# EVALUATING THE ENVIRONMENTAL BENEFITS OF METHANOLYSIS PROCESSES FOR POLYESTER TEXTILE RECYCLING

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## Abstract

The textile industry sector is one of the largest in the world, with a current estimated global market value of USD 1 trillion. A significant issue in this sector is its detrimental environmental impact throughout its life cycle, including production, use, and disposal. Recent research indicates that the textile industry accounts for approximately 5% of the total global waste, generating between 65 and 92 million tons of textile waste annually. It also contributes to 10% of global carbon emissions and 20% of the world's wastewater, with projections suggesting a continued increase in these figures. Polyethylene terephthalate (PET) is one of the most widely used polymers in textile production. However, recovering this polymer and other components from mixed textile waste via traditional methods, such as mechanical recycling, is a challenging task. As a result, most textile waste typically ends up being incinerated, landfilled, or in the environment.

Methanolysis, a novel chemical recycling method, offers a promising alternative for valorizing mixed textile waste. This approach enables the depolymerization of certain plastics, such as PET, into its constituent monomers, which can be directly used for several applications or polymerized back into PET. Four variants of the methanolysis process have been reported: low-temperature, subcritical, supercritical, and vapor-phase methanolysis. A long residence time and unfavorable reaction conditions result in low-temperature and supercritical methanolysis being less desirable. Therefore, in this work, we present a computational framework that integrates process modeling and design with life cycle assessment to quantify the environmental benefits of depolymerizing PET mixed textile waste into dimethyl terephthalate (DMT) and ethylene glycol (EG) through the subcritical and vapor-phase variants of the methanolysis process. We compare our results with the environmental impacts of producing the EG and DMT monomers from fossil sources. Our analysis highlights critical components of the methanolysis processes with the highest environmental burdens. Our study provides valuable insights into the valorization of textile waste through methanolysis processes.

## 1. Introduction and Motivation

The textile sector is an important part of the worldwide economy. However, the production, use, and end-of-life of textiles can result in significant environmental impacts. For example, the production phase generates large amounts of wastewater and carbon emissions. Current estimates indicate that the textile industry accounts for around 10% to 20% of global carbon emissions and of the world's wastewater, respectively [1-3]. Overconsumption of clothing driven by fast fashion and lifestyle trends in the last decades has created a dramatic increase in textile demand around the world. In addition, cheaper fibers, such as PET, have enabled more consumers to participate in low-cost fast fashion trends, exacerbating this problem. This has directly shortened the lifespan of textile materials, leading to a huge amount of textile waste. These factors have led to a linear economy (make, use, and dispose) in the textile sector, which is why the sector is responsible for generating 5% of worldwide waste [4]. The U.S. Environmental Protection Agency reported that textiles accounted for 15.4 million tonnes of the total municipal solid waste in the U.S. in 2018. Alarmingly, less than 1% of this waste is used for fiber-to-fiber recycling, further underscoring the problem at hand [5].

The increasing use of synthetic materials in textile production has worsened the environmental impacts of the sector, since their production depends heavily on non-renewable resources and is associated with high energy demand and emissions. Synthetic fibers are currently the most used materials for textiles. Polyester fibers, such as polyethylene terephthalate (PET), represent more than half of the total production [6, 7]. In 2023, it was estimated that PET constituted approximately 57% of all fibers produced [8]. PET is a thermoplastic polymer derived from fossil sources.

Characteristics such as semi-crystallinity, low density, and high tensile strength make PET suitable for textile production. However, these properties are the reason for their slow degradation and non-biodegradability features. Improper waste management has made them ever-present in our environment, contributing to long-term pollution.

PET is primarily produced from the transesterification reaction of dimethyl terephthalate (DMT) and ethylene glycol (EG) or the direct esterification of terephthalic acid and EG monomers. These monomers are mainly obtained from fossil sources, and their production is not only energy demanding but also involves the use of toxic substances and the release of negative emissions into the environment (land, water, and air). Although PET production consumes only about 0.7% of total global crude oil, it is still reliant on fossil-derived monomers. Therefore, there is a need for alternative production pathways. The current approach to the production of these monomers is based mainly on conventional (brown) chemistry approaches rather than sustainable (green) chemistry principles [9]. It is crucial to move toward a sustainable textile economy to avoid the environmental harm of textile waste and reduce carbon emissions, as well as sustain the growing use of synthetic fibers in textiles.

