Syngas Production via Supercritical Gasification of Leachate: A Review

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Abstract. Leachate is characterized as heterogeneous, recalcitrant wastewater, and potentially harmful pollutant to the environment and human health. It is encountered in landfills municipal disposal, which contains high concentrations of many organic and inorganic compounds such as Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Total Organic Carbon (TOC), and Total Nitrogen (TN). This report provides a comprehensive research review of syngas production characteristics via supercritical gasification (SCG) technology as a treatment before leachate is released to final waste disposal. This technology is worked by gasifying the leachate under supercritical water condition (500°C, 23 MPa) followed by syngas production such as H₂, CO, CO₂, CH₄. First, we compare the essential characteristic of leachate by addressing the value of COD, TOC, pH, TN to understand effect contaminants that will be harmful. Second, the effect of gasification techniques are evaluated to syngas production and carbon gasification ratio. Finally, the landfill leachate treatment using supercritical gasification indicates that this method plays a vital role in treating leachate with syngas production that hydrogen is produced as abundant gas compares to the other. The result also summarizes future development that supercritical gasification technology is feasible to reduce leachate pollutants; therefore, it has the possibility to release into surface water.

1. Introduction

Landfills have been the most common methods to organize waste disposal and often encountered in each province of Indonesia. In fact, 95% of the total municipal solid wastes collected worldwide are disposed of in landfills [1]. Landfills are also contained hazardous residues that are spread around the world [2]. There are three categories of landfills, such as open dumps, controlled dumps, and sanitary landfills. During landfill site operation, rainwater and liquid water percolate through the landfill; then, it dissolves the landfill component as hardened decaying metals, microorganisms, and organic and inorganic compounds [3]. The water which passed through the waste from precipitation and generated from the waste resulting in a liquid phase carrying suspended solids, soluble components of the waste, and products from the degradation of the waste by various micro-organisms is called landfill leachate.

Leachate is a complex wastewater that contained a high concentration of pollutants, which possibly to affect the environment. Hitherto, leachate can percolate from pipe to groundwaters or mix with surface waters, contributes to the pollution of soil, groundwater, and surface water that contribute to human and industrial water usage [4]. Landfill leachate is also consisting of; dissolved organic material, volatile fatty acids and humic-like material, inorganic macro components, xenobiotic organic
compounds which are compounds not degraded by organisms in the environment, and include aromatic hydrocarbons, pesticides, plasticizers, chlorinated aliphatic compounds, etc. [5].

Chian and Dewalle [6] that are described leachate composition for nine sanitary landfills; stated that was leachates prominent characteristics of high concentrations of organic species and heavy metals. They found that the ratio of Chemical Oxygen Demand (COD)/Total Organic Carbon (TOC), which effected to determine the age of landfills, varied from 3.30 (for young landfills) to 1.16 (for older landfills). Several leachate treatment methods can be explained, such as sequencing batch reactor (SBR) and its modification [7], membrane bioreactor (MBR) [8], adsorption [9], and membrane separation [10]. As refined steps, physicochemical techniques usually used combining with biological treatments, and the operation cost was high clearly compared with other treatments (biological process).

On the other hand, leachate also contains high organic matter content that feasible to convert into energy such as syngas via supercritical gasification. Supercritical gasification is a promising technique to convert organic matter to become energy (syngas) [11]. In the process of supercritical condition, water increased simultaneously into a very high temperature and pressure; it shows an excellent promising solvation media for rapid gasification and destruction of aqueous-organic wastes because of the increasing ability of diffusion. Water gain into a critical point ($\geq 374.2°C$ and $22.1$ MPa), Therefore, becomes a supercritical phase that can be is placed into quickly dissolved and gasified organic compounds without drying process [12]. Besides, SCG is also able to remove pollutant, which is contained by leachates such as COD, TOC, and TN parameter [13].

This review aims to: 1) analyze leachate composition and characteristic; 2) discuss the factors which are influenced to gain syngas products; 3) discuss factor of carbon gasification and removal pollutant efficiency during SCG; 4) summarize current challenges and treatment technology facing production landfill leachate.

2. Leachate

Commonly, the leachate comes through natural chemical changes over time that reduces its toxicity [14]. Some of the remaining molecules naturally adsorbed to the clay liner particles and contaminant surface water [15]. The generation of leachate from a sanitary landfill is a complex combination of physical, chemical, and biological processes whereby waste age influences the performance of landfills to generate leachate [16]. Most landfill areas occur at least four phases of decompositions such as 1) an initial aerobic phase, 2) an anaerobic acid phase, 3) an initial methanogenic phase, 4) a stable methanogenic phase [17].

