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Techno-Economic Analysis of Methane Production from Pulp and Paper Sludge

Erfan Hosseini[a], Selen Cremaschi[a*], and Zhihua Jiang[ab]

[a] Department of Chemical Engineering, Auburn University, Auburn, AL, US
[b] Alabama Center for Paper and Bioresource Engineering, Auburn University, Auburn, AL, US
* Corresponding Author: szc0113@auburn.edu.

ABSTRACT

This study investigates the feasibility of valorizing pulp and pulp sludge (PPS) into methane through anaerobic digestion (AD) with a focus on techno-economic analysis (TEA). Three scenarios are evaluated: (A) the base case, (B) sludge AD with alkaline pretreatment using green liquor dregs (GLD), and (C) co-digestion with nitrogen-rich feedstocks. The evaluation is applied to a common PPS, consisting of 70% primary sludge (PS) from the primary clarifier and 30% secondary sludge (SS) from biological treatments from a kraft mill. Theoretical methane potential (TMP) is determined using the Buswell equation. The study highlights the significance of co-digestion with nitrogen-rich feedstocks in enhancing the economic viability of the AD process for PPS, providing valuable insights for sustainable waste management and resource recovery in the pulp and paper industries.

Keywords: Pulp and paper sludge, biomethane, anaerobic digestion, techno-economic analysis, valorization.

INTRODUCTION

The pulp and paper industry (PPI) generates a significant volume of wastewater that undergoes physical and biological treatment [1,2], resulting in a considerable quantity of pulp and paper sludge (PPS) [3], which is one of the major waste streams of PPI [4]. As global energy demand escalates and concerns over energy security and climate change intensify, anaerobic digestion (AD), which transforms organic materials into methane and carbon dioxide in the absence of oxygen [5], has emerged as a versatile technology for renewable energy production. This microbial-mediated process presents a promising solution for managing PPS, offering an alternative energy source to supplement industrial fossil fuels. PPS contains 45-55% organic matter and various nutrients, including nitrogen and phosphorus [6], making it a potential resource for microorganisms in the production of bio-products and biofuels. The conversion of pulp and paper mill by-products, such as sludge, into value-added goods and bioenergy can significantly contribute to the commercial growth of biorefineries. However, for effective utilization, these by-products must meet specific minimum standards. To enhance efficiency and profitability, there is a growing interest in evaluating opportunities to generate new revenues from innovative, value-added products within the existing infrastructure of pulp and paper mills. Consequently, this study aims to identify a promising process pathway for a biorefinery integrated with an existing pulp and paper mill, considering various scenarios in pursuit of improved efficiency and profitability.

METHODOLOGY OVERVIEW

Process synthesis and design

The techno-economic analysis (TEA) of AD for PPS considers three distinct scenarios (Fig. 1), each representing a unique process aimed at evaluating the economic and technical feasibility of methane production. The sludge mixture constituted 70% PS from the primary clarifier and 30% SS obtained from biological treatments. The sludge generation varies widely among mills. In this context, the input mass flow for the sludge is assumed to be 500 tons per day with a dry matter content of 12%. The dry matter content and mass flowrate specified conform to data obtained from a United States Environmental Protection Agency (EPA) investigation encompassing

104 bleached Kraft mills and are consistent with established industry norms [7,8].

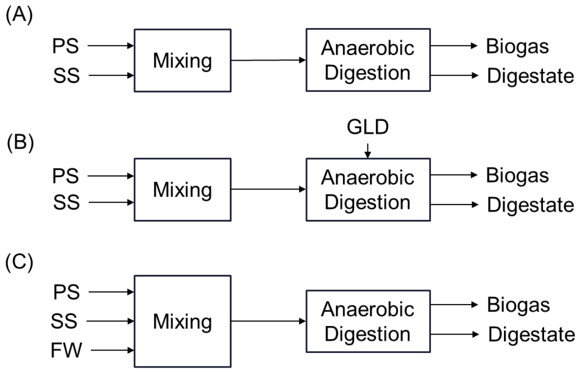

Figure 1. Three TEA scenarios for AD of mixed kraft mill primary and secondary sludge.

Scenario A: Base case

The base case scenario mirrors the conventional operational conditions of AD. As a baseline condition, scenario A serves as a reference point for assessing the shortcomings and limitations of the existing process in terms of economic viability and methane production efficiency.

