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# Maximizing Bio-Hydrogen and Energy Yields Obtained in a Self-Fermented Anaerobic Bioreactor by Screening of Different Sewage Sludge Pretreatment Methods

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**Abstract:** Egypt faces significant challenges in managing its sewage sludge generated in large quantities from wastewater treatment plants. This study investigates the feasibility of utilizing sewage sludge as a renewable resource for hydrogen production through anaerobic digestion at the 100 L bioreactor level. Hydrogen is considered a promising alternative energy source due to its high energy content and environmental benefits. To optimize the microbial degradation process and maximize hydrogen production from sewage sludge, a specialized pretreatment is necessary. Various pretreatment methods have been applied to the sewage sludge, individually and in combination, to study the bio-hydrogen production from sewage sludge. The four methods of treatment were studied in batch assays as a pilot scale. Thermal pretreatment of sewage sludge significantly increases bio-hydrogen production yield compared to other sewage sludge pretreatment methods, producing the highest $H_2$ yield (6.48 $LH_2$/g VS). In general, the hydrogen yield of any type of pretreated inoculum was significantly higher than the untreated inoculum. At the same time, alkaline pretreatment improved the hydrogen yield (1.04 $LH_2$/g VS) more than acid pretreatment (0.74 $LH_2$/g VS), while the hydrogen yield for the combination of pretreatments (shock alkali pretreatment) was higher than both (1.73 $LH_2$/g VS), On the other hand, untreated sewage sludge (control) had almost no hydrogen yield (0.03 $LH_2$/g VS). The self-fermented anaerobic bioreactor improved sewage sludge utilization, increased bioenergy yields, and seems to be promising for treating complex wastes at this scale.

**Keywords:** bio-hydrogen; CSTR; dark fermentation; pretreatment; sewage sludge

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## 1. Introduction

According to the Central Agency for Public Mobilization and Statistics organization, Egypt had over 100 million inhabitants in 2021, making it one of the most populous countries in Africa and the Middle East. The exponential population growth has led to a substantial increase in water consumption, resulting in a significant volume of wastewater during its final cycle process. To address this issue, it is essential to ensure that the wastewater undergoes proper treatment in wastewater treatment plants, particularly addressing the byproduct known as sewage sludge.

Sewage sludge is composed of diverse organic and inorganic constituents and is considered to be a significant societal issue due to the country's population. In Egypt, sewage sludge is generally processed and disposed of in landfills or used as fertilizer [1]. Regarding the situation in Egypt, it has been recently reported that Egypt produced nearly 2.1 million tons of sludge in 2018 in Greater Cairo [2] [3]. The Egyptian situation pushes the researchers to find a solution for achieving the sustainability of the sanitation sector via the proper treatment of sewage sludge [4].

Anaerobic digestion (AD) is a common method for the co-digestion of sewage sludge with different substrates [5–7]. This process usually produces biogas that forms a mixture of methane ($CH_4$), carbon dioxide ($CO_2$), a small percent of hydrogen gas, hydrogen sulfide, and stabilized digestate [8]. Hydrogen, as one of the produced biogases, is considered a valuable energy source. Hydrogen has a higher energy density and is a clean fuel [9]; as a sustainable fuel that generates water and energy, it overcomes the negative effects of the usage of fossil fuels [10]. Hydrogen has many uses as a fuel source for fuel cells and as a feedstock in the chemical industries [11]. Utilizing microbial processes through dark or photo fermentation methods represents an economically viable and highly promising approach for bio-hydrogen production [12]. Recent research findings indicate that the generation of bio-hydrogen via dark fermentation, specifically through the stages of hydrolysis and acidogenesis, represents an efficient approach for harnessing energy from wastewater with a high organic content. Additionally, recent research findings have demonstrated that sewage sludge presents a diminished hydrogen production potential when compared to carbohydrate-abundant substrates, yielding below 1 mmol $H_2$/g volatile solids (VS) [13,14]; this contrasts with 5–10 mmol $H_2$/g VS observed in waste such as agricultural wastes and food waste, as reported by other researchers [15–17].