2.1. Leachate Characteristics and Composition

Several components will be specified characteristics of leachate. Some gases are found in the landfill area, such as methane, carbon dioxide, nitrogen, and ammonia [18]. Methane is often flammable and sometimes even more explosives, but carbon dioxide and nitrogen are nonharmful substances to the environment. The presence of ammonia in air is an irritant and causes burning of the eyes, nose, throats, and lungs.

| Table 1. Gases produced by a municipal solid waste landfill [18]. |
|---------------------------------------------------------------|
| Component            | Percent (dry volume basis) |
|----------------------|----------------------------|
| Methane              | 40-60%                     |
| Carbon Dioxide       | 40-60%                     |
| Nitrogen             | 2-5%                       |
| Ammonia              | 0.1-1.0%                   |

There are three specific periods according to the BOD/COD ratio, such as the acid phase, transient phase, and methanogenic phase showed in table 2. BOD is a biochemical parameter that generally
indicates the biodegradability of organic matter in leachate composition. BOD to COD ratio is an indicator of the proportion of biochemically degradable organic matter to total organic matter. It describes the organic composition in the leachate, and it appears to be a representation of waste stabilization from the early acid phase to the methanogenic phase [19]. If the BOD/COD ratio for untreated wastewater is 0.5 or higher, the waste is considered to be easily treatable by the biological process. If the rate is below about 0.3, either the wastewater may have toxic components or microorganisms may be required in its stabilization.

| Leachate Phase         | Criteria        |
|------------------------|-----------------|
| Acid Phase             | BOD/COD ≥0.4    |
| Transient Phase        | 0.4 > BOD/COD > 0.2 |
| Methanogenic phase     | BOD/COD ≤0.2    |

Period of wastewater placement in landfills shows the extent of microbial activities that affect the quality of leachate. Some parameters, such as COD, BOD, and pH parameter, influence the quality of leachate explained in table 3 [20]. Any wastewater must control pH, CD, and BOD if it is released to the environment. It needs biological, physical treatment to reduce pollutant that contained in wastewater.

| Table 3. The leachate characteristics depend on the age |
|--------------------------------------------------------|
| Young        | Intermediate | Old             |
| Age (year)   | <5           | 5-10            | >10            |
| pH           | <6.5         | 6.5-7.5         | >7.5           |
| COD (mg/l)   | >10,000      | 4,000-10,000   | <4,000         |
| BOD/COD      | >0.3         | 0.1-0.3        | <0.1           |
| TOC/COD      | <0.3         | 0.3-0.5        | >0.5           |
| NH₃-N (mg/l) | <400         | 400            | >400           |
| Heavy metals (mg/l) | Low-medium | Low            | Low            |
| Organic Compound | 80% VFA  | 5-30%          | Humic Acid + Fulvic Acid |
| Bidegradability | Important | Medium        | Low            |

Higher content of BOD, COD, and ammonia will decrease the quality of water and inhibit microorganisms. Therefore, aerobics treatment cannot occur in this condition. Figure 1 describes the main components of leachate.
Table 4 summarizes the composition of leachate in terms of pH, COD, BOD, TOC, Ammonia, TN, TDS, and TC [21-25]. Paula et al. [21] have reported leachate with the old period has explained COD >10,000 mg/l. Other studies conducted by Weijin et al. [22], Gong et al. [23], and Xuejun et al. [24]. It is explained young, intermediate, intermediate, and period characteristics of leachate, respectively. The pH of those studies mentioned above was ranging from 5 to 8. The young period of leachate indicates biodegradable easily and also contains 80% volatile fumic acid. Ammonia, which contained in leachate, is harmful to humans if released directly into the environment because the safe limit of ammonia, which can be inhaled by humans, is two ppm. Leachate has an average value of NH$_3$ about 400, which has a very strong scent. Therefore, leachate must be treated before it released to the environment. Research from Molino et al. did not conduct a review of the content of the contaminants such as COD and BOD but other indicators, total solid content and acetic acid that showed in table 5. The content affects the result of syngas production from the supercritical gasification process.