Scenario B: Sludge AD with alkaline pretreatment

Among the various pretreatment methods, alkaline pretreatment is a promising approach that can successfully improve the enzymatic hydrolysis for many lignocellulosic materials [9,10].

As reported in [9], alkaline pretreatment effectively disrupted the floc structure of pulp and paper sludge, leading to a reduction in fiber size. According to reference [9], effective biodegradation in bioreactors was evidence in terms of soluble chemical oxygen demand (SCOD) removal efficiency, which attained the range of 83–93% in bioreactors with pretreated PPS compared with 70% removal for untreated PPS, indicating a significant improvement of 18.6-32.8% in organic removal. These findings show the efficacy of alkali pretreatment as a promising method for enhancing methane yield in the context of PPS.

An alkaline pretreatment typically involves the use of hydroxides like NaOH or KOH [11]. However, it has been suggested that the GLD, a by-product of the kraft pulping process, could be used as a substitute for powerful and expensive alkaline agents like NaOH [12].

Scenario C: Co-digestion with nitrogen-rich feedstocks

In this scenario, food waste (FW) is specified as a promising nitrogen-rich feedstock for co-digestion with PPS, supported by existing studies on the co-digestion of FW with PPS [13]. Previous work has mentioned nitrogen deficiency as a major challenge in AD of PPS [14,15]. The ideal carbon-to-nitrogen ratio for AD feedstock typically falls within the 20-30% range [16]. A high C:N ratio, as observed in PPS, accelerates nitrogen consumption by methanogens, adversely affecting microbial population growth and prolonging the carbon digestion process. Conversely, a low C:N ratio leads to elevated ammonia release in the digester and inhibits the AD process.

Theoretical methane potential

Theoretical methane potential was assessed using the Buswell equation (Eq. (1)), which relies on the stoichiometric balance between biodegradable organic matter and the resulting gaseous products from anaerobic biodegradation [17]. Volatile solids (VS) represent the organic matter content of the sludge that is readily converted to gas during the anaerobic digestion process, and total solids (TS) refer to the total mass of both organic and inorganic matter present in the sludge. These key parameters, VS and TS, play a significant role in determining the efficiency of anaerobic digestion and the potential methane yield. In the specific case of PPS, the Buswell equation can be applied, assuming a 30% volatile solids (VS) removal based on experimental findings [14]. In this context, biodegradation efficiency signifies the proportion of VS degraded during the process.

$$C_aH_bO_cN_dS_s + \left(a - \frac{b}{4} - \frac{c}{2} + \frac{3b}{4}\right) H_2O \rightarrow \left(\frac{a}{2} + \frac{b}{8} - \frac{c}{4} - \frac{3d}{8}\right) CH_4 + \left(\frac{a}{2} + \frac{b}{8} + \frac{c}{4} + \frac{3d}{8}\right) CO_2 + dNH_3 + sH_2S \tag{1}$$

We can deduce the maximum theoretical methane potential (TMP) using Eq. (2):

$$TMP\left(\frac{m^3}{kgVS}\right) = \left(\frac{\left(\frac{4a+b-2c-3d}{8}\right)\times 22.415}{12a+b+16c+14d}\right) \tag{2}$$

Economic analysis

Gross economic potential (GEP)

The gross economic potential is used to assess the economic benefits and potential value generated by the process. The GEP is defined in Eq. (3).

$$\text{GEP} = \text{VP} + \text{STF} - \text{VF} \tag{3}$$

where VP is the value of products, STF is savings on tipping fee, and VF is the value of feeds. The value of products refers to the economic value generated from the products resulting from the AD process. In the case of converting sludge to methane, VP would include the revenue or value obtained from selling the methane produced, as well as any other by-products that can be monetized, such as organic fertilizers or other valuable substances extracted from the process. Tipping fees are charges imposed for disposing of waste in landfills. By diverting sludge from landfills and converting it into biogas through AD, the process effectively reduces or eliminates the need to dispose of the waste in landfills. This saving is a significant factor in the economic assessment, especially given the specific cost of the tipping fee ($58.47/ton [18]). Value of feeds represents the value of any feeds or materials used in the process. In the context of PPS converted to biogas, the VF is considered zero because PPS is a by-product with no value as a feed or input material.