Textile recycling can help address many of the challenges the textile sector currently faces. For example, textile recycling can help reduce the resource and environmental burden associated with virgin fiber production, as well as reduce textile waste generation and accumulation [10]. However, recycling textiles can be challenging because of their multicomponent nature, making traditional mechanical recycling methods ineffective. Even with the use of mechanical recycling for textiles, economic challenges would persist because the processes remain labor-intensive and the resulting fibers are typically lower in quality than virgin materials, reducing overall profitability. These disadvantages contribute to the low recycling rates of textile materials [11, 12], with approximately 85% of textile waste landfilled or incinerated in the U.S. [13].

Chemical depolymerization may offer a sustainable pathway to recycling the synthetic component of textile materials. This process can depolymerize target polymers, such as PET, into virgin-grade monomers that can be reformed into base polymers for fiber production or used in other applications, such as fuel production, packaging, and pharmaceuticals. About 56% of post-consumer textile waste in the U.S. may be suitable for recycling through chemical depolymerization, potentially generating about $1.5 billion in value [14]. Chemical depolymerization can be classified into glycolysis, methanolysis, ammonolysis, and hydrolysis depending on the solvent used [15]. Due to easy monomer purification, the ability to handle low-purity feedstock, and the use of less toxic solvents, methanolysis offers some competitive advantages over other depolymerization pathways [8].

In this work, we present a computational framework integrating process modeling and life cycle assessment to analyze two variants of the methanolysis process (i.e., subcritical and vapor-phase methanolysis) for producing EG and DMT monomers from PET mixed textile waste. Although two other methanolysis variants exist (i.e., low-temperature and supercritical), they are less attractive for large-scale commercialization due to unfavorable reaction conditions over the former variants [16]. The purpose of our study is to compare the environmental impact of producing these monomers using methanolysis with their production from fossil fuels (virgin EG and DMT monomer production). We offer insights into the specific process variables responsible for the highest environmental impacts, and possible pathways to maximize their environmental benefits.

## 2. Review of Related Work

Recent research has studied the methanolysis of PET derived from both textile and plastic waste, reflecting the growing interest in this depolymerization pathway. DesVeaux, J. S., et al. [17] noted in their study that the methanolysis process outperforms glycolysis and hydrolysis for virgin polyester recycling on a cost and environmental impact basis. Similar conclusions were also derived from McNeeley, A., and Y. Liu's [16]. Experimental work has also been reported for this process. For instance, Hwang, H., and M. Han [18] studied the kinetics of the polyester methanolysis reaction model. They noted that the methanolysis process comprises two reactions: a homogeneous and a heterogeneous reaction. The heterogeneous reaction occurs at the beginning, where liquid methanol diffuses into the solid PET particles. Reduction of the PET chain length through random scission to solid oligomers then occurs. Solid oligomers then dissolve in the bulk methanol solution, forming liquid oligomers. It is at this point that the shift from the heterogeneous reaction to the homogeneous reaction happens. During the homogeneous reaction stage, the liquid oligomer undergoes transesterification to produce DMT and EG monomers. More insights from their study include that the heterogeneous reaction is highly temperature-dependent, and the homogeneous reaction is an equilibrium reaction dependent on the MeOH/PET mass ratio. **Table 1** provides a high-level summary of select recent publications on the methanolysis process, detailing the type of PET material studied, the methanolysis variant considered, and the final product obtained.

