Life Cycle Assessment Framework To Support the Design of Biobased Rigid Polyurethane Foams

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ABSTRACT: A methodological framework implementing laboratory activities and life cycle assessment is presented and applied to determine which parameters should be considered to develop biobased rigid polyurethane foams for thermal insulation with improved environmental performances when compared to their fossil counterparts. The framework was applied to six partially biobased (produced from bio-based polyols obtained from azelaic acid and/or lignin) and one fossil-based formulations. A comprehensive set of impact assessment categories was investigated including uncertainty and sensitivity analysis. Results proved that physical characteristics such as thermal conductivity and density are the most important variable to be optimized to guarantee better environmental performances of biobased polyurethane rigid foams for thermal insulation. Care should be taken with reference to ozone depletion potential, marine eutrophication, and abiotic depletion potential because of the uncertainty related to their results. The methylene diphenyl disocyanate and foam production process were identified as the major sources of impacts. Overall environmental superiority of biobased polyurethanes cannot always be claimed with respect to their fossil counterpart.

1. INTRODUCTION

The scarcity of natural resources and environmental impacts related to the overexploitation of fossil fuels is central to international debate. The United Nations (UN), in 2015, adopted an agenda for the development of more sustainable production processes also known as “Goal 12” of the Sustainable Development Goals (2030 Agenda) by setting specific targets to support the circulation of new materials based on renewable feedstock with improved environmental performances. On the same line, the EU commission has launched the circular economy action plan “Closing the loop” aimed at developing an economy based on recycling, recovery, and reuse with clear attention to the management of natural resources. Among the provisions derived from this action plan, the limitation on the production and marketing of single-use plastic and of the use of nonrenewable resources raised the interest of the market and is considered fundamental for the reduction of negative environmental impacts. The reasons for this interest are manifold: plastics play a fundamental role in our society, with application in almost all areas of our daily life; however, the actual production system and market presents several environmental issues ranging from low recycling rates and high landfilled fraction, without neglecting the dispersion in oceans and the use of hazardous additives.

In this context, the development of new plastic materials with better environmental performances when compared to the traditional fossil-based counterpart is considered a priority. The research community has widely investigated the use of renewable feedstock for the production of biobased plastics so that material such as poly(lactic acid), poly(hydroxyalkanoates), starch plastics, biobased polyethylene (PE), partly bio-based polyethylene terephthalate (PET), and partly biobased polyurethane (PU) are already established in the market.

Despite that the use of biomass is considered to be promising in plastic production because of its renewability and its relatively low price, recent studies based on the application of life cycle assessment (LCA) methodology proved that such materials cannot always guarantee better environmental performances when compared to their fossil counterparts. LCA, as defined by ISO 14040 standard, is the compilation and evaluation of the inputs, outputs, and potential environmental impacts of a product system throughout its life cycle, allowing a comprehensive view on the related potential impacts. Tsioropoulos et al. presented a comparison between biobased high-density polyethylene (HDPE) and biobased PET with the fossil-based counterparts, reporting that biobased materials guarantee to save in nonrenewable energy use and had smaller greenhouse gases (GHG) emissions, but on the other hand increased the net water consumption and emission of eutrophic substances associated with the application of fertilizers during biomass cultivation. Chen et al. provided a...
Comparative analysis between fossil-based PET and PET containing biomass from wood and corn residues investigating climate change, fossil resource depletion, acidification, and ozone depletion impact assessment categories. Formulations containing biomass scored better environmental performances in climate change and fossil resource depletion but they showed higher impacts in acidification and ozone depletion. Semba et al. and Adom et al. analyzing biobased PE, succinic acid, isobutanol, and PET, confirmed that the use of renewable feedstock generally improved the environmental performances of biobased plastics in the climate change impact assessment categories. Helling and Russell focused their assessment on water withdrawals related to different polyols, showing a higher consumption for biobased ones and also a great variability between different feedstocks.

Despite the great attention paid to the investigation of environmental performances of biobased plastics, no study related to the LCA of biobased PU foams has been published yet, neither studies on their comparison with fossil-based PU foams. From the literature review, it emerges that research on biobased PU foams has been more focused on the development of new formulations based on the use of vegetable oil- or lignin-derived polyols.

Considering that PU foams are used in a wide range of applications and that it is the fifth most demanded polymer in Europe (3.74 million tons), to guarantee the sustainability of PU industries in the context of the framework of the circular economy, there is a clear need to investigate the environmental performances of new biobased PU formulations and identify potential pathways to reduce their potential negative environmental impacts.

In light of this lack, the objective of this study, through the development of a framework integrating laboratory testing and LCA, is to determine which parameters should be considered to develop biobased PU foams with improved environmental performances when compared to their fossil counterparts. The biobased foams under study have been prepared after extensive formulation optimization to achieve physical performances at least equal to the fossil-based reference material.

Considering the wide range of applications of PU polymers, this study focuses on PU rigid foams used as the building insulation material. These foams have been prepared starting from biobased polyols, obtained from azelaic acid (AA) and/or lignin. Seven PU formulations were developed and investigated in this study: T01 is based on traditional fossil polyol, B01/B02/B03 are based on AA derived polyol, and BL01/BL02/BL03 are based on AA-derived polyol and contain liquified Kraft lignin from softwood.