Challenges in the dark fermentation of sewage sludge lie in the limited availability of easily fermentable substrates [13], low yielding of bio-hydrogen, and the up-scaling of the fermentative bioreactors [18]. It has been reported that dark fermentation for hydrogen production was improved by pretreating the substrate or inoculum, which helps decrease the activity of the bacteria that produce methane and increase the activity of the bacteria that produce hydrogen [19]. Pretreatment methods, such as thermal, chemical, or enzymatic techniques, enhance the accessibility of fermentable substrates for microorganisms by disrupting the cell structures and liberating the substrates [20]. Consequently, this improves the overall hydrogen production during dark fermentation [13,21]. Several pretreatment processes, such as mechanical [22], thermal [23], chemical [24], or irradiation [25], have been proposed to increase the ability of sewage sludge for fermentation. Sewage sludge has been extensively pretreated with thermal and chemical pretreatments in some research [26–28].

Thermal pretreatment is helpful for the growth of hydrogen-producing bacteria after suppressing the activity of methanogens [29]; however, in the chemical treatment processes that were treated by acetylene, iodopropane, methanol and bromo ethane sulfonic acid (BESA), these served as inhibitors and very toxic for bacteria [30]. Some researchers have demonstrated that alkaline pretreatment methods outperform acidic pretreatment techniques as a pretreatment method for anaerobic sludge [31]. Some researchers have integrated the two distinct pretreatment methods to enhance the sludge solubilization process, increasing the availability of fermentable substrates to produce more biogas [32–34]. Despite the lack of a comprehensive investigation comparing the impacts of various pretreatment methods and their combinations on dark fermentative hydrogen generation from sewage sludge, most prior research has focused on batch processes, with only a limited number of studies reporting continuous reactor experiments and up-scaling of the process [35].

Dark fermentation, a biochemical reaction, is influenced by the pH and temperature during the fermentation phase. The ideal pH and temperature for hydrogen generation are unique for sewage sludge [34,35]. The process of dark fermentation involving carbohydrate-rich substrates is typically conducted under mildly acidic conditions ($pH < 6$) to facilitate the growth of hydrogen-producing microorganisms [36]. They possess the ability to suppress rival microorganisms such as methanogenic archaea [37].

The aim of this study was to study the impact of using multi-pretreatment methods to enhance the self-fermented anaerobic sewage sludge for bio-hydrogen production in a 100 L bioreactor. Hydrogen production from a pretreated 100 L sewage sludge bioreactor was studied with different pretreatments and pH to provide the maximum hydrogen yield. To the best of our knowledge, such a study of the effects of different pretreatments on hydrogen production from sewage sludge using an up-scale of 100 L has not been conducted before. Bio-energy yields produced from the different pretreatment methods from this scale have also been assessed in this study, with analysis of the microbiological examination for the ideal case.

## 2. Materials and Methods

### *2.1. Properties of Initial Sewage Sludge and Experimental Setup*

The excess sludge was harvested from the full-scale anaerobic sludge treatment plant existing in the El-Gabal El-Asfar wastewater treatment plant located in Qalyubia government in Egypt. The main characteristics of the sludge are presented in Table 1. To study the availability of producing of bio-hydrogen at a scale of 100 L of sewage sludge, various pretreatment methods were applied to the sewage sludge mixture, as follows: a control (C) experiment where no pretreatment was performed on the sludge; a thermal pretreatment (TP) experiment involving heat treatment of the sludge mixture at 75 °C for 15 min; a shock alkali pretreatment experiment (SALP), where the sludge underwent alkaline treatment with sodium hydroxide (1 N) to reach pH of 11.5 for 4 h, followed by adding phosphoric acid to reach pH 5.5; an alkaline pretreatment (ALP) experiment, where the sludge mixture was treated with sodium hydroxide (1 N) to adjust pH 11.5; and an acid pretreatment (ACP) experiment, where acidification of the sludge mixture was achieved through treatment with phosphoric acid (pH 5.5).

**Table 1.** Characteristics of mixed sewage sludge.

| Parameters | pH | $COD_{tot}$ (g/L) | $VFA_S$ (g/L) | ALK (g/L) | TS (g/L) | VS (g/L) |
|---|---|---|---|---|---|---|
| Initial sewage sludge (Control (C)) | 7.62 | 32.43 | 0.82 | 2.78 | 26.55 | 17.92 |
| Thermal pretreatment (TP) | 6.40 | 39.70 | 0.74 | 2.67 | 21.00 | 14.20 |
| Shock alkaline pretreatment (SKP) | 6.70 | 33.20 | 0.97 | 2.75 | 26.00 | 18.20 |
| Alkaline pretreatment (KP) | 7.40 | 37.87 | 0.86 | 2.62 | 24.70 | 19.11 |
| Acid pretreatment (AP) | 6.90 | 34.23 | 0.94 | 2.84 | 21.00 | 14.28 |

