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Biohydrogen production using waste activated sludge disintegrated by gamma irradiation

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HIGHLIGHTS

- The waste activated sludge could be disintegrated by gamma irradiation.
- The disintegrated sludge could be used for biohydrogen production.
- Combined alkali-irradiation treatment achieved the highest solubilization of sludge.

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ABSTRACT

The biohydrogen production using the disintegrated and dissolved sludge by gamma irradiation was studied. The experimental results showed that gamma irradiation and irradiation combined with alkali pretreatment could disintegrate and dissolve waste activated sludge for biohydrogen production. The alkali-irradiation treatment of the sludge at pH = 12 and 20 kGy achieved the highest disintegration and dissolution, i.e., it could destroy the cell walls and release organic matters (such as soluble COD, polysaccharides and protein) into the solution. The disintegrated sludge could be used as a low-cost substrate for biohydrogen production.

1. Introduction

Hydrogen is a promising new energy carrier for its environmentally-friendly characteristics [1]. Hydrogen produces only water when it is combusted as a fuel or converted to electricity. Hydrogen can be obtained through various ways, especially from renewable sources like solar, hydro-energy, wind, geothermal and biomass [2–4]. Among various hydrogen production processes, biological method is known to be less energy intensive, for it can be carried out at ambient temperature and pressure. Biological method mainly includes photosynthetic hydrogen production and fermentative hydrogen production. Fermentative hydrogen production has the advantages of high hydrogen production rate and simple operation. In addition, it is of great significance to produce hydrogen from organic wastes by fermentative hydrogen production, because it can not only treat organic wastes, but also produce very clean energy. Therefore fermentative hydrogen production has been received increasing attention in recent years.

Low-cost substrates are especially important for biohydrogen production from the view point of economical consideration. In recent years, different kinds of substrates have been studied for biological hydrogen production, including glucose, sucrose, lactose, xylose [5], sugarcane bagasse [6], wheat and cassava starch [7], sweet potatoes [8], lignocellulosic materials [9–12], microalgal biomass [13–16], industrial wastewater [17–19], municipal solid waste [20–22] and the waste activated sludge from various wastewater treatment plants [23–25]. Hydrogen production from pure substrates may achieve higher production rate, however, using organic wastes as substrate can make the hydrogen gas cheaper and more available. Thus, dark fermentation of organic wastes, including agricultural residues, agro-industrial wastes and organic municipal waste is a promising technology for producing renewable hydrogen [26].

Large quantity of waste activated sludge are produced during the biological wastewater treatment process, their treatment and disposal have become an important environmental problem. However, considering its rich content of organic substances, waste activated sludge is receiving attention for its potential application for different purposes, for instance, as supplementary organic carbon source for biological denitrification [27], as substrate of

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anaerobic fermentation for renewable fuel production [28–30], as fertilizer in agriculture and aquaculture [31]. The biohydrogen production using sludge was studied, but the hydrogen production rate was very low due to its low biodegradability which limits its application as substrate for biohydrogen production.

The main component of activated sludge from wastewater treatment plant is microbial biomass, and the biodegradable organics are encapsulated within microbial cell membranes [32], while the extracellular polymeric substances outside the membranes are non-biodegradable [33]. Therefore, it is necessary to disrupt the microbial cells to release the organic compounds into solution to improve the anaerobic digestion of the waste sludge and to develop the hydrogen production using sludge.

Several methods have been used to pretreat the waste sludge to improve its biodegradability, including mechanical, thermal, chemical, biological and irradiation methods. Mechanical treatment can solubilize the components through physically disrupting the cells [34]. Thermal treatment and microwave disintegrate the chemical bonds of the cell wall and membrane to make the cell content solubilize. Chemical treatment can hydrolyze the cell wall and membrane to release the organic matters [22]. Biological method can disintegrate the activated sludge through the enzyme-catalyzed reactions [35,36].

Ionizing irradiation has been used to treat the activated sludge, which can destroy cellular structure by a number of radical species produced during water radiolysis, and these species can react with the microbial cells with high reactivity [37]. Early studies on applying ionizing radiation for treating waste sludge were carried out in 1970s. However, the main purpose in early studies focused on sludge disinfection [38,39]. With the development of radiation technology, ionizing radiation has been adopted more broadly in treating sludge, such as improving the solubility of sewage sludge [40], eliminating hazardous materials [41], minimizing excess sludge production [42] and enhancing anaerobic treatability of sludge [43,44]. Furthermore, gamma irradiation has been applied in recovering carbon sources from sludge for biological denitrification [27].