Table 1. Brief overview of recent methanolysis studies.

| Type of PET Feedstock | Experimental Work or Process Modeling | Methanolysis Variant | Considers Life Cycle Assessment (LCA) | Product(s) | Impact Categories Considered | Reference |
|---|---|---|---|---|---|---|
| Plastic | Experimental work | Subcritical | No | DMT and EG | - | [18] |
| Plastic and Textile | Experimental and Process modeling | Subcritical | Yes | Recycled PET | 2 | [19] |
| Textile | Experimental work | Subcritical | Yes | DMT, EG, and Heat | 1 | [20] |
| Textile | Process modeling | Vapor-phase | Yes | Recycled PET | 2 | [16] |
| Plastic | Process modeling | Vapor-phase | Yes | Recycled PET | 6 | [21] |
| Plastic | Experimental work and Process modeling | Subcritical | Yes | Recycled PET | 14 | [22] |
| Plastic and Textile | Process modeling | Not stated | Yes | Recycled PET | 3 | [23] |
| Plastic and Textile | Experimental work | Low temperature | No | DMT and EG | - | [24] |
| Plastic | Process modeling | Subcritical | Yes | Recycled PET | 11 | [17] |
| Textile | Experimental work | Low temperature | No | DMT and ethylene carbonate | - | [25] |
| Plastics | Experimental work | Subcritical | No | DMT and EG | - | [26] |
| Textile | Experimental and Process modeling | Low temperature | Yes | DMT and EG | 2 | [27] |

The depolymerization of waste PET plastic and textile materials via methanolysis using a green, efficient, and sustainable catalyst was done in Cao, J., et al. [19]. They synthesized and characterized an oxygen vacancy-rich Fe/ZnO nanosheet catalyst for polyester depolymerization. Using this catalyst, the depolymerization of various PET plastic and textile waste combinations was studied, with the catalyst showing good tolerance and high catalytic activity. An LCA comparing PET depolymerization and repolymerization with PET production from fossil sources was then analyzed. Their analysis shows a 56% reduction in energy consumption and a 44.5% reduction in greenhouse gas emissions. This result highlights the potential of this technique for lowering carbon footprints and enhancing energy efficiency in PET production.

Muangmeesri, S., et al. [20] studied the depolymerization of PET fibers via methanolysis using an organo-catalyst (trimethylamine). To determine the optimal ratio of PET/solvent/catalyst that is most appropriate to obtain a high DMT yield in the process, an optimization study was conducted. This research demonstrated that using toluene as a cosolvent at a 1:1:0.13 ratio with PET/solvent/ catalyst resulted in the highest DMT yield. Using this information, they conducted an LCA to estimate the global warming potential of depolymerizing PET fibers via this method to obtain DMT, EG, and heat (through waste incineration), and compared this with the production of these products from conventional sources. They concluded that 1.88 kg of $CO_2$ equivalent can be avoided when utilizing the depolymerization technique for the production of these materials.

As shown in **Table 1**, there have been numerous approaches to evaluate the efficacy and effectiveness of methanolysis for PET depolymerization. However, a systematic investigation of the environmental impact of DMT and EG monomer production through this depolymerization route is still lacking. Additionally, the process analysis of each stage of the different variants of methanolysis processes has not been well studied. We contribute to addressing these gaps by developing a framework that integrates process design and LCA. A proper understanding of the environmental impacts of monomer production through methanolysis can help reduce the carbon footprint of PET fiber manufacturing. Moreover, this knowledge can facilitate both open- and closed-loop recycling of waste PET fibers, as DMT and EG monomers are used in other products such as dyes, pigments, insect repellents, and pharmaceuticals [14, 28].

# 3. Technology Approach

## 3.1. Process design

The methanolysis processes aim to recover DMT and EG monomers from waste PET fiber materials. We modeled both processes, considering the depolymerization of 200,000 tonnes of PET fiber per year. Process simulations were conducted in UniSim [29] using the extended NRTL thermodynamic model. We selected this thermodynamic package based on its use in similar research [17, 19, 30]. Since PET is not a component in the available database in UniSim, its physical and thermodynamic data were estimated from the literature [12, 31]. Zinc acetate was used as the catalyst in our study based on its use in other similar studies [19, 21]. It is important to note that we analyzed the variants according to the different methanolysis variant descriptions in the work of McNeeley, A., and Y. Liu [12].

