Supercritical Water Gasification of Oily Sludge and Its Life Cycle Assessment

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A supercritical water gasification strategy was employed to investigate the effects of different conditions on hydrogen-rich gas production from oily sludge, based on the Aspen Plus platform. Meanwhile, the entire process was assessed with a life cycle assessment (LCA) method. The results indicated that the gas yield was decreased by increasing the sludge concentration. The yield of hydrogen-rich gas (H_2 and CH_4) was improved with increasing temperature. Higher temperatures were more favorable to gas production. Excessive addition of oxidants could reduce the CH_4 yield, thereby lowering the energy efficiency of the process. The LCA analysis found that, in comparison to the stages of raw material transportation, heat recovery, and wastewater treatment, the effect of more CO_2 produced in cooling and separation stage on global warming potential (GWP) was more obvious. The corresponding process can be improved to mitigate the environmental effect of the whole gasification.

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Keywords: Oily sludge; Supercritical water gasification; Aspen plus; LCA

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INTRODUCTION

Oily sludge is a byproduct of petroleum processing during oil exploitation, transportation, and refinery operations. It usually contains petroleum hydrocarbons, water, and solid particles. This means that oily sludge specimens have characteristics of both oil resources and hazardous waste (Chen *et al*. 2022). The hazardous substances include toxic heavy metals and carcinogenic polycyclic aromatic hydrocarbons. Because of the toxicity, oily sludge exhibits a persistent environmental risk. In China and many other countries, it is classified as hazardous waste. Meanwhile, with high organic content, the quality of the recovered oil is high, and it can be used as an oil resource (Hu *et al*. 2020; Hochberg *et al*. 2022; Xie *et al*. 2023). Therefore, the treatment and reuse of oily sludge has become a focus direction in recent years.

Currently, there are various methods for oily sludge treatment, including biodegradation, pyrolysis and gasification, wet oxidation, and incineration. However, its high viscosity and water content result in low efficiency and high energy consumption when using traditional treatment methods (Hu *et al*. 2013; Li *et al*. 2021; Hochberg *et al*. 2022). Oily sludge often comprises a stable water-in oil emulsion. This makes the dewatering process challenging and requires high input of energy (Huang *et al*. 2024), especially in the gasification process. Fortunately, an alternative treatment approach can be used: Supercritical water can be employed as a green medium that does not require dewatering process now is widely used in oily sludge treatment.

Supercritical water gasification (SCWG) is a chemical process conducted *via* supercritical water conditions, using high-temperature and high-pressure water to convert organic substances in oily sludge into gaseous and liquid products, with the aim of realizing a harmless treatment for the oily sludge. The main advantage is that oily sludge, with a natural water content of 80 % or more, can be converted without drying.

The energy required for heating up the relatively high water content can be recovered by a heat exchanger, which is very important for the overall energy balance. The high water content of the substrate seems to be a significant hardship. Lots of energy is needed to heat the aqueous substrate up to above 500 ℃. Hence, this process usually is combined with a heat recovery or utilization unit for largely recovering energy. Compared to traditional treatment methods, SCWG offers other advantages, such as high efficiency, no need for chemical additives, minimal carbon dioxide emissions, and the generation of useful energy and resources (Jiang *et al*. 2020; Li *et al*. 2020; Zhang et al. 2023). Researchers have conducted experiments using supercritical water for hydrogen-rich gas production from various organic waste materials, including pig manure (Ren *et al*. 2022), kitchen waste (Su *et al*. 2020), sludge (Gong *et al*. 2022), and biomass (Wang *et al*. 2022). Enhancing gasification efficiency and hydrogen production are important for the commercial application of supercritical water for hydrogen-rich gas production. Previous studies have made considerable efforts in optimizing reaction parameters (temperature, pressure, feedstock concentration, and reaction time) and catalyst development (Liu *et al*. 2022; Soltani *et al*. 2023), yielding some notable results.

Life cycle assessment (LCA) is a widely recognized global environmental assessment and management tool (Wang *et al*. 2019b). It integrates the environmental impacts of a product from raw material extraction to final disposal, providing a comprehensive evaluation of a product's environmental footprint and enabling precise assessments of environmental factors (Peng *et al*. 2023). This is attained by quantifying the discharges and emissions that could influence the environment, such as global warming potential (GWP), acidification, and eutrophication (Liu *et al*. 2023).

The LCA study covered the entire life cycle of the transporting and gasifying/degrading of the oily sludge. Its aim was to analyze and quantify the resource consumption and environmental emissions associated with the oily sludge degradation at various stages of its life cycle. This helps to identify the stages that had the most significant environmental effects, from the sludge transportation to the treatment stage, to achieve process improvements to realize greener and cleaner production (Liu *et al*. 2023).