Manufacturing cost

The sizing and cost estimation of the pretreatment reactor followed the methodology proposed by Ulrich et al. [19]. However, in the case of the digester, its large size exceeds the range covered by the graphs outlined. Therefore, for the digester, we referred to a similar farm digester documented in the literature as a case study to estimate costs [20]. Capital cost of this case study is scaled to the capacity of AD systems assumed in our scenarios based on the usual 0.6-power rule (Eq. (4)):

$$CC_A = CC_B \times \left(\frac{Cap_A}{Cap_B}\right)^{0.6} \quad (4)$$

In Equation 4, CC_A and CC_B represent the capital costs of equipment A and B, respectively, while Cap_A and Cap_B denote the capacities of equipment A and B. All cost calculations are based on the chemical engineering plant cost index (CEPCI) of 816 (for 2022). Finally, assuming 18% of the bare module costs for contingency costs and fees based on reference [21], the total module cost (C_{TM}) is calculated as Eq. (5) where n represents the total number of pieces of equipment and $C_{BM,i}$ is the bare module cost for each piece of equipment i:

$$C_{TM} = 1.18 \times \sum_{i=1}^{n} C_{BM,i} \quad (5)$$

In this work, the CAPCOST method was applied to estimate the manufacturing costs [21]. Following the methodology proposed by Ulrich [19], the typical labor requirement for a continuous reactor is estimated at 0.3 workers per unit per shift. The operating labor cost (C_{OL}) estimation assumptions considered are:

- On average, a worker at this plant operates five shifts per week for 52 weeks annually.
- The plant operates 365 days a year with three shifts per day.
- The 2022 Mean Annual Wage for Chemical plant and system operators is reported as $79,290 per year, according to the U.S. Bureau of Labor Statistics [22].

The equation used to evaluate the cost of manufacture (COM) is:

$$COM = \mathit{Direct\ Manufacturing\ Costs} + \mathit{Fixed\ Manufact\ Costs} + \mathit{General\ Expenses} \quad (6)$$

Each individual cost item can be estimated considering the costs of utilities (C_{UT}), waste treatment (C_{WT}), raw materials (C_{RM}) and fixed capital investment (FCI). Turton et al. provide typical ranges for constants (multiplication factors) (Table 8.2. of [21]) to estimate these individual cost items. Since no other information is accessible regarding these costs in our study, we utilize the midpoint value within each range. Depreciation allowance is added separately to compute the cost of manufacturing (COM) using Eq. (7).

$$COM = 0.28 \times FCI + 2.76 \times C_{OL} + 1.23 \times (C_{UT} + C_{WT} + C_{RM}) \quad (7)$$

In Eq. (7), the FCI cost equals the total module cost, given that we are making alterations to an existing facility. In the specific context of our preliminary feasibility study or conceptual design, we adopt a simplified model or scenario wherein the costs of utilities (C_{UT}), waste treatment (C_{WT}), and raw materials (C_{RM}) are ignored. However, it is essential to acknowledge that in real-world scenarios, these costs would typically be significant factors contributing to the overall manufacturing expenses.

RESULTS AND DISCUSSION

Buswell method

Based on the results of the elemental composition analysis for sludge derived from Lopes et al. [23] (refer to table 1), we can calculate the theoretical volume of methane using Eq. (2).

Table 1. Elemental compositions of the primary sludge (PS) and secondary sludge (SS) and mixed sludge (7:3) [23]

Parameters	PS	SS	Mix
VS/TS (g/g)	0.99	0.85	0.97
C (% TS)	44.10	45.20	44.41
H (% TS)	6.04	5.83	5.98
O (% TS)	48.80	29.80	34.86
N (% TS)	0.06	4.85	1.43
S (% TS)	0.40	1.82	0.81
Ash(%TS)	0.60	12.50	12.51
Total	100	100	100

VS: Volatile Solids; TS: Total Solids

According to the data presented in Table 2 calculated using Eqs. (1) and (2), the TMP for the mixed primary and secondary sludge, with a ratio of 7:3, was approximately 130.95 mlCH4/gVS$_{\text{fed}}$, assuming a 30% removal of volatile solids (VS). This aligns with experimental findings documented in the literature [14,24]. This theoretical value provides a preliminary insight into biogas and methane production.

