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Prospective life cycle assessment of bio-based adipic acid production from forest residues

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ABSTRACT

Environmental concerns related to the production of bulk chemicals are growing. Researchers and technology developers are currently looking into alternative production pathways for such chemicals by utilizing renewable resources, such as lignocellulosic feedstocks. Adipic acid is an example of such a bulk chemical, and its conventional fossil-based production emits significant amounts of N_2O, a major greenhouse gas. In this study, a prospective life cycle assessment (LCA) of bio-based adipic acid production from forest residues at an early development stage, situated in Sweden, was conducted. Acid-catalyzed (using SO_2) and alkaline (using $NaBH_4$) pretreatment were employed and scenarios and sensitivity analyses were conducted. The potential environmental impacts of this technology under development were compared to those of conventional adipic acid production. The results show that bio-based adipic acid production has a lower impact on global warming, eutrophication and photochemical ozone creation compared to fossil-based production. In contrast, it has a higher impact on acidification. An increased efficiency of mitigating N_2O emissions from the fossil-based production may alter this comparison. Producing bio-based adipic acid using the alkaline pretreatment has a higher environmental impact than producing it using acid-catalyzed pretreatment. Furthermore, if biomass is used to fulfil process energy demands, instead of fossil fuel, the environmental impact of the bio-based production decreases. It is therefore important to reduce the amount of $NaBH_4$ used in the alkaline pretreatment or to lower the environmental impact of its production.

1. Introduction

The increase of population, the growing concerns about climate change and issues related to non-renewable resource depletion encourage society to find alternative ways to divest from the fossil-based economy. The current fossil-based economy includes a wide range of chemical products, from large-scale bulk to high-value added fine chemicals. Environmental concerns related to the production of these chemicals are growing and potential solutions such as by increasing selectivity, reducing by-product formation, and developing multi-functional catalysts in order to minimize the number of reaction steps, are explored (Hoelderich, 2000). Another solution may be to substitute fossil-based, non-renewable resources and products with renewable ones, which leads to completely different production processes (Sandén and Pettersson, 2014).

Adipic acid ($(CH_2)_4(COOH)_2$) is considered as an important bulk chemical and is a pre-cursor for the production of nylon-6,6. About 80% of the world-wide adipic acid production is used for the production of this polymer (Shimizu et al., 2000), and its production increased almost 9-fold from 2000 to 2010 (Li et al., 2014). There are several industrial-scale production routes to produce adipic acid, and all of them are fossil-based. The most common route is via nitric acid oxidation of cyclohexanol and cyclohexanone, so-called ketone-alcohol (KA) oil (Shimizu et al., 2000). This production route also leads to the formation of nitrous oxide (N_2O), one of the major greenhouse gases (GHG). Its climate impact is 298 times higher than that of carbon dioxide (CO_2) (Myhre et al., 2013). N_2O emissions account for about 9% of global annual greenhouse gas (GHG) emissions, and currently, adipic acid production itself contributes almost 80% of total industrial N_2O emissions (Li et al., 2014).

Despite the availability and implementation of N_2O emission abatement technologies, N_2O emissions remain as a major concern in climate change mitigation. The N_2O emissions due to adipic acid

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production have been a motivation for finding an alternative production process in order to avoid N_2O emissions. For instance, Wang et al. (2013) used a direct synthesis of adipic acid from cyclohexene using H_2O_2 as an oxidant (instead of nitric acid) in a continuous process, and conducted a life cycle assessment (LCA) to determine its environmental impact. Even though the results indicate lower impact on the climate, the authors emphasized that this route has disadvantages in several other life cycle impact categories, which indicate a potential environmental burden shifting. In a more recent study, Han (2016) performed experimental studies on adipic acid production through catalytic conversion of corn stover whose purpose it also was to lower N_2O emissions. However, the study did not quantify the environmental benefits of this production route.

Another way to produce adipic acid is through biological conversion (Draths and Frost, 1994; van Duuren et al., 2011). Draths and Frost (1994) developed the microbial conversion of D-glucose into cis,cis-muconic acid followed by the reduction to adipic acid. van Duuren et al. (2011) performed process simulation and limited LCA studies of adipic acid production via the biological conversion of aromatic feedstocks (benzoic acid, impure aromatics, toluene, and phenol) from lignin to cis,cis-muconic acid. Both studies were conducted at a very early stage in the development of the process. Producing adipic acid via bioprocess pathways looks promising because, not only does it not emit N_2O, it also increases the use of renewable resources (biomass), instead of fossil-based raw materials (KA-oil). Forest residues, which mainly consist of branches and tops from commercial thinning, can be considered as such a promising renewable resource. In Sweden, approx. 8 TWh of bioenergy is already generated from forest residues (Ortiz et al., 2014). Nevertheless, 80% of these residues are still left on the forest floor after harvesting, which suggests their availability as a feedstock (Ortiz et al., 2014). A previous study mentioned that using forest residues as a feedstock for adipic acid production can be considered as a promising alternative pathway (Svensson et al., 2015).

