Deep Decarbonization in the Pulp Industry - Carbon Dioxide Recycling and Utilization in a Lime Kiln via Methanation

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Abstract

In most pulp mills lime kilns are the only source of fossil carbon dioxide emissions. Natural gas is a widely used fossil fuel combusted in lime kilns. For deep decarbonization of the pulp mill, the fossil fuels have to be substituted by alternatives to cut CO₂ emissions from combustion process. Besides biomass-based approaches, the utilisation of hydrogen can be a possible fuel. A complete replacement of the thermal power requirement by hydrogen is not possible due to technical issues in the lime kiln. The utilization of hydrogen is limited to 50 % of the thermal power required in the lime kiln. In order to avoid the CO₂ emissions from the other half of the required fuel, the work proposes a recycling of emitted carbon dioxide from the process for the conversion to synthetic natural gas (SNG).

The process concept is based on the carbon capture unit and the methanation process. Carbon dioxide is captured from the flue gas of the lime kiln via an amine wash. Since the CO₂ concentration is higher than in power plant flue gases, the capture process is expected to be more efficient. The carbon dioxide is reacted with hydrogen to methane (SNG) and water in a series of adiabatic reactors. In a last step, the water is separated from the product methane. Since the reaction of calcium carbonate to calcium oxide in the lime kiln also frees carbon dioxide, the amount of carbon dioxide is enough to satisfy the methane demand of the lime kiln. The production of SNG for the utilization in the lime kiln required only parts of the CO₂ to be separated from the flue gas. Additionally, a simplified lime kiln model (45 MWth) is implemented to calculate the composition of the flue gas for the investigated cases depicted below. As a base case the thermal power is solely supplied by natural gas to the lime kiln. The following cases with the indicated thermal power supply are evaluated:

1) Utilization of 50 % hydrogen and 50 % natural gas (no carbon capture)
2) Utilization of 50 % hydrogen and 50 % SNG (carbon capture rate to supply the amount required for the methanation process)
3) Utilization of 100 % SNG (carbon capture rate to supply the amount required for the methanation process)
4) Utilization of 50 % hydrogen and 50 % SNG (full carbon capture, surplus SNG is fed to the gas grid)

The cases were simulated in a chemical engineering software to calculate mass and energy balances. Based on the simulation the cases 1 – 4 and the base case were evaluated and compared to each other. Besides the comparison of yearly CO₂ emissions, CO₂ abatement cost are calculated for the scenarios. The utility requirements like heating and cooling are compared in term of integrability into the pulp mill. High temperature steam from the SNG process can for example be used in the steam turbine of the pulp mill for electricity generation. The energy balances of the processes are presented and evaluate in terms of efficiency. The carbon capture unit is investigated in detail since the flue gas composition affects the process efficiency significantly.

The investigated concept can also be transferred to the cement industry since processes are very similar. The scale of the process has to be increased for the application in the cement industry.

Keywords: Hydrogen; Carbon Dioxide Capture; Lime Kiln; Methanation; Process Simulation; Deep Decarbonization

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