-

481   er eco-efficiency analyses and can provide additional insights. The application of the present-

482   ed eco-efficiency method without performing the scale-up procedure, i.e. using the lab values,

483   could lead to inefficient improvement recommendations. This has already been shown with

484   the data in the LCA scale-up study of the same case where the relative contributions of the

485   single steps have shifted considerably.[32] The same is true for the scaled-up variable costs and

486   ultimately the eco-efficiency analysis. For that reason, the same limitations that apply to the

487   scale-up framework are also limiting factors for this eco-efficiency method. Those include the

488   limitation to only certain types of processes as well as the uncertainty of predicting future

489   processes.

490   One key advantage over many other methods is that our method provides data with a higher

491   degree of detail at such an early stage. Although it can be applied at a very early stage, i.e. the

492   preliminary design stage according to Azapagic and co-workers,[10] the results in terms of de-

493   gree of detail can be positioned between the early stage and full assessment methods.[9] Also,

494   the costs are obtained without using a separate method to the LCA assessment, meaning that

495   the scale-up is useful for both, resulting in a more straightforward procedure. However, the

496   cost calculation is very simple and limited to considering only variable costs. Expansion of

497   the costs modeling through inclusion of fixed costs and capital expenditures – possibly

498   through combination with existing and established methods – could be a goal for further im-

499   provement. However, one needs to bear in mind that this would result in greater complexity as

500   well.

501   The described analysis provides additional insights and information when compared to a

502   study focusing on the LCA results or costs only. This is not as evident in the here presented

503   case study as the results for improvement recommendations are similar to the LCA scale-up

504   study. However, even in this study, the advantage can be seen. When comparing the LCA im-

505   pact of sodium alginate (6) with the degassing and spinning (7+8) step for the 7P scenario, the

506   values are close to each other making it difficult to identify a clear favorite (especially when

507   uncertainty of the results is considered). However, as soon as the eco-efficiency is assessed

508   the relevance of the sodium alginate becomes much greater and should therefore be priori-

509   tized.

510   We deliberately chose not to include any normalization calculations linked to external data

511   (e.g. environmental impact of a geographical area) as this constitutes a different field of re-

512   search and compromises the results. In our opinion, at this stage the relevance of the impacts

513   is best judged by the assessor him or herself. As it would be reasonable to include a normali-

514   zation step for the alternative selection to put the impacts into relation, this is not necessarily

515   true for the process improvement analysis, which constitutes the core part of this method. As a

516   new process or material – making it difficult to find a normalization reference (especially for

517   the costs) –, we mainly see the contribution of the single steps towards the entire process to be

518   the most relevant part. The advantage hereby is that an optimization from the point of the as-is

519   state is achieved.

520   A broader range of real-case applications of the method will hopefully help to further refine it

521   in future by pointing out its limitation.

522   The presented process improvement eco-efficiency analysis method for process improvement

523   should be applied whenever an LCA scale-up study is performed. Only the addition of the

524   economic perspective allows to obtain a more holistic picture and understanding of the pro-

525   duction process and helps in setting design goals towards more eco-efficiency and, thus, sus-

526   tainability. Given that all this can be achieved with minor efforts once a scale-up study has

527   been performed, our method is a valuable additional step for process eco-design at a very ear-

528   ly stage already.

529   **Supporting Information**

530   Background data for the various production systems and the cost calculations can be found in

531   the Supporting Information document.

532

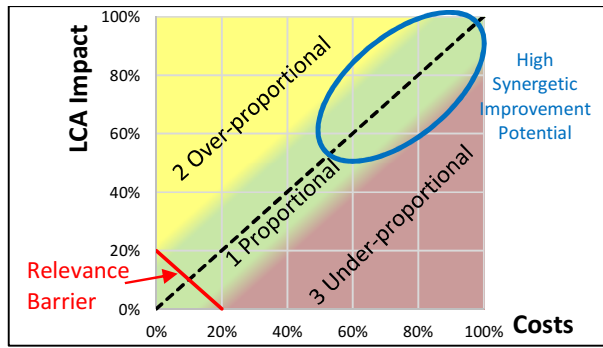533   **Funding Sources**

536

537   **Acknowledgements**

541

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640   TOC/abstract art

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