For the assessment of the environmental performances, considering the conflicting results presented by other references published on other bio-based plastic materials (e.g., climate change vs eutrophication), a comprehensive set of indicators has been used.

### Table 1. Values of the Parameters for Each Formulation

| parameter       | unit         | T01           | B01           | B02           | B03           | BL01          | BL02          | BL03          |
|-----------------|--------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|
| AAperc          | %            | 0.00 × 10^2   | 6.90 × 10^1   | 6.90 × 10^1   | 7.80 × 10^1   | 6.90 × 10^1   | 7.80 × 10^1   | 7.80 × 10^1   |
| polyol mass     | kg           | 1.00 × 10^-3  | 1.00 × 10^-3  | 7.00 × 10^-3  | 5.00 × 10^-2  | 7.40 × 10^-2  | 5.30 × 10^-2  | 1.10 × 10^-1  |
| glycerine ratio | %            | 0.00 × 10^-6  | 0.00 × 10^-6  | 4.30 × 10^-1  | 1.00 × 10^-6  | 4.30 × 10^-1  | 1.00 × 10^-6  | 1.00 × 10^-6  |
| lignin perc     | %            | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 1.90 × 10^-6  | 1.90 × 10^-6  | 1.00 × 10^-6  |
| sulfuric mass   | kg           | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 3.00 × 10^-3  | 1.50 × 10^-3  | 0.00 × 10^-6  |
| NaOH mass       | kg           | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 6.00 × 10^-6  |
| raw material distance | km | 1.00 × 10^1 | 1.00 × 10^1 | 1.00 × 10^1 | 1.00 × 10^1 | 1.00 × 10^1 | 1.00 × 10^1 | 1.00 × 10^1 |
| liquefaction energy | W h | 0.00 × 10^-6 | 0.00 × 10^-6 | 0.00 × 10^-6 | 0.00 × 10^-6 | 0.00 × 10^-6 | 0.00 × 10^-6 | 0.00 × 10^-6 |
| reaction_PandL  | kg           | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  |
| reaction_MDl    | kg           | 1.60 × 10^-1  | 1.80 × 10^-1  | 3.70 × 10^-1  | 4.40 × 10^-1  | 1.50 × 10^-1  | 3.00 × 10^-1  | 4.00 × 10^-1  |
| reaction_additives | kg | 8.00 × 10^-3  | 7.70 × 10^-3  | 1.10 × 10^-3  | 1.20 × 10^-2  | 8.70 × 10^-3  | 7.60 × 10^-3  | 7.70 × 10^-3  |
| PU_density      | kg/m³        | 4.20 × 10^-2  | 4.60 × 10^-2  | 3.60 × 10^-2  | 3.80 × 10^-2  | 3.50 × 10^-2  | 4.10 × 10^-2  | 3.10 × 10^-2  |
| PU_lambda       | W/(K m)      | 2.70 × 10^-2  | 2.60 × 10^-2  | 2.60 × 10^-2  | 2.80 × 10^-2  | 3.50 × 10^-2  | 2.90 × 10^-2  | 2.70 × 10^-2  |
| waste_distance  | km           | 1.00 × 10^1   | 1.00 × 10^1   | 1.00 × 10^1   | 1.00 × 10^1   | 1.00 × 10^1   | 1.00 × 10^1   | 1.00 × 10^1   |
| glycerine mass  | kg           | 0.00 × 10^-6  | 0.00 × 10^-6  | 3.00 × 10^-5  | 5.00 × 10^-5  | 3.20 × 10^-5  | 5.30 × 10^-5  | 1.10 × 10^-5  |
| lignin mass     | kg           | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 0.00 × 10^-6  | 1.90 × 10^-6  | 1.70 × 10^-6  | 1.50 × 10^-6  |
| raw material transport | t km | 1.00 × 10^-1  | 1.00 × 10^-1  | 7.00 × 10^-5  | 5.00 × 10^-5  | 9.30 × 10^-5  | 7.00 × 10^-5  | 1.50 × 10^-5  |
| produced_PandL  | kg           | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  | 1.00 × 10^-1  |
| PU_produced     | kg           | 2.70 × 10^-3  | 2.90 × 10^-3  | 4.80 × 10^-3  | 5.50 × 10^-3  | 2.50 × 10^-1  | 4.10 × 10^-1  | 5.00 × 10^-1  |
| PU_required     | kg           | 1.20 × 10^-1  | 1.20 × 10^-1  | 9.40 × 10^-3  | 1.10 × 10^-3  | 1.20 × 10^-3  | 1.20 × 10^-3  | 8.40 × 10^-3  |
| waste_transport | t km         | 1.20 × 10^-1  | 1.20 × 10^-1  | 9.40 × 10^-3  | 1.10 × 10^-3  | 1.20 × 10^-3  | 1.20 × 10^-3  | 8.40 × 10^-3  |
derived polyols and BL series contain lignin-based polyols). The reason of these results is the larger amount of reactants required to produce B01, due to its slightly higher density, B03 and BL02, due to their slightly higher conductivity, and BL0, due to its higher conductivity and density.