The subcritical methanolysis variant was modeled using the process described in Cao, J., et al. [19]. Waste PET fiber, zinc acetate catalyst, and methanol were mixed and heated prior to their introduction into the reactor. In the reactor, the depolymerization reaction occurred at 160 ºC and 17 bar. Following the reaction, the undissolved solid catalyst was filtered out using a solid-liquid separator. Due to the exothermic nature of the reaction, the liquid mixture's temperature is greater than the boiling point of methanol, and as such, some portion of methanol is removed from the process liquid in the flash drum. Upon cooling of the liquid mixture, DMT starts to crystallize and is subsequently filtered out in the separator. Water is added to the liquid mixture to help crystallize the remaining DMT in the liquid mix, and this is also filtered out. Both DMT streams are then combined and collected as a pure DMT-rich stream with > 99% purity. The liquid mixture containing water/EG/methanol was separated using a series of distillation columns. Following distillation, high-purity EG and methanol were obtained for the process. We considered a scenario where we recycled 95% of methanol (based on results from simulation) and 98% of the solid catalyst (based on recycling rate in the literature [16]) back into the process loop.

The vapor-phase methanolysis variant was modeled using the process described in McNeeley, A., and Y. Liu [16] and Uekert, T., et al. [21]. Both works discussed the Eastman methanolysis patent [32] but interpreted it slightly differently. The process starts with solubilizing the waste PET material in a melt mixture made up of bis-2-hydroethyl terephthalate (BHET), DMT, and EG in the dissolver. This mixture is then fed into a reactor, where it comes in with superheated vapor methanol and zinc acetate catalyst. The depolymerization reaction occurs at around 300 ºC and 3 bar. Due to the operating conditions of the reactor, most of the product formed immediately gets flashed and exits at the top of the reactor, leaving unreacted PET and catalyst to exit at the bottom. The top outlet of the reactor is then cooled before being sent to a series of distillation columns for component separation.

The first distillation column is operated with a partial reflux condenser. A rich methanol stream exits at the top of the condenser, and a mixture of DMT and EG exits at the bottom. The melt mixture, comprising mostly BHET and some EG and DMT components, leaves the bottom of the column. The rich methanol stream is then connected to a second distillation column, where the remaining EG and DMT are removed, yielding a pure methanol stream exiting the top of the second column. The bottom stream exiting this column (containing only EG and DMT) is then recombined with the stream exiting the bottom of the condenser in the first column in a crystallization unit. In the crystallization unit, methanol was used as the crystallization agent to crystallize the DMT component in the mixture. After the crystallization process, a separator was used to filter out solid crystallized DMT. A third distillation column was used to help separate uncrystallized DMT from the EG and methanol streams. The stream exiting at the bottom of the third column is subsequently combined with the solid DMT stream to obtain a pure DMT product stream. Finally, EG is separated from methanol in the fourth distillation column, following which a pure methanol stream exits at the top of the column, and a pure EG product stream is collected at the bottom of the column. Here, we considered a scenario where we recycled 95% of methanol (based on the simulation results) and 98% of the solid catalyst and melt stream (based on recycling rates reported in the literature [16]) back into the process loop.

## 3.2. Life cycle assessment (LCA)

The goal of this LCA study is to compare the environmental impacts of the production of DMT and EG monomers from PET depolymerization via the subcritical and vapor phase methanolysis processes against fossil-derived DMT and EG monomers. The system boundary for the LCA, as shown in **Figure 1**, represents a cradle-to-gate system with a functional unit of 1 kg DMT and 1 kg EG production. For this study, the conventional "cut-off" approach is used to incorporate the impact of waste PET fiber. This implies that the waste PET fiber imposes no environmental burden from its previous life/lives and thus has no impact within the studied recycling system. The factors to estimate the environmental impacts of the processes are obtained from the Environmental Footprint database [33]. The annual utility and solvent usage of the methanolysis processes used for the LCA are summarized in Table 2. These inputs were obtained from the earlier-described process simulation work. Since DMT and EG were co-products from both methanolysis processes, a mass-based allocation was done to estimate the individual contribution of each monomer produced using the methanolysis processes based on the method described in Matthews et al. (2014) [34].