Table 4. Comparison of Leachate Composition

| Content   | Paula et al.[21] | Weijin et al[22] | Gong et al [23] | Xuejun et al [24] |
|-----------|------------------|------------------|-----------------|-------------------|
| pH        | 7.7              | 5-7              | 8.04 ± 0.02     | 7.5               |
| COD (mg/l)| 2265             | 42536-44073      | 6633 ± 330      | 10000             |
| BOD (mg/l)| 443              | -                | -               | 2000              |
| TOC (mg/l)| 646              | 14860-16120      | 4079.0 ± 376.0  | -                 |
| NH$_3$(mg/l)| 410             | 2008-2275        | 393 ± 54        | 500               |
| TN (mg/l) | -                | 1703-2279        | 3800 ± 82       | 700               |
| TDS (mg/l)| 5410             | -                | -               | -                 |
| TC (mg/l) | 5568             | -                | 9650.3 ± 467.2  | -                 |
According to Tables 4 and 5, leachate with the high value of COD, BOD, and TOC (Total Organic Carbon) content is considered to convert into energy, especially in the gas phase (syngas).

### 2.2. Leachate Treatments

There are certain treatments to process leachate are discussed. Biological treatment is the most cost-effective to destruct the organic leachate constituents. Aerobic treatment is easier to control and has a longer industrial waste treatment history. Anaerobic processes are good to consider because of low energy consumption and produce minimal sludge. Nowadays, microbial interaction is important to develop and control mixed microbial cultures. The role of genetically engineered organisms is uncertain, given their instability and the complexity and variability of leachate composition.

The coagulation technique has been demonstrated for removing both heavy metals, colloidal material, and entrained oil. Lime is the most commonly used coagulant. Carbon adsorption and membrane processes have the capability of removing a variety of organic and inorganic leachate constituents. However, coagulation, adsorption, and membrane processes do not destruct the partitioned leachate constituents so that further processing or disposal is needed necessarily [26].

Alternative chemical or thermal leachate oxidation treatment may be suitable for certain types of leachates but operate at more extreme conditions and thus require more stringent control and safety precautions. Thermal treatment, which is proposed, is supercritical gasification. Table 6 explains the comparison of performance removal contaminants of leachate in certain treatment techniques. SCG shows result better than other techniques. Supercritical gasification not only reduces pollutant TOC, COD, and BOD contained on leachate but also produced clean energy (H2).

| Treatments (COD:>1500mg/l) | Technology | Performance removal |
|---------------------------|------------|---------------------|
|                           |            | COD(%)  | TOC(%)  | TN(%)  |
| Biological Treatment      |            |         |         |        |
| Aerobic Treatment         | Active Sludge Reactor | 75-97   | -       | -      |
| Anaerobic Treatment       | Digester   | 56-70   | -       | -      |
| Physical/Chemical Treatment |          |         |         |        |
| Floatation                | Coagulant FeCl₃/Al₂(SO₄)₃ | 55      | -       | -      |
| Coagulation               |            | -       | -       |        |
| Absorption                | Adsorbent CaCO₃ (2-4 mm) | 90      | -       | -      |
| Membran Filtration        | Nano Filtration | 66      | -       | -      |
| Thermal Gasification      | Supercritical (Batch reactor) | 80-91   | 85-90   | 10     |

**Table 5. Leachate Composition**

| Content                   | Molino et al. [25] |
|---------------------------|--------------------|
| Water (%w/w)              | 86                 |
| Total Solid Content (%w/w)| 9.5                |
| Lactic Acid (%w/w)        | 8.8                |
| Acetic Acid (%w/w)        | 0.9                |
| Ethanol (%w/w)            | 0.2                |

**Table 6. Comparison of leachate treatment technology**
3. Supercritical Gasification System

Supercritical gasification (SCG) is a novel technology that can convert organic matter into syngas. The gas-phase consists of methane, hydrogen, carbon monoxide, carbon dioxide. SCG produces clean gas because there is no production of NOx. Pradecta et al. [27] investigated the phenomenon of biomass processing supercritical gasification that has obviously result in hydrogen production and less contain CO₂ with catalyst addition [27]. This research emphasizes to utilize a huge amount of biomass waste in the form of empty fruit bunches (EFB) since the moisture content is around 65%. It needs necessary to be pre-processing before EFB considered as a fuel. This Result elucidates that SCG is a suitable technology to utilize wet biomass waste to become clean energy that contained high H₂ and less CO₂ content. Figure 2 explains the experimental apparatus using in the previous experiment [27].

Steam is a promising gasification agent that produces a high content of H₂ and a high caloric value of syngas compared with air and oxygen. Although air is readily available, nitrogen dilutes the caloric value of the produced gas and leads to difficulty in gas separation. Oxygen is an expensive alternative that produces a medium heating value gas; however, it also leads to difficulty in gas separation. The biomass chemical composition, reactor configuration, gasification temperature, pressure, and addition of catalyst all work in contribution to selectively isolate H₂, deconstruct recalcitrant compounds, and produce a high energy and gasification efficiency value [28].