The B02 formulation, on the other hand, scores the best performances with the following environmental impact reductions when compared to the performances of the fossil-based formulation (T01): CC (−30%), ODP (−14%), POF (−31%), AE (−28%), FE (−44%), ME (−24%), ADPFF (−31%) and ADP (−8%). Finally, BL03 resulted to have higher impacts with respect to T01 in the ODP (+10%) category but have lower impacts in the others: CC (−32%), POF (−33%), AE (−29%), FE (−43%), ME (−11%), ADPFF (−36%), and ADP (−16%).

Results of the contribution analysis (Table 2) allowed the identification of the life cycle stages that mostly influenced the impact assessment results.

The CC, POF, AE, and ADPFF impact assessment categories resulted to be strongly influenced by the use of nonrenewable resources related to the application of MDI in all of the biobased PU formulations and the use of fossil-based polyol in the case of T01. In these categories, considering that the use of MDI increased per unit of polyol (from an MDI/polyol ratio of 1.64 for T01 to 4.40 for B03 and 3.97 for BL03, depending on the nOH of the polyol), the bio-based formulations scored performances similar to those of T01.

The ODP, FE, and ADP categories resulted to be more influenced by the impacts deriving from “foam production” and “polyol production” life cycle stages. In these cases, considering that impacts were directly proportional to the quantity of materials, formulations with better physical properties resulted to be favored.

The ME category resulted to be more influenced by the impacts deriving from end of life management (in particular the disposal in landfill) followed by the production process and use of aniline in the MDI’s production process. Also, in this case, the reason of such results was directly related to the physical properties of the PU formulations.

Focusing on the comparison between the biobased PU formulations that presented overall better performances, B02 and BL03, the introduction of lignin (considering both the

Figure 1. Results of the life cycle impact assessment. All the results are referred at the functional unit.
production and liquefaction processes) does not make the polyl more environmentally sustainable than the one derived from AA. This trend is confirmed by the contribution analysis; the overall contribution of polyl, lignin, and liquefaction to the impacts of BL03 is higher than the contribution of polyol in B02 in all of the impact assessment categories considered.

Focusing on a wider analysis of the literature, in one hand, the results of this study partially confirm those already published. This is the case of the reduction of impacts guaranteed by biobased formulations in the CC category which are consistent with the results obtained by Tsiropoulos et al.\(^9\) (bio-HDPE and bio-PET), Adom et al.\(^{12}\) (bio-PE, succinic acid and isobutanol), Semba et al.,\(^{11}\) and Chen et al.\(^{10}\) (bio-HDPE and bio-PET). Results in the ADPFF category confirm the conclusion of Chen et al.\(^{10}\) and Adom et al.\(^{12}\).

On the other hand, the increase in impacts in ODP and AE reported by Chen, et al.\(^{10}\) and in the case of ME reported by Tsiropoulos, et al.\(^9\) are not confirmed in this study. This discrepancy is due to the different functional units adopted; in those studies, in fact, impacts were reported per unit of mass without considering the physical properties (e.g., density and conductivity) which are instead fundamental for the use of PU in building insulation.

As already mentioned in Section 2, the sensitivity analysis has been performed to identify the most influencing parameters among AAperc, lignin perc, reaction_PandL, reaction_MD1, PU_density, and PU_lambda (Table 3).

Results proved that physical properties of the foam (PU_lambda and PU_density) linearly affected the quantity of material required and therefore indirectly affected the impacts in all of the impact assessment categories. Among other parameters, only reaction_PandL and reaction_MD1 showed the relevant value of sensitivity, in particular for the impact categories ODP, FE, ME, ADPFF, and ADP. Parameters related to the formulation of PU (AAperc and lignin perc) have a small influence on the final results. Results of the study proved that the impacts, in the case of biobased PU formulation, were more influenced by the amount of foam required to satisfy the functional unit, rather than its formulation. For this reason, when developing a new formulation from renewable feedstock, it is important to achieve physical performances at least equal to the fossil-based reference material.