Based on the prior research, Aspen Plus software was used to investigate the effects of various sludge concentrations, temperatures, and oxidation coefficient on the gas product formation during supercritical water gasification of oily sludge. Subsequently, using the LCA method, the environmental impact of the entire gasification process was assessed, and the specific stage was determined to improve the process to minimize the environmental effect of the whole gasification. This study can provide theoretical support for the efficient and eco-friendly degradation of oily sludge.

EXPERIMENTAL

Materials

The main components of the oily sludge, as shown in Table 1, were derived from measurements taken in the authors' previous studies. The characterization analysis of the dried oily sludge had been conducted through approximate and ultimate analysis, which were measured using an elemental analyzer (FlashEA 1112; Thermo Fisher Scientific, Corston, UK and an approximate analyzer (Thermo Fisher Scientific, Corston, UK; 5E-MAG670), respectively. The high heating value of the dried sludge was determined using the same method. In the Aspen Plus software (AspenTech, Aspen Plus V11, MA, USA) simulation process, oily sludge was defined as a non-conventional solid. Dried oily sludge was mixed with water in mixer M1 to form an oily sludge slurry.

Table 1. Chemical Composition of the Oily Sludge

Moisture (%)	Ash (%)	Ultimate Analysis (%)					HHV (MJ/kg)
		C	H	N	S	O	
3.68	61.32	28.18	0.66	1.55	4.87	3.43	9.970

HHV: high heating value

Aspen Plus Simulation

In this study, the PR-BM (Cao *et al*. 2017) method, known for its good thermodynamic property predictions for hydrogen gas production, was chosen. The gasification reaction system for oily sludge is illustrated in Fig. 1. Initially, sludge with a flow rate of 150 kg/h and water with a flow rate of 850 kg/h were mixed and homogenized in a mixer under conditions of 25 °C and 0.1 MPa, and then pressurized to 24 MPa using a pump. The raw materials entered a heat exchanger (HX) and an electric heater (EM) and were heated to the reaction temperature. Simultaneously, oxygen, pressurized to 24 MPa at 25 °C, was introduced into the yield reactor (RYIELD) using a pump (P_{ox}). In the yield reactor (RYIELD), the sludge was reformed into elemental species (C, O_2, H_2, N_2, and S), and then reacted with the input oxygen in a Gibbs reactor (RGIBBS) based on the principle of minimizing Gibbs free energy (Cao *et al*. 2018; Hantoko *et al*. 2019; Magdeldin and J¨arvinen 2020).