### *2.2. CSTR Setup and Operation*

To produce hydrogen from the mixed sewage sludge, the pilot-scale CSTR with a total volume of 100 L and a working volume of 70 L was utilized. The area of the reactor had three sampling or feeding ports on the side, a bottom port for sludge removal, and 4 holes on the upper side of the reactor were used for gas collection, temperature, and pH monitoring. Mechanical mixing of the CSTR content was carried out by external pumped recirculation. The temperature was maintained by using electrical heaters. The resulting gas was collected into a 25-L steel container, and after that, the gas was received using gas bags, as shown in Figure 1. The CSTR was fed with mixed sewage sludge that had been pretreated and operated in batch mode.

### *2.3. Analysis and Calculations*

Gas production in the continuous stirred tank (CSTR) reactor was assessed on each operating day using the water displacement technique. The pH value was measured using a JENWAY 3510 device (Long Branch, NJ, USA), and chemical oxygen demand (COD) was measured using the HACH method [5]. Volatile fatty acids (VFAs), ammonia nitrogen, total Kjeldahl nitrogen (TKN) (steam distillation, Behr S-1, Dusseldorf, Germany), total solids (TS) (drying oven, DHG-9055A, Dobetter Group of Corporations, Shanghai, China), and volatile solids (VS) (Vulcan A-550, Ransom & Randolph GmbH, Rötha, Germany) were measured according to the standard methods for the examination of water and wastewater [21]. Briefly, total solids were measured by drying the samples at 105 °C for 24 h, and the solid contents were determined from the difference between the sample weights before and after drying. Volatile solids content was calculated from the loss on ignition after ashing the dried residue at 550 °C for 2 h. Volatile solids were a part of the total solids, and some calculations were used from the standard methods for the examination of water and wastewater for measuring these processes [21]. Biogas composition was determined using a portable biogas 5000 gas analyzer (Geotech, Geotechnical Instruments (UK) Ltd., Coventry, England). The bacterial community was identified from the hydrogen reactors using the Biolog GEN III system (BIOLOG, Hayward, CA, USA), as discussed previously in the literature [38].

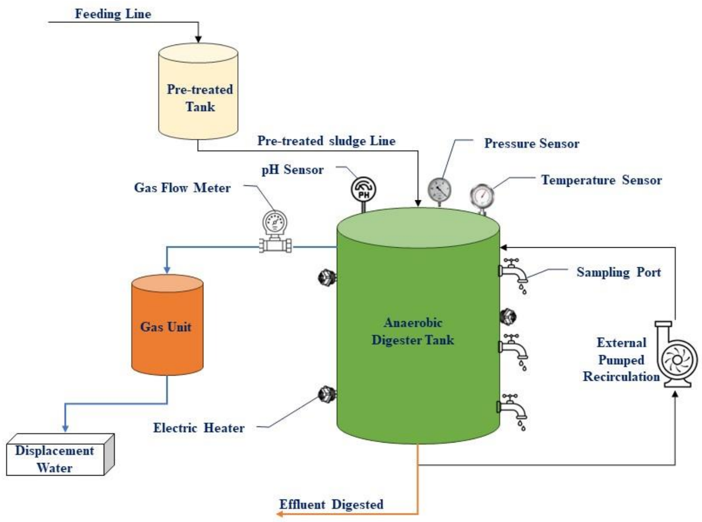



**Figure 1.** Schematic diagram of the CSTR producing hydrogen from mixed sewage sludge.

## 3. Results and Discussion

### *3.1. Bio-Hydrogen Production from a 100 L Sewage Sludge Bioreactor Using Different Pretreatment Methods*

The potential for bio-hydrogen production was evaluated through batch assays on the pretreated mixed sewage sludge with different initial treatments, as described before (C, TP, SALP, ALP, and ACP). The hydrogen yields of the four kinds of pretreated and non-pretreated sludge in anaerobic fermentation are shown in Figure 2. As per the results of the batch experiment, the pretreatment methods used in the mixed sewage sludge leads to production of different bio-hydrogen gas, and consequently, production of different hydrogen yields.