### Table 2. Results of the Contribution Analysis for the Formulation T01, B02, and BL03

| imp. cat. | foam | polyl [%] | lignin [%] | liquefaction [%] | MDI [%] | foam production [%] | end of life [%] |
|-----------|------|-----------|------------|------------------|--------|--------------------|---------------|
| CC        | T01  | 34        | 0          | 0                | 58     | 7                  | 1             |
|           | B02  | 5         | 0          | 0                | 85     | 8                  | 1             |
|           | BL03 | 3         | 2          | 6                | 80     | 7                  | 1             |
| ODP       | T01  | 26        | 0          | 0                | 6      | 57                 | 11            |
|           | B02  | 31        | 0          | 0                | 7      | 53                 | 9             |
|           | BL03 | 12        | 11         | 28               | 5      | 37                 | 6             |
| POF       | T01  | 32        | 0          | 0                | 49     | 18                 | 1             |
|           | B02  | 4         | 0          | 0                | 74     | 21                 | 1             |
|           | BL03 | 2         | 2          | 5                | 70     | 20                 | 1             |
| AE        | T01  | 32        | 0          | 0                | 59     | 8                  | 1             |
|           | B02  | 5         | 0          | 0                | 84     | 10                 | 1             |
|           | BL03 | 3         | 2          | 9                | 77     | 9                  | 1             |
| FE        | T01  | 44        | 0          | 0                | 10     | 45                 | 1             |
|           | B02  | 15        | 0          | 0                | 18     | 66                 | 1             |
|           | BL03 | 7         | 2          | 16               | 16     | 58                 | 1             |
| ME        | T01  | 14        | 0          | 0                | 15     | 19                 | 52            |
|           | B02  | 3         | 0          | 0                | 20     | 20                 | 56            |
|           | BL03 | 1         | 8          | 16               | 16     | 16                 | 43            |
| ADPFF     | T01  | 36        | 0          | 0                | 59     | 4                  | 1             |
|           | B02  | 5         | 0          | 0                | 89     | 5                  | 1             |
|           | BL03 | 3         | 2          | 3                | 88     | 5                  | 1             |
| ADP       | T01  | 7         | 0          | 0                | 9      | 82                 | 2             |
|           | B02  | 17        | 0          | 0                | 10     | 72                 | 1             |
|           | BL03 | 9         | 1          | 7                | 10     | 71                 | 1             |

### Table 3. Absolute Values of Sensitivity*\(^a\)

| imp. cat. | AAperc | lignin perc | reaction_PandL | reaction_MD1 | PU_density | PU_lambda |
|-----------|--------|-------------|----------------|--------------|------------|-----------|
| CC        | 1.71 × 10^{-2} | 4.07 × 10^{-3} | 6.82 × 10^{-2} | 7.29 × 10^{-2} | 1.01 × 10^{0} | 1.01 × 10^{0} |
| ODP       | 5.58 × 10^{-2} | 3.15 × 10^{-2} | 3.90 × 10^{-1} | 3.41 × 10^{-1} | 1.05 × 10^{0} | 1.05 × 10^{0} |
| POFF      | 2.61 × 10^{-2} | 8.35 × 10^{-3} | 6.23 × 10^{-2} | 6.61 × 10^{-2} | 1.00 × 10^{0} | 1.00 × 10^{0} |
| AE        | 1.97 × 10^{-2} | 2.05 × 10^{-3} | 4.42 × 10^{-2} | 5.14 × 10^{-2} | 1.00 × 10^{0} | 1.00 × 10^{0} |
| FE        | 2.85 × 10^{-2} | 2.09 × 10^{-2} | 1.57 × 10^{-1} | 1.35 × 10^{-1} | 1.00 × 10^{0} | 1.00 × 10^{0} |
| ME        | 3.43 × 10^{-3} | 4.05 × 10^{-2} | 1.68 × 10^{-1} | 1.45 × 10^{-1} | 1.00 × 10^{0} | 1.00 × 10^{0} |
| ADPFF     | 3.84 × 10^{-2} | 5.50 × 10^{-3} | 1.14 × 10^{-1} | 1.14 × 10^{-1} | 1.00 × 10^{0} | 1.00 × 10^{0} |
| ADP       | 2.26 × 10^{-2} | 1.46 × 10^{-2} | 1.15 × 10^{-1} | 9.96 × 10^{-2} | 1.01 × 10^{0} | 1.02 × 10^{1} |

*\(^a\)High values mean a greater relevance of the parameter for the indicated impact category.
Results of the uncertainty analysis are reported in (Figure 2) and (Table 4). Uncertainty, that is related at the secondary data used in this study, was higher in the formulation containing lignin (BL03), whereas T01 and B02 showed similar values of coefficient of variation (CV).

Considering the results of uncertainty analysis, impacts of B02 were confirmed to be lower than T01 (without overlap of error intervals) in the categories CC, POF, AE, and ADPFF, while impacts of BL03 were confirmed to be lower only in the CC and ADPFF. With reference to ODP, FE, ME, and ADP, due to an overlap in the 95% confidence interval, reduction in environmental impacts cannot be always confirmed.

Considering that uncertainty resulted to be influenced by the use of secondary data, the collection of primary data becomes a priority for future study and to confirm if formulations such as B02 and BL03 can guarantee environmental performance improvements when compared to T01.

### 3. CONCLUSIONS

The overall results of LCA show that formulation containing azelaic-acid derived polyol (B02) and lignin-based polyol (BL03) can score reductions of impacts in all the of the impact categories when compared to their fossil counterpart (T01). B02 presented better overall performances with a reduction of impacts between −8 and −44%. However, considering the results of uncertainty analysis, reduction in environmental impacts (absolute term) was confirmed only in the categories CC, POF, AE, and ADPFF for B02 and CC and ADPFF for BL03. Such results should be confirmed by improving the use of primary data to limit the uncertainty of results. Overall results also show that the introduction of lignin is not always