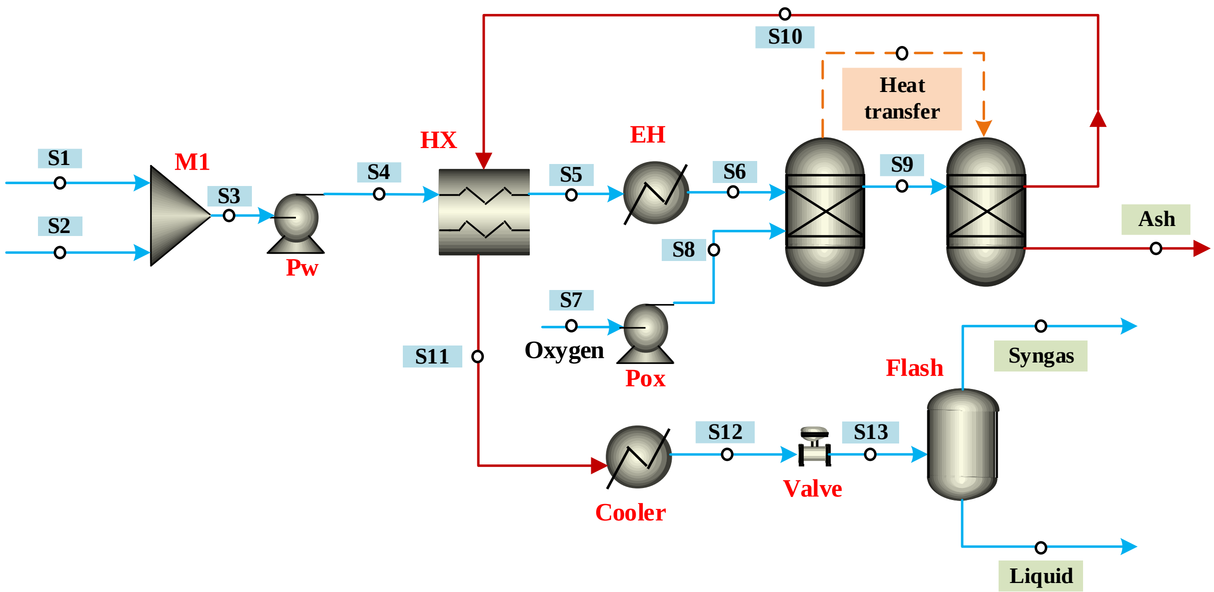

Fig. 1. The flow diagram of the oily sludge gasification and degradation

Possible reaction products at the outlet of the Gibbs reactor were defined as H_2O, CO, CO_2, N_2, N_2O, NO, NO_2, NH_3, SO_2, SO_3, H_2, CH_4, C_2H_4, C_2H_6, and solid carbon (Zhang *et al*. 2017; Wang *et al*. 2019a; Yuksel *et al*. 2019). At this stage, the product stream from the RGIBBS reactor had a high temperature, and it was preheated in the heat exchanger (HX) to recover and utilize the residual heat. Finally, the reaction product stream was cooled to 25 °C in a cooler, reduced to 0.1 MPa through a valve, and separated into gas and liquid phases in a FLASH (Thermo Fisher Scientific, Corston, UK) unit. The parameters for the gasification and degradation reactor are provided in Table 2.

Table 2. Reactor Parameter Settings

Equipment	Specification
Pump	Efficiency: 0.8; Pressure: 24 MPa
Heat exchanger	Cold flow: S4, S5; Hot flow: S10, S11; Preheated temperature: 200 °C
Electric heater	450 to 560 °C
RGibbs	Temperature: 450 to 560 °C; Pressure: 24 MPa
Cooler	Temperature: 25 °C
Valve	Pressure: 0.1 MPa
Flash	Temperature: 25 °C; Pressure:0.1 MPa

Life Cycle Assessment

The life cycle environmental impact assessment process for the gasification and degradation of the oily sludge followed four steps defined in accordance with the ISO 14040 (2006) standards, which were as follows: (1) Goals and scope definition; (2) Life cycle inventory (LCI); (3) Life cycle impact assessment (LCIA); and (4) Interpretation of the results.