The sludge without pretreatment was used as a control for the whole experiment, producing hydrogen gas and the following hydrogen yield, as shown in Figure 2. It shows that the control batch gave a small amount of hydrogen gas that accounted for 0.03 (L $H_2$/g VS). Figure 2 shows that the hydrogen yield obtained after each pretreatment increased significantly compared to the yield from the raw mixed sludge. Temperature can influence the metabolic activity of fermentative microorganisms, which can affect the hydrogen production rate. Hydrogen is usually produced as a byproduct by fermentative microorganisms in their metabolism, which involves breaking down organic matter in the sewage sludge in the absence of oxygen. These microorganisms' metabolic activity can be influenced by several factors, including temperature [39]. As shown in Figure 2, the highest $H_2$ yield was produced by TP of the anaerobic sludge at 6.48 (L $H_2$/g VS). This indicates that heat pretreatment success fully eliminated $H_2$-consuming bacteria (HCB) and improved the $H_2$ production rate and hydrogen production yield.

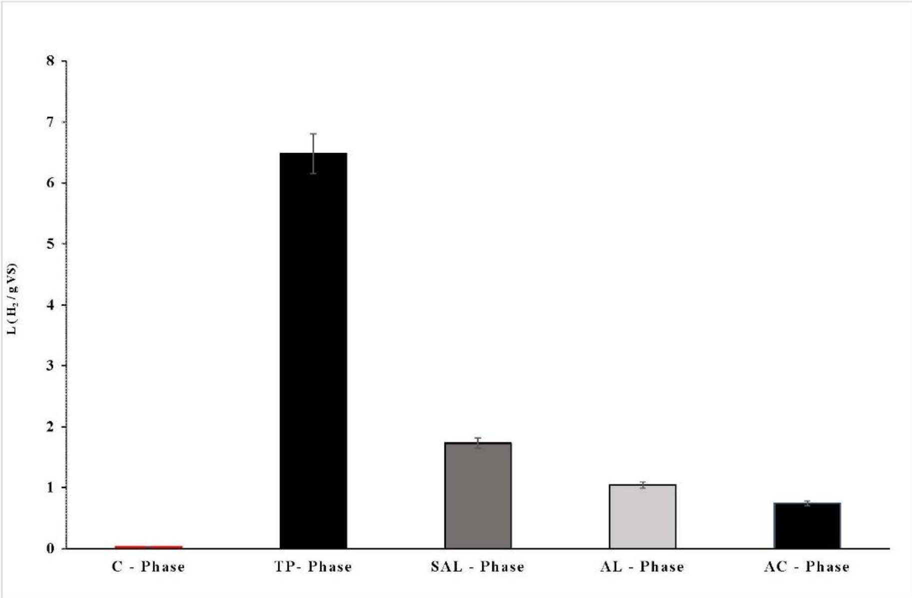



**Figure 2.** Effects of various pretreatments on total hydrogen yields.

Figure 2 also shows the effect of initial pH on the hydrogen yield in the cases of SALP, ALP, and ACP. The variations were significant because the higher pH could further disrupt the microbial cell of sludge and gave different values for the bio-hydrogen gas, as stated by some researchers [30,40]. The hydrogen yield of various pretreated sludge at various initial pH was completely different at this scale. At an initial pH of 5.54 (acidic), the hydrogen yield was 1.73 L $H_2$/g VS; however, the hydrogen yield decreased to 1.04 L $H_2$/g VS when the reactor was working at an alkaline pH (pH = 11.34). These results are better than the results that were provided by Zhang et al. (2007) [41] at pH 4.0, who provided an accumulative hydrogen yield of only 0.15 L $H_2$/g VS at the same pH while undertaking an anaerobic digestion process for cornstalk waste; however, the results of this study were without any substrate and was self-fermented. Another researcher, Antonopoulou et al. (2010), reported an optimum pH range of 4.7–5.3 for bio-hydrogen production from sweet sorghum extract in a continuous stirred tank bioreactor [42], and Assawamongkholsiri et al. (2013), reported a maximum hydrogen production potential of 481 mL $H_2$/L from an activated sludge pretreated with acid (0.5% (w/v) HCl) for 6 h [43]. Another study prepared by Zhang et al. (2003) reported that the optimal initial pH for converting starch to hydrogen was found at 6.0 under thermal conditions [44]. On the other hand, our study results did not correspond to the results obtained by Cai et al. (2004), who indicated that the optimum pH for producing hydrogen gas at the rate of 11.68 mL $H_2$/g VS was 11.5 and, in contrast, when they used a pH of 7.0, the biohydrogen gas was only 1.46 mL $H_2$/g VS [45]. The hydrogen yield was only 0.74 L $H_2$/g VS in the case of shock alkaline pretreatment. These experimental results suggested that all pretreatments could enhance hydrogen production from sewage sludge by anaerobic fermentation at different rates, and the heat pretreatment was the best, with good availability to do this in the real field as well. The values in parentheses for the maximum $H_2$ yield are the incubation pH at which the maximum yield was obtained.