Table 2. Biogas product yield and composition from the Buswell equation

Biogas	Yield (ml/gVS$_{fed}$)	Composition (%)
CH_4	130.95	49.12
CO_2	126.41	47.42
NH_3	7.43	2.79
H_2S	1.79	0.67
$Biogas$	265.63	100

Techno-economic analysis

GEP of the different scenarios

For the base case scenario, the methane yield amounts to 130.95 mlCH$_4$/gVS$_{fed}$, equivalent to 269,141.78 ft^{3}CH$_4$/day and 17.46 tons PPS conversion per day. In a simplified context, assuming full upgrading and considering the lower heating value of methane at 910 Btu/ft^3 [25], the resulting energy production would be approximately 244.92 million Btu/day. Considering a natural gas price of $6.45 per million Btu [26] and the average cost of landfilling municipal solid waste in the US in 2022 at $58.47 per ton [18], the GEP of the base case is 0.95 million dollars per year.

In the second scenario, given the absence of specific Volatile Solid Removal (VSR) data in the literature, we approximated the improvement in VSR by referencing the enhancement in Soluble Chemical Oxygen Demand (SCOD) removal. Both SCOD and VSR serve as crucial indicators of organic material removal efficiency, and according to the literature [27], they tend to exhibit similar trends. According to [9] the highest organic conversion rate corresponds to a 33% increase in SCOD removal efficiency. This translates to approximately a 40% conversion of volatile solids for the pretreated PPS in our study, leading us to anticipate a methane yield of 174.60 mlCH4/gVS$_{fed}$, equivalent to 358855.71 ft^{3}CH$_4$/day and 23.28 tons PPS conversion per day. Taking these factors into account, the GEP for the second scenario is estimated to be $1.26 million per year.

According to the literature, AD of food waste gave a specific methane yield of 470 mlCH$_4$/gVS$_{fed}$, which is equal to approximately 70% of the theoretical value (660 mlCH$_4$/gVS$_{fed}$ with a biogas methane content of 58%) based on the Buswell equation. Also, the food waste exhibited a TS content of 23.9% and a VS content of 21.6% [28]. Assuming the absence of any synergistic effects, the biodegradation efficiency, calculated by averaging the efficiencies of AD for both FW and PPS, stands at 50%. Using the Buswell equation and the elemental composition for the mixture of PPS and FW at a TS ratio of 1:1 (refer to Table 3), the cumulative methane yield reaches 291.76 mlCH$_4$/gVS$_{fed}$, aligning with the findings reported in the literature [29]. With a fixed ratio of 0.94 gVS/gTS [23,28] and a total solids content of 12%, the calculated methane yield amounts to 581,107.51 ft^3/day, resulting in a converted PPS of 28.2 tons/day. Consequently, for the third scenario, the GEP is $1.85 million per year.

Table 3: Elemental compositions of the food waste and mixed Pulp and paper sludge

Parameters	FW [28]	FW:Mixed PPS (1:1)
VS/TS (g/g)	0.90	0.94
C (% TS)	51.10	47.75
H (% TS)	6.41	6.19
O (% TS)	32.50	33.68
N (% TS)	3.10	2.26
S (% TS)	0.00	0.40
Ash (% TS)	6.89	9.72
Total	100	100

Sizing and Capital Cost

Digester

Considering 12% TS and an organic loading rate (OLR) of 5 kgVS/m^{3}day, the required digester volume is 11,220 m^3 or approximately 3 million gallons (MG) [30].

The hydraulic residence time (HRT) was calculated as 22.44 days for a 3 MG digester processing 500 tons of feed per day, aligning with established industry standards. The cost estimation is derived from the Synergy Biogas, LLC Case Study [20]. The purchased cost from the case study has been adjusted to suit the assumed capacity of the AD systems in this study. The capital cost for a digester vessel with a volume of 2.2 million gallons, as indicated in the report from the year 2011, is specified at $1.25 million. The cost was adjusted to fit the 3 MG capacity using Eq. (4) yielding a value of $1.50 million. Subsequently, converting this cost estimation to the 2022 price, the adjusted cost would be approximately $2.01 million.

Pretreatment reactor

The sizing of the pretreatment reactor, determined in accordance with established norms, for a given input flow rate (q) and hydraulic retention time (HRT) of 1 hour is computed using Eq. (8), resulting in a volumetric capacity of 20.83 m^3 (Equivalent to a process vessel with a height of 5 m and an inside diameter of 2.3 m).

$$Volume(m^3) = HRT \times q \tag{8}$$

Based on Ulrich's graphs (see Fig 5.44 of reference [19]), the estimated purchase cost for a carbon steel vessel of this size under atmospheric pressure is approximately $150,000.