| Impact Category | Foam | Mean | Median | SD   | CV   | 2.50% | 97.50% |
|-----------------|------|------|--------|------|------|-------|--------|
| CC [kg CO₂eq]  | T01  | 4.88 × 10³ | 4.87 × 10³ | 4.47 × 10⁻² | 9.16 × 10⁻¹ | 4.80 × 10⁰ | 4.98 × 10⁰ |
|                 | B02  | 3.42 × 10³ | 3.42 × 10³ | 4.86 × 10⁻² | 1.19 × 10⁰ | 3.35 × 10⁰ | 3.51 × 10⁰ |
|                 | BL03 | 3.32 × 10³ | 3.31 × 10³ | 5.44 × 10⁻² | 1.64 × 10⁰ | 2.31 × 10⁰ | 4.44 × 10⁰ |
| ODP [kg CFC-11eq] | T01  | 6.46 × 10⁻⁴ | 6.31 × 10⁻⁴ | 1.56 × 10⁻⁵ | 2.41 × 10⁻⁵ | 3.90 × 10⁻⁴ | 9.74 × 10⁻⁴ |
|                 | B02  | 5.50 × 10⁻⁴ | 5.40 × 10⁻⁴ | 1.27 × 10⁻⁵ | 2.30 × 10⁻⁵ | 3.31 × 10⁻⁴ | 8.45 × 10⁻⁴ |
|                 | BL03 | 7.15 × 10⁻⁴ | 6.96 × 10⁻⁴ | 3.27 × 10⁻⁵ | 4.58 × 10⁻⁵ | 1.12 × 10⁻⁴ | 1.44 × 10⁻⁴ |
| POF [kg NMVOCeq] | T01  | 1.71 × 10⁻² | 1.70 × 10⁻² | 1.70 × 10⁻³ | 9.88 × 10⁻⁵ | 1.68 × 10⁻⁴ | 1.75 × 10⁻⁴ |
|                 | B02  | 1.18 × 10⁻² | 1.18 × 10⁻² | 1.46 × 10⁻³ | 1.24 × 10⁻⁴ | 1.16 × 10⁻² | 1.22 × 10⁻² |
|                 | BL03 | 1.18 × 10⁻² | 1.21 × 10⁻² | 8.97 × 10⁻⁴ | 7.63 × 10⁻⁴ | −6.32 × 10⁻³ | 2.78 × 10⁻³ |
| AE [mol H₂eq]  | T01  | 2.42 × 10⁻² | 2.42 × 10⁻² | 3.67 × 10⁻⁴ | 1.52 × 10⁻⁴ | 2.37 × 10⁻² | 2.51 × 10⁻² |
|                 | B02  | 1.73 × 10⁻² | 1.73 × 10⁻² | 3.45 × 10⁻⁴ | 1.99 × 10⁻⁴ | 1.68 × 10⁻² | 1.82 × 10⁻² |
|                 | BL03 | 1.76 × 10⁻² | 1.77 × 10⁻² | 6.66 × 10⁻³ | 3.78 × 10⁻⁴ | 4.34 × 10⁻³ | 2.95 × 10⁻³ |
| FE [kg P₄O₁₀]  | T01  | 6.06 × 10⁻⁴ | 5.60 × 10⁻⁴ | 1.91 × 10⁻⁵ | 3.16 × 10⁻⁵ | 3.72 × 10⁻⁴ | 1.15 × 10⁻⁴ |
|                 | B02  | 3.39 × 10⁻⁴ | 3.00 × 10⁻⁴ | 1.67 × 10⁻⁵ | 4.92 × 10⁻⁵ | 1.49 × 10⁻⁴ | 7.88 × 10⁻⁴ |
|                 | BL03 | 3.44 × 10⁻⁴ | 3.07 × 10⁻⁴ | 1.52 × 10⁻⁵ | 4.41 × 10⁻⁵ | 1.65 × 10⁻⁴ | 7.52 × 10⁻⁴ |
| ME [kg N₂eq]   | T01  | 1.53 × 10⁻² | 1.46 × 10⁻² | 3.73 × 10⁻³ | 2.44 × 10⁻³ | 1.02 × 10⁻² | 2.43 × 10⁻² |
|                 | B02  | 1.15 × 10⁻² | 1.09 × 10⁻² | 3.01 × 10⁻³ | 2.61 × 10⁻³ | 7.34 × 10⁻⁴ | 1.91 × 10⁻² |
|                 | BL03 | 1.35 × 10⁻² | 1.34 × 10⁻² | 4.45 × 10⁻³ | 3.29 × 10⁻³ | 5.12 × 10⁻³ | 2.24 × 10⁻² |
| ADPFF [MJ]     | T01  | 9.53 × 10¹  | 9.53 × 10¹  | 5.80 × 10⁻¹ | 6.09 × 10⁻¹ | 9.43 × 10⁻¹ | 9.66 × 10⁻¹ |
|                 | B02  | 6.55 × 10¹  | 6.54 × 10¹  | 7.04 × 10⁻¹ | 1.08 × 10⁰ | 6.43 × 10⁻¹ | 6.69 × 10⁻¹ |
|                 | BL03 | 6.09 × 10¹  | 6.08 × 10¹  | 6.84 × 10⁻¹ | 1.12 × 10⁰ | 5.97 × 10⁻¹ | 6.25 × 10⁻¹ |
| ADP [kg Sb₄eq] | T01  | 6.16 × 10⁻⁶ | 5.60 × 10⁻⁶ | 2.89 × 10⁻⁶ | 4.68 × 10⁻⁶ | 2.71 × 10⁻⁶ | 1.32 × 10⁻⁵ |
|                 | B02  | 5.74 × 10⁻⁶ | 5.13 × 10⁻⁶ | 2.60 × 10⁻⁶ | 4.53 × 10⁻⁶ | 2.60 × 10⁻⁶ | 1.26 × 10⁻⁵ |
|                 | BL03 | 5.45 × 10⁻⁶ | 5.16 × 10⁻⁶ | 3.58 × 10⁻⁶ | 6.58 × 10⁻⁶ | −6.46 × 10⁻⁷ | 1.33 × 10⁻⁵ |
preferable to the use of only AA-derived polyol. In addition, lignin-containing foams are characterized by greater uncertainty.