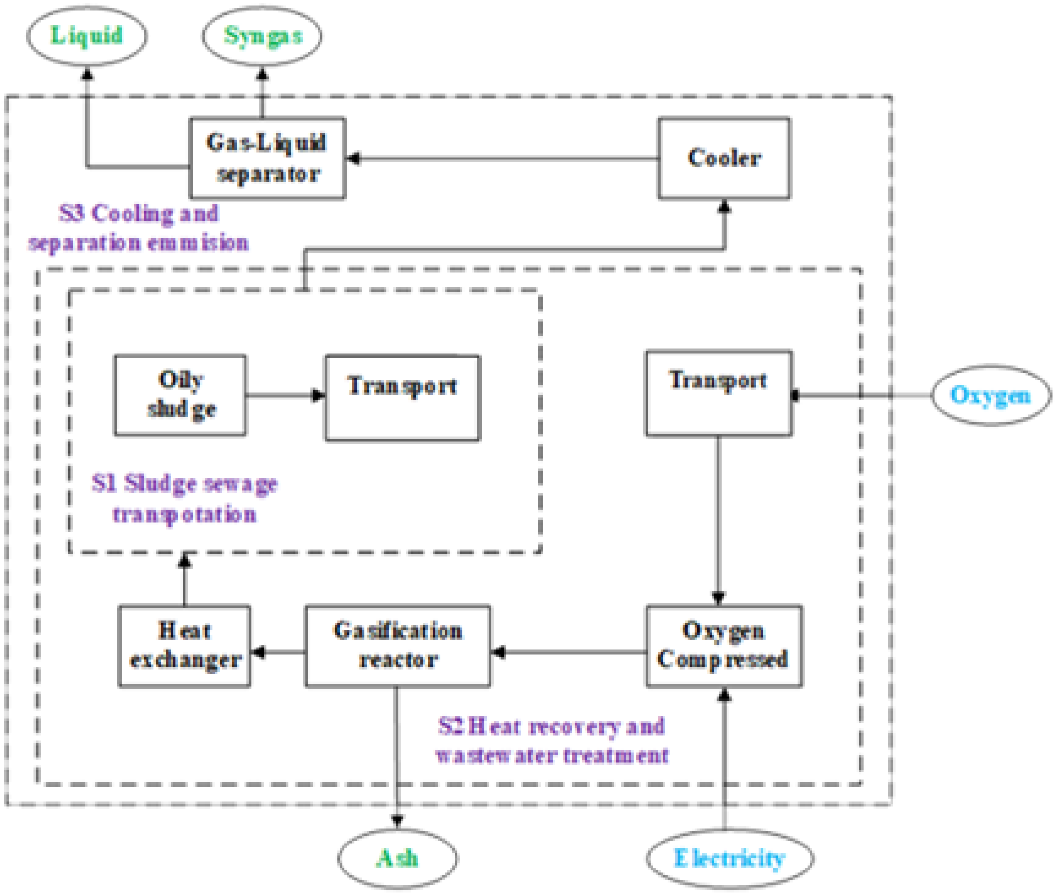

Fig. 2. System boundary for the oily sludge gasification and degradation process

Goals and scope definition

In LCA, the entire life cycle of a product, from cradle to grave, is typically considered as the system boundary for the research. Because the usage stage of the output products from the gasification and degradation processes of oily sludge was not addressed in this study, the scope of the assessment range was settled as from "cradle" to "gate." Figure 2 defines the system boundary for the entire life cycle of the oily sludge gasification and degradation system. This boundary primarily included S1: Oily wastewater transportation, S2: Heat recovery and wastewater treatment stage, and S3: Cooling and separation stage.

Life cycle inventory

Once the purpose and scope of the analysis inventory have been established, the data inventory can provide detailed analysis information for the LCA. In this study, based on the data obtained from the gasification and degradation process of the oily sludge, a life cycle inventory was created, as presented in Table 3.

Table 3. Life Cycle Inventory

Input	Value	Unit
Sludge sewage	150.000	kg
Water	850.000	kg
Oxygen	11.400	kg
Electricity-Pump	8.312	kWh
Electricity-Heater	565.121	kWh
Electricity-Oxygen	3.000	kWh
Output	Value	Unit
CO	0.037	kg
CO_2	80.847	kg
H_2	0.430	kg
CH_4	26.955	kg
Ash	91.974	kg

RESULTS AND DISCUSSION

Gasification Results

Effect of sludge concentration on gas production

The sludge concentration is a key parameter influencing the products of the gasification. Sludge with a relatively low water content normally might contain high organic matter, from which more value-added products can be recovered. In principle, such compounds might be utilized by degradation of the oily sludge in the SCWG process. However, excessive sludge concentration can lead to issues such as pipeline blockages. When the water content in oily sludge is less than a certain threshold (81.7%), severe carbon accumulation will occur, with over 10% (w/w) of the carbon in the sludge being converted into carbon deposits, then causing blockage in reaction system. Therefore, the selected sludge concentration was 5% to 15% (w/w) to avoid the pipeline blockage. The effect of the different sludge concentrations on the gasification and degradation products of the oily sludge is shown in Fig. 3.

The conditions for the gasification reaction included a temperature of 500 °C, pressure of 24 MPa, and an oxidation coefficient of 0.1. Gas production was defined as the ratio of the molar flow rate of product gases to the mass of wastewater treatment. As shown in Fig. 3, as the sludge concentration increased, the total gas yield gradually decreased. The yield decreased from 13.2 mol/kg of the sludge processed to 1.42 mol as the sludge concentration changed from 5% (w/w) to 15% (w/w). The reduction in H_2 production was more obvious compared to the decrease in CO_2 production, which ranged from 12.2 to 17.5 mol/kg. This might be attributed to the decreasing water content in the system, which hampers the water-gas shift reaction responsible for H_2 generation. Simultaneously, the reduction in water content promoted methane formation in the reaction. Therefore, as the sludge concentration increased from 5% (w/w) to 15% (w/w), CH_4 production was predicted to increase from 5.87 to 11.2 mol/kg.