Table 2 compares the maximum hydrogen yield in sewage sludge fermentation. It can be seen from Table 2 that the maximum hydrogen yield in the present work (6.48 (L H2/g VS/100 L)) was higher compared with sewage sludge fermentation in other studies. This result indicated that the real field experiment (100 L) was the best, with a good availability of hydrogen compared to other laboratory studies, as shown in Table 2.

**Table 2.** Comparison with the reported data of hydrogen yield using sewage sludge.

| Substrate | Fermentation Conditions | Maximum Hydrogen Yield | References |
|---|---|---|---|
| Thermal pretreated sludge | Batch 37 °C | 12.23 mL/g-$VS_{added}$ | [46] |
| Ultrasound pretreated sludge | CSTR 36.5 °C | 25.2 mL/g-$VS_{added}$ | [47] |
| Alkaline pretreated sludge | Batch 37 °C | 11.68 mL/g-$VS_{added}$ | [48] |
| Thermal pretreated sludge | Batch 37 °C | 60 mL/g $VS_{added}$ | [49] |
| Thermal pretreated sludge | Batch 36 °C | 20.8–51.7 mL/g VS | [50] |
| Thermal pretreated sludge | CSTR 55 °C | 64.8 mL $H_2$/g VS | our study |

### *3.2. Effect of Pretreatment on the Composition and Characteristics of Mixed Sewage Sludge*

To enhance the bio-hydrogen production, various pretreatments were applied to the sewage sludge—some of them acted individually (thermal, alkaline, and acidic), and one case was in combination (alkaline + acidic) compared to the control experiment. Because the mechanisms of pretreatments were different, the produced bio-hydrogen from the sewage sludge was also different [51]. Table 3 summarizes the effects of the five pretreatments on the sludge after anaerobic fermentation. Because these pretreatments can break part of the bacteria structures and the microbial cells of the sludge, biogases are produced and detoxification of the waste happens, as recorded by many researchers previously [52].

**Table 3.** The effect of different pretreatments on mixed sewage sludge characteristics.

| Item | Control (C) | Thermal (TP) | Shock Alkaline (SKP) | Alkaline (KP) | Acid (AP) |
|---|---|---|---|---|---|
| pH | 7.45 | 6.50 | 6.05 | 8.91 | 5.40 |
| $COD_{tot}$ (g/L) | 27.80 | 27.60 | 23.76 | 25.17 | 22.94 |
| ALK (g/L) | 2.49 | 2.82 | 2.79 | 2.08 | 2.18 |
| TS (g/L) | 21.89 | 15.43 | 27.10 | 34.00 | 18.65 |
| VS (g/L) | 13.98 | 9.88 | 15.99 | 21.76 | 10.26 |
| Max $H_2$ yield (L $H_2$/g VS) | 0.03 | 6.48 | 1.73 | 1.04 | 0.74 |

Hydrogen was produced at an acidic pH and alkaline pH as the lysis of the cell strengthened along with the acidic and alkaline pHs, but not at a neutral pH, as compatible with the results of many previous studies [53–55]. To date, some studies have demonstrated that in alkaline conditions, the VFA production was improved significantly, and the yield was even over 3 or 4 times that at pH 5.0, or under uncontrolled pH conditions [24,56], and this is consistent with the results of our study since the hydrogen productivity in alkaline pretreatment is almost twice as high as in acidic (Table 3). This can be used to explain why the combination of solubilized protein from sludge and the formation of VFAs and ammonia during bio-hydrogen fermentation occurs.