For a further improvement of the environmental performances, a reduction of the MDI/polyol ratio in the biobased formulations is desirable (i.e., use of low nOH polyols is preferable), while the significant contribution of the fossil polyol justifies the efforts made to replace it with a renewable counterpart. Impacts related to the foam production process are strictly related to the quantity of foam required, for this reason, obtaining adequate physical performance is essential, in particular, lowest thermal conductivity at the lowest possible density.

The sensitivity analysis proved that the density and conductivity of the foam are the key parameters that influence the overall impacts. This is further confirmation that an introduction of renewable feedstock end in itself is not sustainable if not supported by the achievement of physical properties similar to fossil-based foams.

The results of this study confirm those presented in the literature for the categories CC and ADPFF, whereas for the others categories (ODP, AE, and ME), the results are only partially verified. In fact, the expression of the functional unit according to the insulating properties of the foam highlights that the improving of physical performance lead to a reduction in the required foam amount to be used, which is reflected in a proportional decrease in impacts.

Results of this study support the development of future research, indeed there is a clear need to improve the quality of data related to the production of biomass used as feedstock to confirm the results scored by biobased PU in ODP, FE, ME, and ADP categories.

4. EXPERIMENTAL SECTION

4.1. General Methodological Framework. To investigate the objective of the study, the general structure of LCA studies has been adapted to better integrate the laboratory activities implemented in the development of the new biobased PU formulas.

LCA studies are structured in 4 phases. The goal and scope definition are implemented to clarify the objective of the study, to determine the main methodological boundaries (e.g., impact assessment categories to be investigated and data quality criteria for data collection) as well as the life cycle processes to be included in the analysis (also referred as system boundaries). Another fundamental step of this phase is the definition of the so-called functional unit which is the measuring unit that quantifies the function of the product under study. Its definition is very important to allow for comparisons of different products with similar functions (e.g., PU rigid foams based on fossil or biomass). The inventory analysis phase includes the data collection and modeling of all of the input and outputs of material, energy, and other elementary flows that can cause potential environmental impacts. In this study, the inventory phase is supported by the collection of primary data generated through laboratory activities that allowed the collection of relevant information on the bio-based and fossil-based PU formulas. The outputs of the foam production and foam characterization, which included the quantification of mechanical and physical properties, were therefore used as input to inventory analysis and modeling and the following impact assessment phase. In this part of the study, inventory data are characterized into potential environmental impacts. Finally, the interpretation phase is applied to discuss the validity of the results concerning the goal and scope of the study and to identify the criteria (e.g., life cycle stages, mechanical and physical parameters) to be considered in the development of new biobased PU foams with improved environmental performances. Results of the interpretation stage were used as inputs to guide the development of the new formulations in the laboratory activities.

4.2. Goal and Scope Definition. The goal of the LCA study is to compare the environmental performances of the new biobased PU formulations used for building insulation, containing different quantities and types of renewable feedstocks (polyols and lignin). To guarantee a solid comparison of the performances of the new bio-based formulas with the fossil-based one, it was decided to use the product category rules (PCR) for insulation materials set by The International EPD System. These rules are a set of defined sectorial parameters and requirements, determined by the market in a participatory process, that standardizes the application of LCA of a specific product.

According to the selected PCR, the functional unit, referred also as the declared unit, is defined as the amount of foam needed to achieve 1 m²·K/W of thermal resistance $R$, defined as

$$ R = \frac{L}{\lambda} $$

where $\lambda$ is the thermal conductivity [W/(K·m)] and $L$ is the thickness of the insulation layer [m]. The considered system boundaries (Figure 3) include all of the processes that take place along the life cycle of the product under study (with the exception of the use phase), supply of raw materials (petroleum-based polyol, bio-based polyol, methylene diphenyl diisocyanate—MDI—, kraft lignin, glycerine), manufacturing (both lignin liquefaction and foam production), waste processing, and disposal. In accordance with the PCR, the use phase has not been considered, as it is not associated with energy or resource consumption nor with other flows relevant from an environmental point of view.

To guarantee the validity of comparison’s results, biobased and reference foams were produced in laboratory applying the same procedure, in such a way as to have the same level of
primary data quality. Secondary data were chosen, where possible, to be representative of the European area. The detailed list of the considered sources is present in the Section 4.3.2 “life cycle inventory and modeling”. Model has been created using SimaPro software.