Table 2. Annual utility and solvent usage of the methanolysis processes.

| Utility/Solvent | Unit | Subcritical Methanolysis | Vapor-phase Methanolysis |
|---|---|---|---|
| Electricity | MJ | 4,991,184 | 215,136 |
| Steam | MJ | 9,700,960,000 | 3,700,336,000 |
| Cooling water | m3 | 382,400,611 | 23,010,020 |
| Methanol | tonne | 60,000 | 67,200 |
| Water | tonne | 405,600 | - |
| Catalyst | tonne | 31 | 31 |
| Melt | tonne | - | 924 |

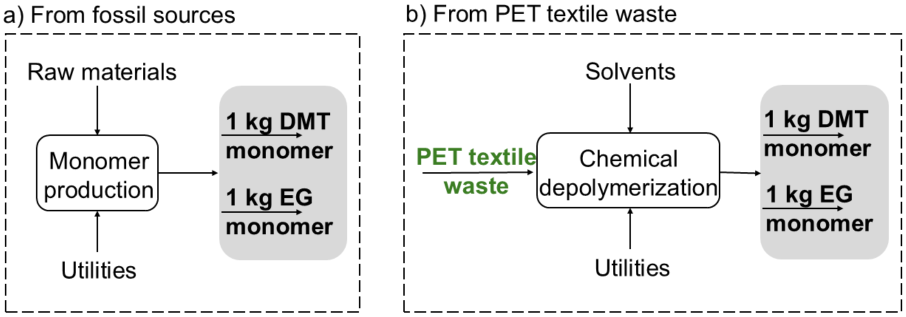

*Figure 1. System boundaries for the LCA study. (a) EG and DMT monomer production from fossil sources. (b) EG and DMT monomer production via the methanolysis processes.*

# 4. Discussion

Both processes (subcritical and vapor-phase methanolysis) were modelled, with a waste PET feed stream of 25,000 kg/hr as input. The yields of the DMT and EG monomers increased by 0.16% and 0.73%, respectively, for the vapor-phase methanolysis process. This slight difference in yield is likely due to the PET solubilization step performed during the modeling of this variant. For both of the processes considered, the purity of the monomers was over 99%, highlighting their efficiency. The subcritical methanolysis process used nearly three times more steam compared to vapor phase methanolysis, with the distillation column separating the EG/water mixture, accounting for about 55% of the total steam used in the process. As a result, the subcritical process required more electricity and cooling water for operation. Based on our process modeling results, the subcritical methanolysis variation is more energy- and resource-intensive than the vapor-phase configuration.

**Figure 2** presents a comparison of the cradle-to-gate climate change impact of DMT and EG monomer production via subcritical and vapor-phase methanolysis variants with those from fossil sources. The impact per monomer produced through the methanolysis processes was allocated based on mass-based allocation calculations to quantify the environmental impact attributed to their respective productions. The impacts were expressed per functional unit: 1 kg of DMT and 1 kg of EG monomer production. For the methanolysis processes, the impacts related to solvent (melt, water, methanol), catalyst, and process utilities (steam, cooling water, and electricity) are also shown in **Figure 2**.

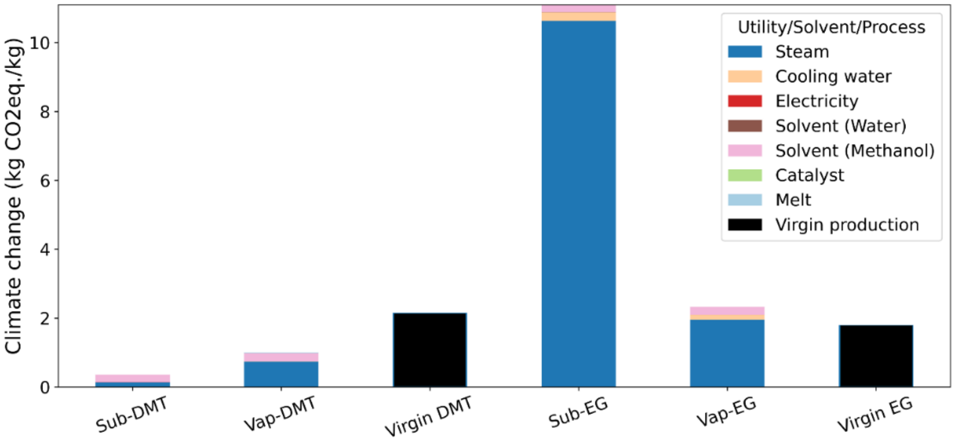

*Figure 2. Climate change impact of the production of EG and DMT monomers through the subcritical (Sub-DMT/EG) and vapor-phase (Vap-DMT/EG) methanolysis processes, and through the production from fossil sources (Virgin DMT/EG).*