For Scenario B, the total bare module cost for this initial TEA is assessed by combining the purchased costs of digester and pretreatment reactor, totaling $2.16

Table 4: Comparative economic analysis of a PPS-based biogas plant in different scenarios.

Scenarios	C_{TM} ($MM)	Labor cost ($MM)	COM ($MM)	GEP ($MM)			Payback period (years)	CFRR (%)	NPV ($MM)
				VP	STF	Total			
A	2.37	0.12	0.99	0.58	0.37	0.95	Undefined	NA	-2.04
B	2.55	0.20	1.26	0.77	0.49	1.26	Undefined	NA	-1.98
C	2.37	0.12	0.99	1.25	0.60	1.85	4.3	22.55	2.50

NA: Not Applicable; C_{TM}: Total Module Cost; COM: Cost of Manufacturing; GEP: Gross Economic Potential; CFRR: Cash Flow Rate of Return; NPV: Net Present Value

million. Utilizing Eq. (5), the total module costs for scenarios A, B, and C amount to $2.37 million, $2.55 million, and $2.37 million, respectively.

Operating Cost

Labor cost

Based on the specified assumptions, the base case scenario requires 1.5 full-time operators, whereas the scenario involving alkaline pretreatment necessitates 2.5 full-time operators due to an additional reactor.

According to the 2022 yearly mean wage estimates from the US bureau of labor statistics, the reported annual mean wage for chemical plant and system operators is $79,290 [22]. Using this information, the estimated annual labor cost is $118,935 for the base case and scenario C and $198,225 for the scenario incorporating alkaline pretreatment.

Fixed capital investment

The fixed capital investment (FCI) for the process is equal to the total module cost for each scenario. Finally, using Eq. (7), the total manufacturing cost is estimated to be $0.99, 1.26, and $0.99 million for scenarios A, B, and C respectively.

Cash Flow Analysis

The feasibility of different scenarios is analyzed based on key parameters – i.e., net present value (NPV), cash flow rate of return (CFRR), and payback period. Table 4 summarizes revenue generation through each scenario, and other critical parameters for a 20-year project life (years after Startup). The assumed values, parameters, and equations for the analysis are summarized in Table 5.

Scenario C stands out as the most promising scenario as it was the only one to yield a positive NPV. This favorable outcome is principally due to its high conversion rates and methane production yield, resulting in increased revenue. Although Scenario B has higher conversion rates than the base case scenario, its overall performance is still overshadowed by higher total module cost and comparatively higher COM due to higher labor costs associated with the additional reactor.

Further research is needed to explore the synergistic effects of co-digesting FW and PPS, ensuring a more comprehensive understanding of their potential economic benefits in AD processes. In addition, to enhance the robustness of our findings and pave the way for future advancements, several additional aspects demand further attention. Considering the potential benefits of co-digesting FW and PPS, along with alkaline pretreatment, presents an opportunity to reveal novel insights for optimizing AD processes. For more accurate cost estimation, it's crucial to integrate biogas upgrading, utility and chemical expenses (covering items such as inoculum for bacterial cultures, buffering agents, antifoaming agents, and, where applicable, enzymes) into the analysis. These efforts will collectively contribute to advancing the economic and environmental sustainability of AD processes in the PPI.

Table 5: Parameters and equations employed for the cash flow analysis

Parameter	Unit	Value
Project life	Years	20
Construction period	Years	3
Taxation rate	%	21
Annual interest rate	%	10
Depreciation method	-	MACRS (5-year)
Annual interest rate	%	10
Salvage value	$	0.1*FCI [21]
Working capital	$	0.1*(C_{RM} + FCI + C_{OL}) [21]

CONCLUSION

This study investigates the potential of AD to valorize PPS through TEA. Three distinct scenarios are compared: (A) the base case, (B) sludge AD with an alkaline pretreatment, and (C) co-digestion with nitrogen-rich feedstocks.

The study highlighted the enhanced economic viability of PPS digestion through the integration of food waste (Scenario C). Alkaline pretreatment (Scenario B) also showed potential with a 33% increase in volatile solids conversion.

Further research is needed to investigate the synergistic effects of co-digesting food waste and PPS in AD and integrate biogas upgrading, utility, and chemical expenses in the TEA. Conducting experiments to optimize process parameters such as temperature, pH, TS content, OLR, and retention time could enhance methane production efficiency in the pulp and paper industry's AD processes. In addition, exploring potential applications for the AD digestate could lead to additional revenue streams or beneficial reuse options.

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