$COD_{tot}$ reduction is a significant parameter for measuring the productivity performance of bio-hydrogen in the study [57]. $COD_{tot}$ reduction was monitored throughout the anaerobic process period. As shown in Figure 3A, each pretreatment process enhanced the reduction of total chemical oxygen demand at the self-fermented bioreactor work, as indicated in Table 3. The thermal pretreatment (TP) achieved the most significant decrease, resulting in a COD total reduction from 39.70 to 27.60 g/L (2.15 times higher than the untreated sludge); however, the decrease in $COD_{tot}$ was from 33.2 g/L to 23.7 g/L in the shock alkaline phase, and the reduction was from 31.8 g/L to 25.1 in the alkaline phase. In the acidic phase, the reduction of COD was recorded from around 30 g/L to around 23 g/L. In the context of hydrogen production from sewage sludge, a decrease in the COD can be advantageous as it indicates that microorganisms utilize the organic matter to generate hydrogen through the process of anaerobic fermentation [58–60].

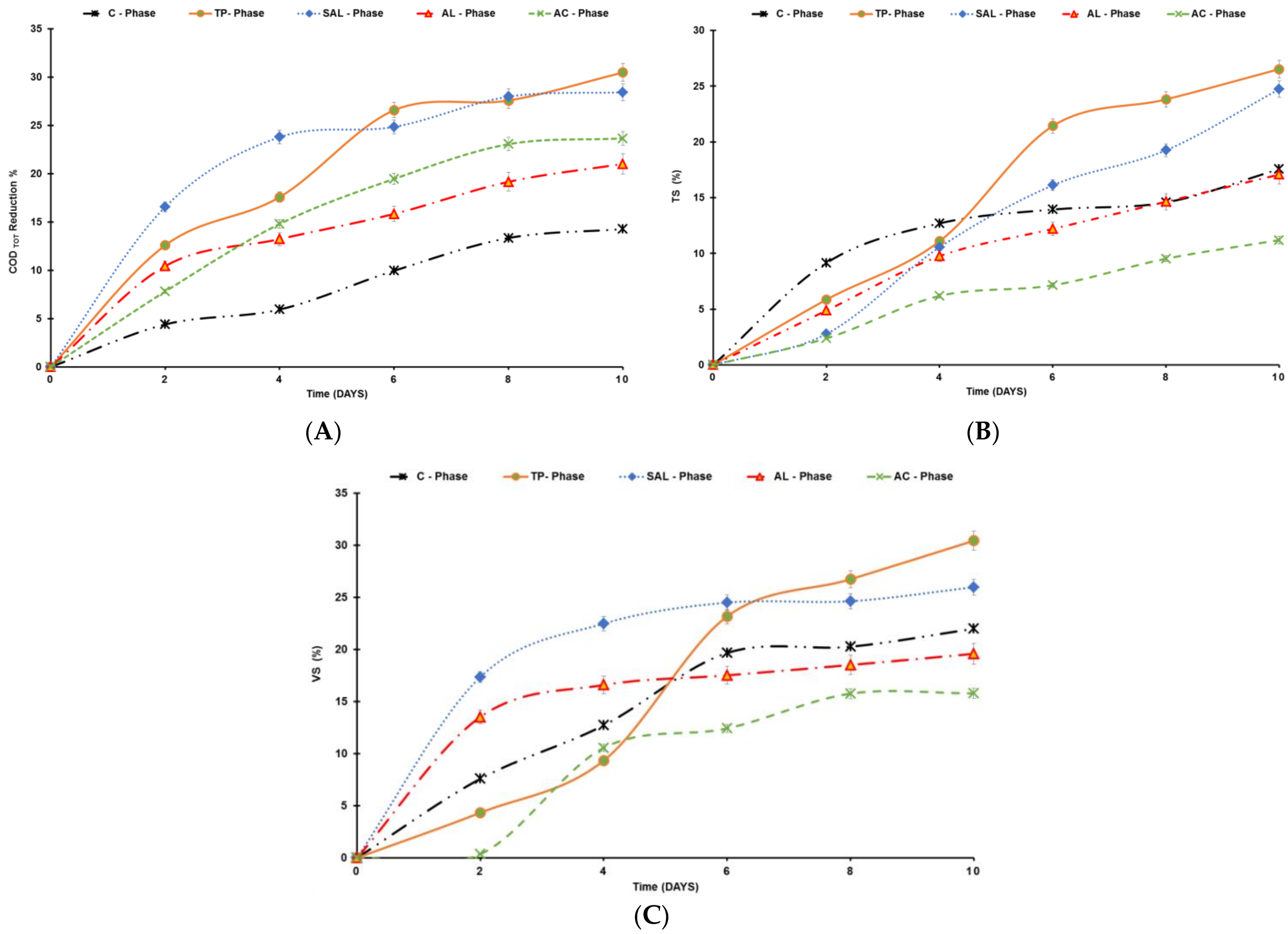


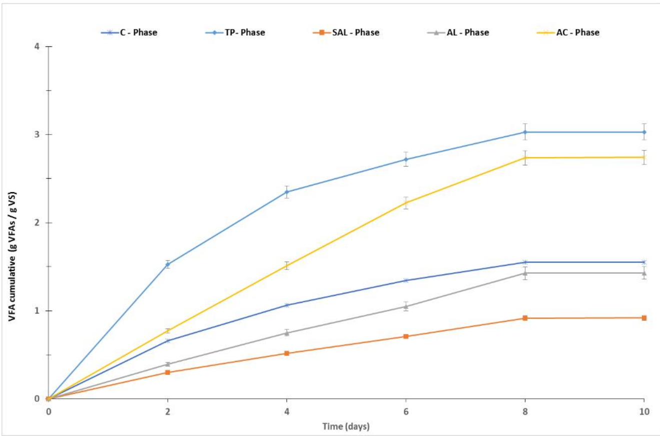

**Figure 3.** The effects of different pretreatments on (**A**) COD, (**B**) TS, and (**C**) VS reductions from the self-fermented CSTR reactor (100 L).

The total solid (TS) content plays a significant role in bio-hydrogen production from sewage sludge through anaerobic fermentation. Optimizing the TS content within an appropriate range is crucial for maximizing the bio-hydrogen yield and overall process performance [61]. Complex organic compounds in the waste material are broken down into simpler compounds during anaerobic fermentation, such as volatile fatty acids and carbon dioxide. The result is a reduction in total solids content (TS), as larger molecules are transformed into smaller ones or gases that can be easily separated from the liquid fraction. As shown in Figure 3B, each distinct pretreatment procedure contributed to an augmentation of the reduction for (TS) by a factor ranging from 11% to 27.5%. The most significant decrease was achieved through shock alkaline pretreatment (SKP), resulting in a TS reduction from 36 to 27.1 g/L; however, the results of volatile solids through the different pretreatments were variable. Volatile solids play a crucial role in the fermentation process as they refer to the