For DMT production, we observe that both subcritical (Sub-DMT) and vapor-phase (Vap-DMT) methanolysis processes generate lower $CO_2$ emissions per kg of monomer produced than virgin DMT monomer production. The subcritical and vapor-phase methanolysis processes generate 83% and 54% fewer emissions, respectively. This can be attributed to fewer unit separations used for DMT monomer extraction and to the ease of DMT purification from the reaction mixture. For the Vap-DMT process, the most contributory process unit and solvent to the overall climate change impact were the column used for DMT/EG/methanol separation and the methanol solvent used as the DMT crystallizing agent, which together accounted for 83% of the total emissions from the process. Overall, the avoided impacts for DMT production from these methanolysis processes highlight their feasibility as an alternative source for producing this monomer.

Regarding the climate change impact of producing the EG monomer, we highlight that the virgin production of this monomer has lower environmental impacts than its production via the subcritical (Sub-EG) and vapor-phase (Vap-EG) methanolysis processes. We note that steam plays a significant role in these processes. About 96% and 84% of the total climate change impact from Sub-EG and Vap-EG processes, respectively, is generated by steam consumption. We also note that the climate change impact for the Sub-EG process is significantly higher than that of the Vap-EG process (4.8 times) and the virgin process (6 times). To better understand the behavior of these processes, we analyze the contribution of each process unit, solvent, and utilities used for both processes. We present these results in **Figures 3** and **4**. These results provide insights into the process tasks that generate the highest climate change impact and identify potential areas for improvement.

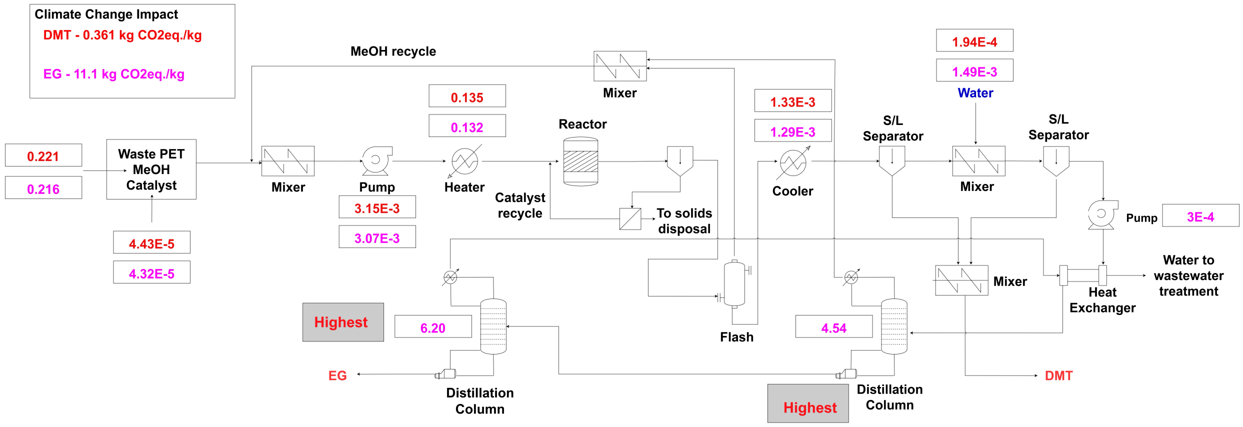

*Figure 3. Climate change impact per process unit, utilities, and solvent for the subcritical methanolysis process*