Figure 2. Supercritical experiment apparatus

3.1. SWOT Analysis

The process of supercritical gasification has been studied in numerous laboratory-scale experiments. The comparison to conventional treatment of leachate such as biological, physical and chemical techniques shows some advantages for the supercritical gasification, some weakness of this technology also summarized in Table 7 as follows:

Table 7. SWOT Analysis of Leachate SCG

| Strength | Weakness | Opportunity | Threat |
|----------|----------|-------------|--------|
| • Complete solution with the realization full conversion | • It has a higher energy requirement. Require higher energy compare to | • Production of gas with high energy content and amount | • Competition with other treatment methods |
| other leachate treatment | (biological, chemical) |
|-------------------------|-----------------------|
| • The resultant liquid product contains less than total solid matter | • The cost of gas generated \((\text{H}_2, \text{CH}_4)\) relatively is higher than the cost of other treatment |
| • It takes a short time to implement (seconds and minutes) and generates more energy | • Increased independence in the production of \(\text{H}_2\) and energy |
| • It has a lower reactor volume than other methods | • The people’s tendency to adopt new technology hardly |
| • The product is a clean fuel with lower emissions of nitrogen oxides | • The technology should be operated by taking high safety precautions |
| • Produce syngas production with Increased \(\text{H}_2\). It is renewable energy. | • Can eliminate pathogenic microorganisms and harmful pollutant the presence of many landfills |
| • Pathogenic microorganisms and toxic compound can be eliminated | • The increased of nitrate \((\text{NH}_3)\) concentration in some condition |
| • It reduces the needs of landfill areas and the use of fossil fuels | |

3.2. Current Leachate Treatment by using SCG

Figure 3 explains the study mapping of current technology treatment, categorized into feedstock, parameter control, and type of reactor. Most experimental works on gasification mostly use biomass and waste product as raw materials. In major, there are many variations in control parameters, but many studies often used temperature, pressure, catalyst addition as control on the SCG process. Besides, several types of reactors implemented in the experiment condition are batch reactor and a continuous tubular flow reactor. Temperature and pressure can affect the more reactive and rapidly chemical process. While the addition of catalyst conduces of increasing syngas production (maximize hydrogen and minimize \(\text{CO}_2\) production in several studies). Red box symbols show several current reviews in the previous experiment in the diagram.
Figure 3. Study mapping of supercritical gasification

The current experiment model in supercritical gasification has used several important parameters. Weijin et al. [22] show their experiment with adjusting residence time, the addition of catalyst that impact on resulting high H₂ production in SCG of leachate. H₂ concentration increase to 79.4% while the addition of NaOH catalyst. The experiment was suggested 15 min of retention time that increases hydrogen production and carbon gasification ratio. Varying flow rate and addition of catalyst that was conducted by Molino et al. [25] manifest production of syngas with Higher Heating Value (HHV) about 15-17 MJ/kg. The addition of the Nickel-based catalyst can be increasing CH₄ syngas fraction up to 50%. There are several methods that were adjustment of water oxidant coefficient into the gasification process by Xuejun et al. [24], Paula et al. [21], Gong et al. [23]. Their experiments that use a combination of water oxidation have better results of removal pollutant efficiency than conventional supercritical gasification. So, high syngas production and removal pollutant efficiency can be achieved by controlling several parameters, which are very influential such as catalyst, coefficient of oxidation, retention time, and feedstock concentration. Table 8 shows the differentiation of the experimental apparatus, which was used by researchers.

**Table 8. Experimental Facilities**

| Feedstock            | Reactor type          | Reactor Volume (mL) | Temperature (°C) | Pressure (MPa) | Substance Addition | Heating rate | Flow rate | Reference          |
|----------------------|-----------------------|---------------------|------------------|---------------|-------------------|--------------|-----------|--------------------|
| Leachate TOC: 14860-16120 mg/l | Batch Reactor with stainless steel HC276. | 600 mL             | 500              | 29            | Catalyst          | 3°C/min      | -         | Weijin et al. [22] |
4. Production of Syngas via SCG Carbon

This review summarizes several kinds of research on syngas production from the previous scholar. Figures four to seven show \( H_2 \), \( CH_4 \), \( CO_2 \), and CO production [22] [23] [25]. The increase in temperature could affect to generate more \( H_2 \) and \( CO_2 \). This is due to supercritical gasification that reacted to a temperature between 600-800\(^\circ\)C stimulate to increase the production of \( H_2 \) and \( CO_2 \) [29]. Otherwise, the concentration of CO and \( CH_4 \) slightly decreased as the increasing temperature of the reactor.