organic matter that can be broken down and converted into biogas through anaerobic digestion [62]. As shown in Figure 3C, our study found that each distinct pretreatment procedure contributed to an augmentation of the reduction for (VS) by a factor ranging from 15% to 30.4%, as demonstrated in Table 3. The most significant decrease was achieved through shock alkaline pretreatment (SKP), resulting in a VS reduction from 21.60 to 15.99 g/L (1.38 times higher than the untreated sludge), as indicated in Table 3. This was due to the increase in the available solids in the shock alkaline phase due to the addition of amounts of chemicals, which make it available for bacteria; the same observation was made by El-QLish et al., 2020 [63].

*3.3. Volatile Fatty Acids Production at Different Pretreatments from the Self-Fermented CSTR Reactor (100 L)*

During anaerobic fermentation, microorganisms break down organic matter to produce different byproducts. One of these byproducts is VFAs, which are organic acids with a relatively low molecular weight. VFAs are formed through the breakdown of complex organic compounds. The production of bio-hydrogen through anaerobic fermentation has the potential to have a significant effect on the production and concentration of volatile fatty acids (VFAs). The production of VFAs can depend on several factors, such as the type of organic matter being fermented, pH and temperature of the fermentation environment, and the microbial community present during fermentation [64].

Figure 4 shows the VFA yields and confirms that the yield and the produced VFAs were at the maximum value in the TP phase (2.9 g VFAs/g VS). This value was decreased in the case of ASL to 0.7 g VFAs/g VS, and this was comparable to the case of the AL-phase (1.2 g VFAs/g VS). The VFA yields were 2.6 g VFAs/g VS in the AC phase. In CSTRs, the hydrogen yield was not constant and did not correlate with the concentration of VFAs in the fermentation process, suggesting the occurrence of the homoacetogenesis process, as happened in [65]. Similar results were reported by Wan et al. 2016 [35], who obtained a hydrogen yield of 0.85 mmol $H_2$/g VSS from sewage sludge in a batch under thermophilic conditions (55 °C), and reported that the thermophilic temperature was the optimum case for producing hydrogen from the anaerobic fermenter.