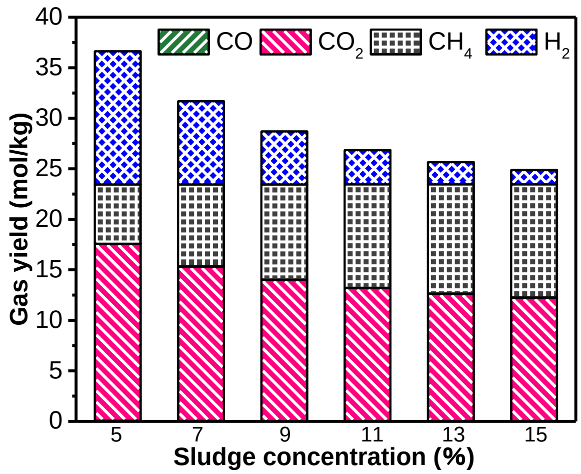

Fig. 3. Gas yield in different system

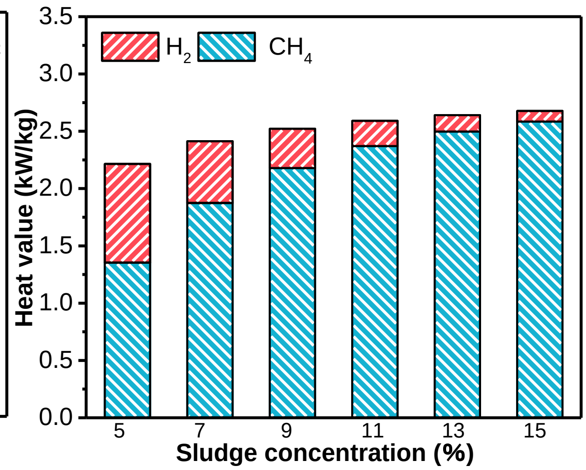

Fig. 4. Energy output in different system

The higher heating value of the fuel gases is defined as the ratio of the heating value of fuel gases to the mass of the sludge processed, and it is shown in Fig. 4. The chemical heats of H_2 and CH_4 were 32.7 and 14.4 kW/kg, respectively. When the sludge concentration was 15% (w/w), each kilogram of sludge could generate 2.68 kW of heat. As the sludge concentration increased, the total heat output of the system increased. This phenomenon was more pronounced when the sludge concentration was within the range of 5% (w/w) to 9% (w/w), going from 2.21 to 2.52 kW/kg.

Higher water content can increase the yield of H_2, as seen from Fig. 4, but it can also raise the cost of the oily sludge gasification process. From an energy output perspective, low sludge concentration was not conducive to the production of methane, as it reduced the energy efficiency of the system. Therefore, when applying SCWG strategy to oily sludge degradation process, it was suggested that the selected sludge concentration must be 15% (w/w).

Effect of gasification temperature on gas production

Temperature is one of the key factors influencing the reaction rate and product distribution in supercritical water gasification. An elevated temperature can enhance the reaction rate, and under high temperature, supercritical water can more efficiently convert organic substances into gaseous products, such as hydrogen and methane, thereby increasing the energy recovery efficiency.

The effect of different gasification temperatures (460 to 540 °C) on the degradation products of the oily sludge are shown in Fig. 5. The other gasification conditions were pressure 24 MPa, sludge concentration 15% (w/w), and oxidation coefficient 0.1. As shown in Fig. 5, the molar yield of the total gas products increased with the temperature increasing, which can be related to the promotion of the free radical reactions in supercritical water system. As the temperature was increased, the organic carbon proportions in the gas phases increased, which indicated that the enhanced temperature would promote the hydrolysis and steam-reforming reaction.