Although radiation has been used in treating sludge for various purposes, no study has been performed to explore the possibility of fermentative hydrogen production from ionizing radiation pretreated sludge.

The objective of this study was to investigate the possibility of hydrogen production using disintegrated sludge from gamma irradiation (alone or combined with acid/alkali treatment), the disintegrated sludge may be a low-cost substrate for biohydrogen production.

2. Materials and methods

2.1. Sludge

The sludge used in this study was collected from an aeration tank of a sewage treatment plant located in Beijing, which adopts an A^2/O (anoxic–anaerobic–oxic) process. The physicochemical characteristics of the activated sludge were measured, including pH, settling velocity (SV), suspended solid (SS), volatile suspended solid (VSS), total chemical oxygen demand (TCOD), soluble chemical oxygen demand (SCOD), total phosphorus (TP), total nitrogen (TN), polysaccharides and protein (Table 1).

2.2. Disintegration of sludge

Sludge was divided into three groups, the pH was adjusted to 2.0, 7.0 and 12.0 with NaOH and HCl, respectively, and stored in 1 L sealed bottles.

Table 1
Physicochemical characteristics of the activated sludge.

Item	Average
pH	6.8
SV (mL/100 mL)	52
SS (mg/L)	10,620
VSS (mg/L)	6640
TCOD (mg/L)	7240
SCOD (mg/L)	302
TP (mg/L)	66
TN (mg/L)	22
Polysaccharides (mg/L)	9.8
Protein (mg/L)	50.0

For gamma irradiation, different doses, i.e., 10 kGy, 20 kGy and 30 kGy were applied at ambient temperature. Gamma irradiation was carried out using a ^{60}Co-source in the Institute of Nuclear and New Energy Technology (INET), Tsinghua University, the radioactivity was around 1.26×10^{15} Bq. The absorbed dose was measured using a standard Fricke dosimeter.

2.3. Inoculum

Anaerobic sludge was taken from an anaerobic digester of a sewage treatment plant located in Beijing, and the concentration of mixed liquor volatile suspended solids (MLVSS) of the sludge was 2.42 g/L. The sludge was stored at 4 °C until being used.

Before inoculation, the seeding sludge was pretreated with 5 kGy gamma irradiation at ambient temperature (around 25 °C) to restrain the activity of methanogens and to enrich the hydrogen producers.

2.4. Biohydrogen production

According to the sludge disintegration results, sludge treated at 20 kGy and 30 kGy and pH = 12.0 were used as substrate to examine their hydrogen production possibility.

Batch experiments of biohydrogen production were performed in 150 mL Erlenmeyer flasks, silicone rubber stoppers were used to ensure the tightness. For control test, 0.1 g of glucose was used as sole carbon source. For test groups, 75 mL of 20 kGy irradiation and alkali hydrolyzed sludge (I20), 75 mL of 20 kGy irradiation and alkali hydrolyzed sludge with 0.1 g glucose (IG20), 75 mL of 30 kGy irradiation and alkali hydrolyzed sludge (I30), 75 mL of 30 kGy irradiation and alkali hydrolyzed sludge with 0.1 g glucose (IG30) were used as substrate, respectively. 15 mL seeding sludge mentioned above were inoculated to each batch and all of the groups were filled with de-ionized water to total volume of 100 mL.

Before fermentation, the initial pH of media was adjusted to 7.0 with 1 mol/L HCl and 1 mol/L NaOH, respectively. To ensure the anaerobic environment in each batch, argon gas was passed through the medium for 3 min to drive away the residual oxygen. During the incubation, bottles were placed in a reciprocal shaker (120 r/min) at constant temperature of 36 °C for 48 h. Each batch test was conducted in three replicates.

The cumulative hydrogen production was fitted with the Gompertz equation:

$$H = P \cdot \exp\{-\exp[R_m \cdot e(\lambda - t)/P + 1]\}$$

where P is the hydrogen production potential (mL); R_m is the maximum hydrogen production rate; and λ is the lag time.

2.5. Analytical methods

Samples were collected and filtered through a 0.45 μm filter membrane to obtain a filtrate for analyses. The physicochemical