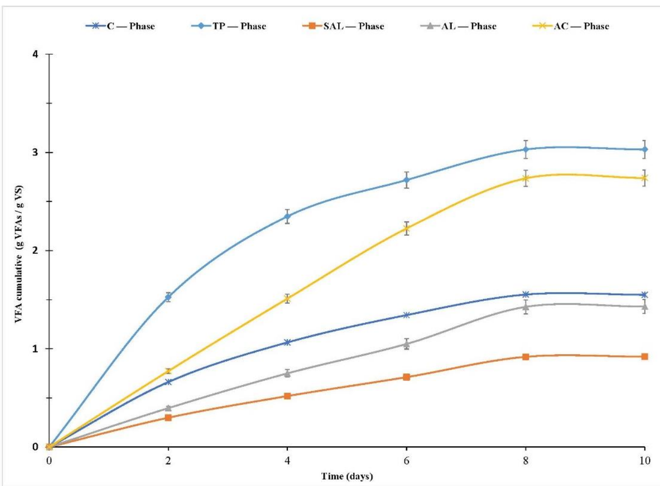




**Figure 4.** The effects of different pretreatment on VFA yields from the self-fermented CSTR reactor (100 L).

### 3.4. Microbiological Examination of Sludge in Thermal Pretreatment as the Optimum Phase

Examination of the microbial community taken from the TP phase, using Biolog GEN III, confirmed the presence of *Bacillus licheniformis* and *Bacillus pseudomycoides*; this observation was compatible with Shah et al. [66], who found that two *Bacillus* sp. strains produced a $H_2$ yield through the fermentation of food waste. These bacterial strains could produce up to 61 mL of $H_2$/g-VS and are considered good biomarkers for the development of relevant $H_2$-producing inoculants; these results are consistent with Hassan et al. [18], who found that the fermentation process to produce more hydrogen could happen through *Bacillus Licheniformis* species reported that the anaerobic digester also contained *Bacillus pseudomycoides*.

### 3.5. Total Energy Yields from the 100 L Sewage Sludge Self-Fermented Bioreactor in Different Pretreatment Methods

Table 4 shows the energy yield values of the self-fermented anaerobic CSTR using the heating values of 120 kJ/g VS fermenter for hydrogen gas [67]. The energy yields from the hydrogen fermenter was 64,300 kJ/g VS fermenter and only 300 kJ/g VS fermenter for the TP and the control phase, respectively, and this confirms that the thermal pretreatment can maximize the energy yields compared to the control phase. The energy yield increased by a factor of 24.6, 33.4, and 57.6 for acidic, alkaline, and shock alkaline phases, respectively, as a result of using the pretreatment in the preceding hydrogen production. Hassan et al., 2022 [7] used a novel technology for increasing the energy yields from the hydrogen fermenter and suggested that the thermal pretreatment would maximize the energy yields from the food waste and using such technologies (electrodialysis), the VFA concentrations will also be increased; however, in this study, the maximization of energy yields happened for a fermenter with 100 L, not 5 L, as in other studies.

**Table 4.** Energy yields from the 100 L sewage sludge self-fermented bioreactor at different pretreatment methods.

| Pretreatment Phase | Hydrogen Yield (mL/g VS) | Energy Yield (K j/g VS) |
|---|---|---|
| Control | 30 | 300 |
| Acidic | 740 | 7400 |
| Alkaline | 1004 | 10,040 |
| Shock alkaline | 1730 | 17,300 |
| Thermal | 6480 | 64,800 |

## 4. Conclusions

The present study systematically reviews, for the first time at a pilot scale, the effect of different sewage sludge pretreatments on dark fermentative hydrogen production from mixed sewage sludge at a bioreactor level 100 L scale. The thermal pretreatment of sewage sludge for the inoculum can increase the bio-hydrogen production yield with the production of the highest $H_2$ yield; however, the bio-hydrogen yield of any type of pretreated sludge was significantly higher than the untreated inoculum, proving the effectiveness of the pretreatment methods on the bio-hydrogen production yield. Overall, the sewage sludge or even industrial sludge is considered to be a very good source for the production of valuable products [68].

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**Data Availability Statement:** The datasets used and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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