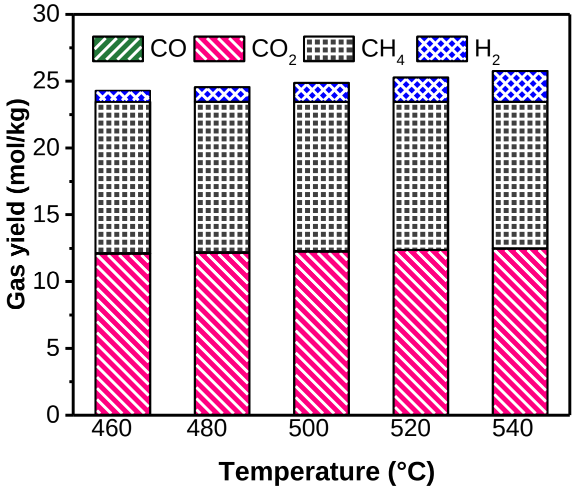

Fig. 5. Gas yield in different temperature

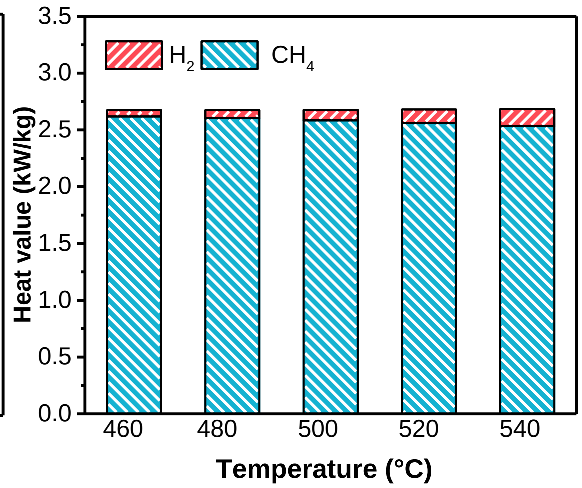

Fig. 6. Energy output in different temperature

Figure 5 shows that the produced CO_2 in this study was relatively high. This may be due to the lack of any catalyst being used in the reaction to promote the formation of H_2. Gong *et al*. (2022) reported that CO_2 yield obtained in gasification of sewage sludge was high without catalyst addition. After using the catalyst, the H_2 yield was increased, with the gradually decreased CO_2 production. The high CO_2 content also may be caused by the low reaction temperature. The free-radical reaction will be more conductive for H_2 formation when the temperature exceeds 600 °C. Zhang *et al*. (2016) reported that H_2 production was highest in the temperature range of 590 to 660 °C.

The overall energy output of the system changed from 2.67 to 2.68 kW/kg, with a minimal variation of 0.01 kW/kg as the temperature changed from 460 to 540 °C (Fig. 6). This minor increase of the energy output may be caused by the enhanced yield of the hydrogen at higher temperatures.

Within the temperature range of 460 to 540 °C, the endothermic reaction of steam reforming will intensify as the temperature increases and more hydrogen is generated. Under mild temperature conditions, the decarboxylation reaction intensifies, and it is more prone to form CO_2. Thus, it is predicted that a mild temperature is not conducive to the formation of hydrogen and is more facilitated to the production of CO_2.

In contrast, considering the ability of the reaction facility to resist high temperature and high pressure under actual conditions, the temperature of the reaction system should be controlled within the tolerable range of the reaction device. Therefore, the optimal sludge treatment temperature can be selected as the middle reaction temperature at 500 ℃ .

Effect of oxidation coefficient on gas production

The oxidation coefficient is a critical factor that influences gasification products. Different oxidation loadings can lead to different reaction rates influencing the formation of the products.

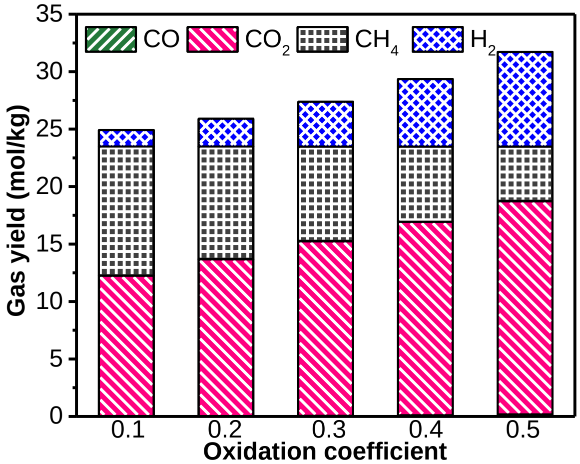

Fig. 7. Gas yield in different system

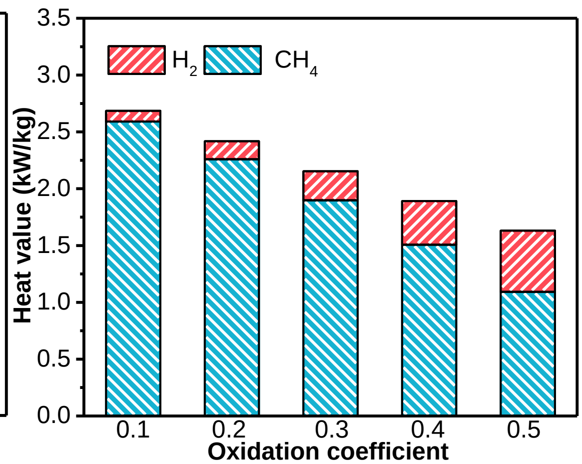

Fig. 8. Energy output in different system

Figure 7 shows the molar flow rates of the products in the gasification and degradation system of oily sludge at a temperature of 500 °C, a pressure of 24 MPa, and a sludge concentration of 15% (w/w), with variations in the oxidation factor from 0.1 to 0.5. When more oxidant was provided, the yield of gasification products CO_2 and H_2 increased, while the yield of CH_4 decreased. The total gas yield increased from 24.4 to 31.53 mol/kg. With the increase in oxidant, the yield of CO_2 rose from 11.33 to 17.87 mol/kg, gradually becoming the primary carbon products. Combustible gas H_2 increased from 1.4 to 8 mol/kg, while CH_4 decreased from 11.33 to 4.33 mol/kg. Therefore, the increase in hydrogen yield comes at the cost of reduced methane production.