To avoid the issue of environmental burden shift that emerged from literature review, it was decided to investigate a comprehensive set of impact categories (Table 5). In detail, impact categories were selected according to the PCR’s requirements, whereas the impact assessment methods were chosen following the ILCD recommendations (Table 5). In addition to the categories required from PCR, marine eutrophication was also considered because it emerged to be significant for biobased material production as shown by Tsiropoulos et al.

### 4.3. Inventory Analysis. 4.3.1. Laboratory Activities and Material Characterization.

PU foams have been obtained starting from commercial EMEROX polyols (supplied by Emery Oleochemicals, USA) as well as lignin-based polyols, synthesized in our laboratories. The last ones have been obtained by liquefaction of kraft lignin from softwood (Indulin AT, MeadWestvaco Corporation, USA) in presence of suitable solvents (glycerine and EMEROX polyols) and acid or base catalyst (sulfuric acid 98% or sodium hydroxide pellet, Sigma-Aldrich). Although the detailed development and optimization of the lignin liquefaction process will be the subject of another paper, generally speaking, the liquefaction process used in this work is very similar to what already reported in the literature.

To avoid the issue of environmental burden shift that emerged from literature review, it was decided to investigate a comprehensive set of impact categories (Table 5). In detail, impact categories were selected according to the PCR’s requirements, whereas the impact assessment methods were chosen following the ILCD recommendations (Table 5). In addition to the categories required from PCR, marine eutrophication was also considered because it emerged to be significant for biobased material production as shown by Tsiropoulos et al.

#### Table 5. List of the Considered Impact Categories and Suggested Methods

| impact category | units | suggested method |
|-----------------|-------|-----------------|
| emission of GHG (CC—climate change) | kg CO₂eq | GWP100 |
| emission of ozone-depleting gases (ODP—ozone depletion potential) | kg CFC-11eq | WMO |
| emission of acidifying gases (AE—accumulated exceedance) | mol H₂SO₄ | AE |
| evapotranspiration potential in nitrate equivalents (ME—marine eutrophication) | kg Pₘₐₓ | ReCiPe |
| ozone creating potential (POF—photochemical ozone formation) | kg NMVOCₘₐₓ (non methane volatile organic carbon) | ReCiPe |
| depletion of abiotic resources (elements) | kg Sb eq | CML2002 |
| depletion of abiotic resources (fossil) | MJ | CML2002 |

#### Table 6. Main Data about Raw Materials

| polyol | nOH [mg KOH/g] | bio-based content [%] |
|--------|----------------|----------------------|
| Isoexter 4530 | 510 | 0 |
| Isoexter 4537 | 350 | 0 |
| Isoter 842G | 160 | 0 |
| EMEROX 14511 | 110 | 78 |
| EMEROX 14535XP | 355 | 69 |
| glycerine | 1810 | 100 |
| lignin 1 | 400 | 87 |
| lignin 2 | 629 | 87 |
| lignin 3 | 856 | 87 |

*a*From technical datasheet. *b*Glycerine can be sourced from EMEROX production which starts from natural oils. *c*Based on raw materials used.

#### Table 7. Foam Nomenclature

| foam | chemical composition of polyols |
|------|--------------------------------|
| B01 | EMEROX 14511 |
| B02 | EMEROX 14511, EMEROX 14535XP |
| B03 | EMEROX 14535XP, glycerine |
| BL01 | lignin, EMEROX 14511, glycerine, acid catalyst 3% |
| BL02 | lignin, EMEROX 14511, glycerine, acid catalyst 1% |
| BL03 | lignin, EMEROX 14511, glycerine, base catalyst 1% |

TEGOSTAB B 8469; and both were kindly supplied by Evonik (Germany).

After that, isocyanate (VORANATE M600, Dow Chemicals) has been added; the mixture has been mixed for 15 s by using a high-speed mechanical mixer and then poured in an aluminium mould. The water amount has been varied in order to obtain foams with a density within 36 ± 5 kg/m³.

Foam properties have been measured according to ISO standards: apparent densities according to ISO 845; and thermal conductivity according to ISO 8301, one day after production, at a mean temperature of 10 °C and temperature difference of 20 °C.

#### 4.3.2. Life Cycle Inventory Analysis and Modeling.

As described in Section 4.1, the inventory analysis consists in the collection and modeling of all of the input and output of the processes included in the system boundaries. To populate the life cycle inventory, data from different sources were collected (Table 8).

Primary data on the different formulations, liquefaction process, and physical and mechanical characteristics of the rigid foams described in Section 4.2 were collected directly from the laboratory activities. The amount of foam needed to satisfy the functional unit that was derived by applying eq 2:

\[
M_{PU} = \rho \cdot V_{PU} = \rho \cdot A \cdot L = \rho \cdot A \cdot L \cdot R
\]

(Table 8).

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where $M_{PU}$ [kg] and $V_{PU}$ [m$^3$] are, respectively, the mass and the volume of the foam, while $\rho$ [kg/m$^3$] is the density.

The life cycle inventory of kraft lignin is taken from Culberston et al.\textsuperscript{31} while data about the composition of AA-based polyols are made available by the manufacturer. For transportation processes, having no more specific information, a reference distance of 100 km and EURO4 lorry (16–32 ton) have been arbitrarily assumed. Data on the considered end of life scenario, according to ecoinvent 3.5,\textsuperscript{32} have been modeled with the following values: 35% incineration, 32.5% disposed in inert landfill, and 32.5% disposed in sanitary landfill.