Considering that adipic acid bio-conversion technology is still at an early development stage, its environmental performance needs to be evaluated with assessment tools such as LCA, using a prospective perspective, in order to guide its further development. To the best of the authors' knowledge, there is only one study that conducted a limited LCA of bio-adipic acid production (van Duuren et al., 2011), which implies that a more comprehensive prospective LCA study is needed. Therefore, the purpose of the LCA study reported in this paper is to assess the potential environmental impacts and to identify the environmental hotspots of bio-based adipic acid production from forest residues in Sweden at an early development stage, and to compare it with the existing fossil-based production process. The LCA results are intended to guide the decision makers for further development towards a more environmentally benign adipic acid production technology and process.

2. Materials and methods

2.1. System description

The technical system under study is depicted in Fig. 1.

2.1.1. Forestry activities

In the upstream activities, firstly the forest residues from Norway Spruce harvesting are collected by a forwarder at the harvesting site and brought to the roadside. The forestry activities were assumed to take place approx. 50 km from the adipic acid plant, situated in Örnsköldsvik, Sweden. It was assumed that the timber extraction produced two different products: roundwood, which is the main part of the trees and the forest residues. Economic allocation is used to attribute environmental impacts from forest residue extraction.

2.1.2. Pretreatment

The pretreatment fractionates the forest residues into cellulose, hemicellulose, free sugars, lignin, and other materials. There are two types of pretreatment used in this study: the acid-catalyzed and alkaline pretreatments. Sulfur dioxide (SO_2) is used for the acid-catalyzed pretreatment process. The pretreatment was conducted by adding steam at 212 °C and at 20 bar. The steam is generated by burning lignin and methane produced as a byproduct from the fermentation and anaerobic digestion (Fig. 1), respectively.

The alkaline pretreatment is performed by using a 7% (m/m) solution of sodium borohydride ($NaBH_4$) based on the mass of the forest residues. The process is then followed by mild steam explosion (STEX) at 110 °C. Next, the alkaline pretreatment by using sodium hydroxide (NaOH) is performed.

2.1.3. Neutralization and upstream separation process

For both acid and alkaline pretreatment processes, the neutralization step is needed in order to adjust the pH to approx. 4.8, which is optimal for the hydrolysis and fermentation process. In case of acid-catalyzed pretreatment, NaOH was added, while hydrochloric acid (HCl) was used to neutralize the pretreated material after the alkaline pretreatment. All materials then go into the upstream separation process (see Fig. 1) where the water with free sugars is discharged into the anaerobic digestion process which produces the biogas to fuel the steam generation. The rest of the materials are sent into the fermentation reactor.

2.1.4. Hydrolysis and fermentation

The flow into the hydrolysis and fermentation process is the pretreated forest residues which contains cellulose, hemicellulose, lignin, and other components (bark, ash, free sugars, etc.). The hydrolysis process produces fermentable sugars, undigested holocellulose, and unfermentable free sugars. The other components, including lignin, are only slightly affected by the hydrolysis process.

In the case of acid-catalyzed pretreatment, the Cellic Ctec2 enzyme product is used to hydrolyze cellulose and hemicellulose. In the case of alkaline pretreatment, there is no enzyme used (Jedvert et al., 2012; Jedvert, 2014). The fermentation consists of the conversion of C_6 sugars to adipic acid via lysine (Burgard et al., 2013), and it was assumed that this two-step process can take place in one fermentation reactor to produce adipic acid. The required nutrient for the fermentation process was assumed to be ammonium sulfate ($(NH_4)_2SO_4$).

2.1.5. Downstream process

The downstream process unit operations and their energy demands were based on a simulation study performed by Schweigler (2016). The evaporation of water and the concentration of adipic acid are the most energy intensive parts of the downstream process. Lignin is produced by the downstream process as by-product. The filtration unit produces relatively pure adipic acid while the other flows (water with dissolved chemicals and materials) are either recycled to increase the recovery of adipic acid, or are flowing to the anaerobic digestion (see Fig. 1). The steam needed in the downstream process is produced by burning fossil fuel and lignin. Meanwhile, the undigested part of hollocellulose and process water are sent to a wastewater treatment plant, which is considered outside of the system boundary.