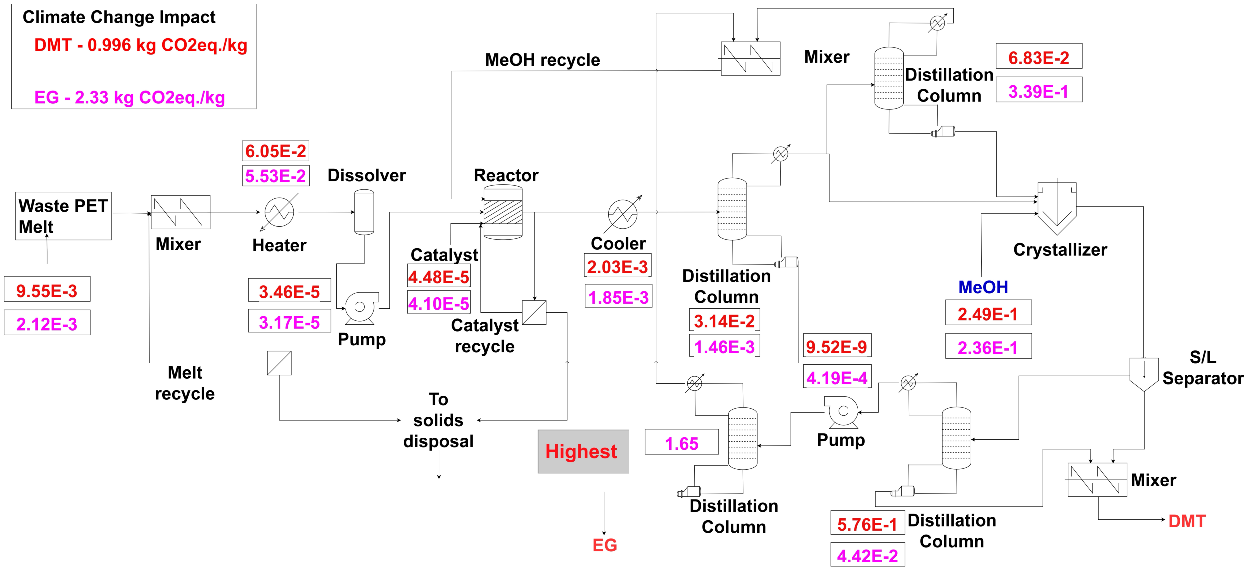

*Figure 4. Climate change impact per process unit, utilities, and solvent for the vapor-phase methanolysis process*

For EG monomer production via the subcritical methanolysis process presented in **Figure 3**, we see that the distillation columns separating the water/EG/methanol mixture are responsible for the highest impacts. These columns combined contribute to about 97% of the total emissions from the process. The high utility demand of these distillation columns makes the subcritical methanolysis process generate a significant amount of greenhouse gas emissions as compared to the other processes. This demand is related to the EG/ water separation, as it is an energy-intensive process. This result is in agreement with recent research [35], which conducted an LCA study of different microwave-assisted glycolysis pathways for PET depolymerization. In this regard, improving the energy demand of the distillation columns could be achieved by using a different DMT crystallization agent, rather than water, which is currently being used. Similarly, the EG monomer production via the vapor-phase methanolysis process is presented in **Figure 4**. In this process, the column separating methanol/EG generates the highest environmental impact. This column contributes to about 71% of the total climate change impact of the process. As stated earlier, this variant is more energy efficient than the subcritical process, which contributes to its lower climate change impact. We note that this variant still generates a higher impact for EG production than the virgin production process.

## 5. Conclusions & Recommendations

We developed a computational framework integrating process modeling and LCA to provide insights into two variants (subcritical and vapor-phase) of the methanolysis process for depolymerizing PET waste fiber. The proposed framework helps compare these variants to identify process operating conditions with the lowest environmental impact for DMT and EG monomer production. We compare the results of our process simulation and analysis with the virgin production of these monomers from fossil sources. From these results, we conclude that across both of the methanolysis processes, steam plays a significant role in their environmental performance. The subcritical and vapor-phase processes for DMT production had 83% and 54% fewer emissions, respectively, than the virgin production process. As for the EG monomer production, no climate change impact benefits were obtained when compared to the virgin production process. Our framework may serve as a guide to the design of textile recycling using methanolysis processes, considering its environmental impacts at each process stage. Future work will include analyzing various steam sources (e.g., renewables) to determine the extent to which steam affects the methanolysis process's environmental impact. Lastly, we intend to extend the impact categories currently considered beyond climate change. These additions would make our analysis more robust and help us avoid issues that can arise from problem-shifting, which is often attributed to studies with single impact categories.

## 6. Acknowledgements

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