All other useful data for a complete description of the processes under study are acquired from the ecoinvent 3.5\textsuperscript{32} database. In two cases, it was necessary to modify the processes to make them more representative of the system under study: in the ecoinvent's original polyl production process, the formulation has been modified according to that provided by the manufacturer and in the foam production process, the formulations elaborated in laboratory have replaced the original one.

In order to guarantee homogeneity, the considered foam production process and end of life scenario are the same for all of the formulations and have been taken from ecoinvent 3.5\textsuperscript{32}.

To simplify the management of data coming from laboratory activities, a parametric approach has been adopted in the modeling of environmental impacts. Parametrization refers to the practice of presenting LCA information using raw data and formulas instead of computed numbers in the unit process dataset within database.\textsuperscript{33} This method also allows the creation of a data entry scheme able to support the entire design. The model process, without the need to directly modify the model of a data entry scheme, has been adopted in the development of new biobased PU formulations with improved environmental performances, a sensitivity and uncertainty analysis were performed.

### 4.4. Impact Assessment

In order to identify the main sources of impacts for each category along the entire life cycle, a contribution analysis has been performed for the formulation (materials) presented in Section 4.3.1.

The life cycle processes were classified into six stages:

1. Polyol production includes the extraction of raw material, polyol production (both fossil and bio-based), and transport to the foam production site;
2. Lignin production includes the production of the lignin and the transport to the foam production site;
3. Liquefaction describes the lignin liquefaction in the polyol–glycerine (50/50) mixture. This stage includes also the production and the transport of the glycerine, the acid catalyst (1 or 3 wt %) and the energy required for the microwave oven;
4. MDI production includes the MDI production and transportation to the foam production site;
5. Foam production, in addition to the process itself, it also includes transport to the site of use;
6. End of life includes all the processes from the deconstruction to the final disposal.

### 4.5. Interpretation

To discuss the results of the impact assessment and determine the parameters to be monitored in the development of new biobased PU formulations with improved environmental performances, a sensitivity and uncertainty analysis were performed. Sensitivity analysis was specifically used to investigate the validity of results and to identify the most relevant parameters of the model, verifying how the final results vary when the initial parameters change. An arbitrary value of variation equal at ±20% has been considered following a common practice in LCA studies.\textsuperscript{34} Sensitivity S has been calculated according to eq 3

\[
S = \frac{\Delta I_i/I_{0,i}}{\Delta p_j/p_{0,j}}
\]

(3)

where $\Delta p_j/p_{0,j}$ is the normalized variation of the parameter $j$ with respect to the starting value $p_{0,j}$ and $\Delta I_i/I_{0,i}$ is the normalized variation of the quantified impact for the impact category $i$ with respect to the starting value $I_{0,i}$. The investigated parameters were chosen in such a way as to be the representative of the physical properties and formulation (AAperc, lignin perc, reaction_PandL, reaction_MDI, PU_\text{density}, PU_\text{lambda}).

The uncertainty analysis was performed to further validate the results of the study. Where direct uncertainty measures were not available, the approach of the pedigree matrix\textsuperscript{35} was

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**Table 8. Sources of the Inventory Data Used in the Model**

| process/material | source |
|------------------|--------|
| kraft lignin     | Culberston et al.\textsuperscript{31} |
| tallow           | modified by ecoinvent 3.5\textsuperscript{32} |
| polyol production from AA | modified by ecoinvent 3.5\textsuperscript{32} |
| lignin liquefaction | primary data |
| foam formulation and properties | primary data |
| foam production | modified by ecoinvent 3.5\textsuperscript{32} |
| all of the others processes/products | ecoinvent 3.5\textsuperscript{32} |

**Table 9. List of the Independent Parameters Implemented in the Model**

| parameter       | unit       | description                                      |
|-----------------|------------|--------------------------------------------------|
| AAperc          | %          | percentage of AA in the starting polyol          |
| polyol mass     | kg         | quantity of polyol used for the liquefaction     |
| glycerine ratio |            | ratio between glycerine mass and polyolmass      |
| lignin perc     | %          | percentage of lignin respect (polyol mass + glycerine mass) |
| sulfuric mass   | kg         | quantity of sulfuric acid used for the reaction  |
| NaOH mass       | kg         | quantity of sodium hydroxide used for the reaction |
| raw material distance | km | distance considered for the transport of raw material |
| liquefaction energy | W h | quantity of energy used by microwave oven for liquefaction |
| reaction_PandL  | kg         | quantity of polyol with lignin required for foam production |
| reaction_MDI    | kg         | quantity of MDI required for foam production     |
| reaction_additives | kg    | mass of additives required for foam production   |
| PU_density      | kg/m$^3$   | density of the produced foam                     |
| PU_lambda       | W/(K m)    | conductivity of the produced foam                |
| waste_distance  | km         | distance considered for the waste transport      |

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adopted, and following a common practice in LCA, especially when secondary data are used, a Monte Carlo analysis was performed.

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