There are three phases of reaction that occur in the supercritical gasification, such as hydrolysis, pyrolysis (solid-phase reaction), and gas-phase reaction. A hydrolysis reaction transpires a chain-breaking reaction of organic matter that occurs between 270-570\(^\circ\)C [30]. Leachate contains high organic matter that indicates by BOD and TOC parameter. Next, the solid-phase reaction occurs decomposition process that several reactions such as boudard, carbon water, and hydrogenation reaction. Water-gas shift and methanation is prominent reaction to produce \( H_2 \), \( CO_2 \), and \( CH_4 \) that have a possibility to promote high energy density and high heating value of syngas.

![Figure 4. \( H_2 \) production via SCG](image1)

![Figure 5. \( CH_4 \) production via SCG](image2)
The addition of catalyst can improve reaction and push to produce more H$_2$; CH$_4$ also reduce CO and CO$_2$ content. Weijin et al. [22] utilize NaOH, KOH, K$_2$CO$_3$, Na$_2$CO$_3$, and also Molino et al. [25] utilize Ni-Al$_2$O$_3$ as a catalyst for SCG process. The catalytic reaction shows better results than the non-catalytic reaction that proven by high H$_2$, CH$_4$, and low CO, CO$_2$ content. H$_2$ production reaches 78% of syngas volume, which uses the KOH catalyst more while the highest content of CH$_4$ is 50% of syngas volume that uses Ni-Al$_2$O$_3$ catalyst. According to the hydrogen production, the effect of a catalyst on landfill leachate supercritical gasification is in the following order: NaOH > KOH > Na$_2$CO$_3$ > K$_2$CO$_3$ > Ni-Al$_2$O$_3$. The Catalytic reaction on SCG of leachate has a great effect on the high production of syngas. Therefore pressure has a little effect on syngas production because of pressure push to only kinetic reaction [31].

5. Carbon Gasification and Removal Pollutant Efficiency

5.1. Carbon Gasification Efficiency

Carbon gasification ratio of landfill leachate that has studied by Weijin et al. and Molino et al. show in figure 8 below. Carbon gasification is affected by the type of reactor, temperature, and addition of the catalyst. The result indicates that the increase in temperature could affect on increasing carbon gasification efficiency. Also, the batch reactor shows better performance than continuous flow (tubular reactor). The batch reactor process has some superior features that impact reaction processes such as easier and more accurate flow and sampling, more straightforward determination of mass balances[32].
5.2. Pollutant Efficiency Removal

The pollutants such as TOC, BOD, and TN can be reduced obviously by using the supercritical gasification process, as shown in figure 9. Several compositions of leachate can dissolve and transform into the gas phase (syngas) because of the increasing ability to dissolve and diffusion at supercritical water [33]. The organic compound is more easily fractionated in supercritical water conditions than another thermal process [34].

![Figure 9. Pollutant efficiency removal of SCG](image_url)

The results show the significant efficiency of removal pollutants among certain technology supercritical gasification of leachate. The highest value of removal TOC composition is studied by Gong et al. [23], with the value of is 95.8% removal efficiency. This result is caused by the high operational condition of 723°C and 25 MPa. Water in supercritical condition has good transportability with low viscosity and high diffusivity that all parameter is contributed to dissolving several organic compounds [35]. Paula et al. give the best result of removal COD composition, which value is 98.8% removal efficiency, which is caused by the presence of ozonation processing and water oxidation.

COD, TOC, and TN are heterogeneous compounds that can be pollutant if it is not treated well. Supercritical gasification has the potential to treat several pollutants contained in leachate. However, the performance of supercritical gasification treats TN not yet satisfactory. Furthermore, it is required to combine with other processing treatments such as biological treatment or chemical treatment for better performance of removal pollutants.

6. Conclusion and Future Development

Landfill leachate using supercritical gasification is showed high content of syngas and good removal pollutant efficiency. The addition of catalyst promotes to higher production of H₂ and CH₄ that is based on water gas shift (CO+H₂O→H₂+CO₂) and methanation reaction (CO+3H₂→CH₄+H₂O). Increasing the temperature could affect more production of H₂ and CO₂. The type of reactor, such as transpiring wall and continuous tubular flow, can affect more production of hydrogen because this process occurs continuously, but it is needed more cost of installation. Actually, the batch reactor can produce more hydrogen, but it is not suitable for a continuous process.

SCG process is a feasible process to process very wet feedstock like leachate or algae. Landfill leachate supercritical gasification also reduces harmful pollutant, and it will be safe to release for local environment but its needed any research further. Hydrogen from landfill leachate supercritical gasification can be used for fuel cell application in long term scenarios.
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