It is clearly observed in Fig. 8 that the oxidation coefficient noticeably affects the system’s energy output. The system’s energy output decreased from 2.69 to 1.63 kW/kg. Therefore, excessively high oxidation coefficients were detrimental to the system’s energy output.

The addition of a small amount of oxidant led to a predicted considerable increase in CO_2 production and a predicted increase in H_2 yield. A higher oxidation coefficient was more favorable for the formation of hydrogen, but also resulted in more carbon dioxide generation, leading to a decrease in the energy density of the produced gas. Hence, the optimal sludge treatment oxidation coefficient can be selected at 0.1, taking a comprehensive approach to promote gas production efficiency and quality while minimizing environmental pollution.

Life Cycle Impact Assessment

In this section, the CML-IA baseline method was used for LCA assessment process (Frontera *et al.* 2020). The CML-IA is a database that contains characterisation factors for life cycle impact assessment. It is the most common impact categories used in LCA. This assessment quantified the system’s environmental effects into several impact categories. The primary focus of this study was the global warming potential (GWP) environmental indicator.

Global warming potential

Based on the optimal process parameters obtained in the above section, which included: a sludge concentration of 15% (w/w), gasification temperature 500 °C, and an oxidation coefficient 0.1, an LCA model to focus on analyzing the environmental impact category of GWP was established.

The GWP is an indicator used to measure the impact of different greenhouse gases on the global climate environment during their cumulative emissions. The intergovernmental panel on climate change (IPCC) proposed the GWP model, which is calculated in terms of CO_2-equivalents (eq.), providing a standardized method for converting emissions of different types of greenhouse gases into equivalent CO_2 emissions. This quantifies the contribution coefficients of various emissions to global warming. The GWP is a quantitative metric that helps scientists, policymakers, and industry professionals better understand the impact of various emissions on global climate change and take specific actions to reduce and control greenhouse gas emissions. The primary emission factors and characterization factors for GWP, as obtained from the SimaPro database, are presented in Table 4.

Table 4. Major Emission Factors and Characteristic Equivalent for GWP

Emission Factor	Characteristic Equivalent (kg CO_2 eq)		
	20 year	100 year	500 year
CO_2	1	1	1
CO	1.5714	1.5714	1.5714
CH_4	62	25	7
NO_2	275	296	156

Based on the quantified GWP characterization values for each production stage of the gasification and degradation of oily sludge, the GWP results were calculated using SimaPro software (PRé Sustainability, SimaPro 9.2, Amersfoort, Netherlands), as shown in Fig. 9.

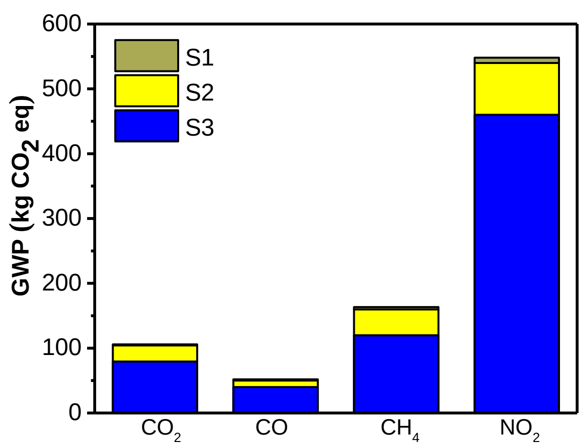

Fig. 9. GWP characteristics value in different stages

The GWP values for each stage of the oily sludge gasification and degradation system were 15, 155, and 699 kg CO_2 eq, respectively, with the GWP contributions in the order of S3 > S2 > S1. The largest contribution to greenhouse gas emissions comes from the CO_2 generated during the cooling and separation stage (S3). This is mainly due to the production of CO_2 in the S3 stage. Therefore, future research should focus on further utilizing the gases generated in the gasification and degradation process to reduce greenhouse gas emissions.

Sensitivity analysis

In the LCA model, there is a certain degree of uncertainty in the data. Therefore, sensitivity analysis is necessary to verify the impact of changes in system input or output parameters on LCA results. This study employed the commonly used local sensitivity analysis in LCA research to examine the effects of variations in key gasification process parameters, including temperature, sludge concentration, and oxidation coefficient on LCA results. As shown in Fig. 10, increasing sludge concentration could lead to a gradual decrease in GWP values, indicating a reduction in the negative environmental impact of the gases produced during the gasification and degradation process in these two impact categories. Therefore, increasing the sludge concentration during gasification can reduce the negative environmental impact. The effects of temperature on the GWP in different gasification stages are shown in Fig. 11. There was a slight decrease in GWP values as the temperature increased. Therefore, the impact of temperature on GWP in the context of the gasification and degradation of oily sludge was relatively minor.

