Superstructure-based process synthesis and optimization of hydrogen production via biomass gasification

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Achieving carbon neutrality by 2050 is the ambitious goal of the Green New Deal, the current EU policy in response to the global climate crisis (Hafner and Raimondi 2020). Decarbonization of mobility is considered particularly challenging since this sector is nowadays heavily relying on fossil energy sources accounting for around 20% of the global anthropogenic CO_2 emissions (Lepage *et al*, 2021). Hydrogen is foreseen to have a pivotal role in the decarbonization of mobility. Indeed, it is a clean, efficient, versatile fuel and a raw material to other endpoints such as ammonia and methanol. Market forecasts show that hydrogen production will have in the next years a growth of 5-10% per year in concomitance with the creation and retrofitting of supporting infrastructure at the production, distribution, and consumption stages (Lepage *et al*, 2021; MISE, 2020). In order to cope with sustainability requirements, the growing hydrogen economy should rely on technologies with low life cycle Greenhouse Gas (GHG) emissions such as water electrolysis and biomass gasification (Lepage *et al*, 2021; Olateju and Kumar, 2015; Cao *et al*, 2020). Water electrolysis has advantageous techno-economical properties for large-scale applications and offers the possibility to convert surplus electrical energy from intermittent sources like wind and solar farms (Olateju and Kumar, 2015). On the other hand, biomass gasification has the advantage of relying on non-intermittent input materials and being applicable on a wide scale-range offering the opportunity of an hydrogen production diffused in the territory (Olateju and Kumar, 2015; Parkinson et al., 2019). However, field-scale applicability of biomass gasification for hydrogen production is not yet achieved. One of the reasons is a lack of knowledge regarding the relationships between several operating parameters and techno-economic-environmental performance of biomass gasification systems (Cao *et al*, 2020).

Within this context, useful hints can be gained through conceptual process design. Indeed, the present contribution lays in this domain and focuses on hydrogen production via biomass gasification. Our aim is to design different process configurations and classify them according to techno-economic-environmental performance at optimal operating conditions. This should provide a valuable support for early-stage decision-making in planning and retrofitting plants. We use a superstructure-based approach for process synthesis and optimization, whereas with superstructure is meant a set of all unit operations (e.g., gasification reactor, adsorber, filtering unit, reformer etc.) which can be employed in the process. Our work is inspired by analogous studies on superstructure-based synthesis and optimization of biorefineries: Ma and You (2019) designed and economically optimized a process for poultry litter valorization to char, gasoline, heat, and power including gasification units in the superstructure; Wang *et al* (2013) identified the best conceptual processing route for gasification-based production of diesel and gasoline from lignocellulosic biomass according to both economical and environmental aspects; Zhang *et al* (2014) performed a similar study including hydrogen production from biomass gasification. They found that this processing route is not favorable in terms of both economical and environmental aspects compared to natural gas reforming and pyrolysis-based routes due to the high utility consumption. However, they included in the superstructure only high and low temperature steam gasification units. In our work we include in the superstructure a broader range of units namely steam, air, oxygen, steam/air, and steam/oxygen mixture gasification operated with different catalysts.

According to the above cited studies, Mencarelli *et al* (2020) and Quaglia *et al*, (2015), performing superstructure-based process synthesis and optimization generally involves the following methodological steps:

1. Superstructure definition:
   In the framework of our work, we define a superstructure including unit operations needed to treat lignocellulosic biomass to yield pure hydrogen via gasification plus natural gas reforming units to provide a benchmarking process configuration. The superstructure is made up of subsets corresponding to processing steps (e.g., gasification, syngas cleaning, water-gas-shift reaction, syngas purification, combined heat, and power production) which include unit operations. The identification of appropriate unit operations related to each processing step is performed through systematic review of technical reports and scientific articles.
2. Formulation of a Mixed Integer Non-Linear Programming (MINLP) problem:
   This step is performed analogously to Wang *et al* (2013) and Zhang *et al* (2014). The optimization objectives functions are the Net Present Value (NPV) and the Global Warming Potential (GWP). The former is maximized, while the latter is minimized. All unit operations are modeled with linear equations

to facilitate the problem solvability. Each unit operation of the superstructure is associated with a binary integer variable, whose value indicates selection or rejection. Other process variables (e.g., flow rates, capacities) are continuous. Constraints are formulated as both equalities and inequalities. The whole MINLP problem is formulated in GAMS. The necessary process parameters and specifications are collected from databases, technical reports, and scientific articles following the hierarchical procedure proposed by Quaglia *et al* (2015).

3. Solution of the MINLP problem:
   In the framework of our work, we use the global optimizer BARON which implements solution algorithms of the branch-and-bound type (GAMS, 2021). The two problem's objectives, NPV and GWP, are reformulated to a single objective through the ε-constraint method as proposed by Wang *et al* (2013) and Zhang *et al* (2014).

Solving the optimization problem yields different process configurations at optimal processing conditions. Thereby, it is possible to compare process configurations according to the economic and environmental performance of hydrogen production. Future work should extend the superstructure to further processing steps such as hydrothermal carbonization, and further input raw materials such as municipal organic waste.

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