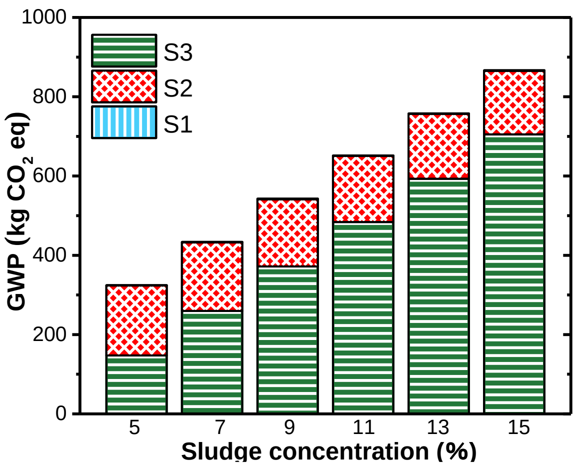

Fig. 10. Effect of sludge concentration on GWP

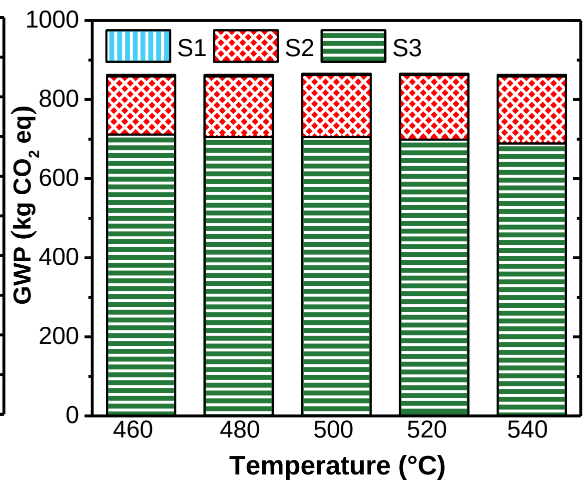

Fig. 11. Effect of temperature on GWP

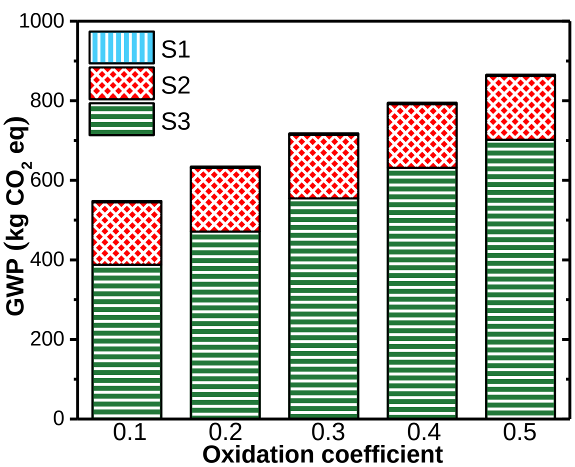

Fig. 12. Effect of oxidation coefficient on GWP

As the oxidation coefficient increased, GWP values gradually rose (Fig. 12), indicating an increase in the negative environmental impact of the gases produced during the gasification and degradation process in these two impact categories. Hence, in the gasification and degradation of oily sludge, it is necessary to control the value of the oxidation coefficient to lower GWP values and minimize the negative environmental impact.

CONCLUSIONS

In this study, based on previously published data, the gasification and degradation processes of oily sludge were simulated using Aspen Plus software, and gas production predictions under different conditions were obtained. Based on the optimal gas production process conditions, a life cycle assessment of the treatment process was carried out, analyzing the process impact on global warming. Additionally, a sensitivity analysis of the LCA results was performed. The main results were as follows:

1. Under the conditions of a temperature of 500 °C, sludge concentration of 15% (w/w), and an oxidation coefficient of 0.1, the gasification of oily sludge achieved optimal gas production. The total gas yield was considerably influenced by the sludge concentration. A small amount of oxidation was beneficial for sludge gasification; however, more loading of oxygen may reduce the system's output of the fuel gas.

2. Under the optimal parameters, LCA analysis results revealed that in the three stages of oily sludge gasification, the stage with the highest contribution to the global warming index was the cooling and separation stage (S3).

3. Sensitivity analysis of the primary influencing factors in the gasification process indicated that in the process of oily sludge gasification, it is necessary to optimize environmental impact factors by adjusting parameters such as temperature, sludge concentration, and oxidation coefficient. Comprehensive environmental management and monitoring should be carried out in the practical implementation of the process.

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