e-ISSN No.: 2582-4228

Journal of Indian Association for Environmental Management

Journal homepage: www.http://op.niscair.res.in/index/php/JIAEM/index

Enhancement of the Bio-hydrogen Production from Complex Food Wastewater using the Hematite and Nickel Oxide Nanoparticles

Shriram S. Sonawane[a*], Parag P. Thakur[a], Abhijit Gadhe[a]

[a]Department of Chemical Engineering, Nano Research Project Laboratory,
Visvesvaraya National Institute of Technology, Nagpur, MS 440010, India
*Corresponding author mail id: shriramsonawane@gmail.com

Submitted: March 03, 2021 Revised: December 06, 2021 Accepted: December 07, 2021

Abstract: **Nanoparticles are useful to improve the mass transfer rate. In this article, we have explored the potential of the nickel and hematite particles for the bio-hydrogen production increment. Batch fermentation test is used in the experimentations, and distillery wastewater is used as a feed for the process. During the batch fermentation test, the complex distillery wastewater served as a substrate. The current study's findings strongly suggest that adding Fe_2O_3 and NiO nanoparticles together improve hydrogen recovery from complex distillery effluent by 1.2–4.5 orders of magnitude as compared to use only nanoparticles. Ferredoxin, hydrogenase enzymes, oxidoreductase and ferredoxin are important for the increase in hydrogen production.**

Keywords**: Hydrogen yield, hematite nanoparticles, nickel oxide nanoparticles, wastewater**

I. Introduction

The need for novel energy sources is increasing in the modern era. The use of non-renewable energy sources is declining over the globe. The use of hydrogen as an energy source is encouraged by various researchers. Hydrogen energy is considered a better energy option due to its clean nature. However, there is currently no sustainable supply of hydrogen to meet current demand[1]. Dark fermentation of wastewater is a mostly used method for hydrogen production. This method received a lot of attention because of the abundance of substrates and the potential to produce hydrogen energy.[2]. According to Central Pollution Control Board (CPCB, 2009–2010), waste containing a high amount of carbohydrates is considered a complex waste. As a result, proper wastewater treatment is required before disposal. As a result of the abundance of biodegradable sugars in complex distillery wastewater, such as sucrose, glucose, xylose, and hemicelluloses,[2] bio-hydrogen generation from wastewater can become a potential technique for long-term energy production and waste stabilization.

The presence of numerous parameters, including iron and Nickel, in the hydrogenase, a major enzyme in hydrogen generation routes, suggests that these metals have a significant influence on enzyme-catalyzed reaction and microbial performance [3]. Nanotechnology has recently attracted a lot of attention for its potential to improve process efficiency by increasing the important enzymes. An exhaustive literature search reveals that the use of iron and nickel nanoparticles to boost bio-hydrogen production from diverse pure carbon sources such as glucose, sucrose, and other sugars has been widely reported [4-10]. However, it has received very little attention in terms of its relevance for improving bio-hydrogen production from actual industrial wastewater, resulting in grey regions for long-term hydrogen production from complicated wastewaters. Furthermore, a thorough review of the literature revealed that only one study focused on the use of iron nanoparticles to improve bio-hydrogen generation. Malik et al. [10] investigated the influence of iron oxide nanoparticles on bio hydrogen generation from distillery effluent, finding an HY of 1.97 mmol/g COD at a concentration of 50 mg/L, which was significantly lower than the theoretical maximum yield. Furthermore, despite their ability to improve hydrogenase activity, HY, and SHPR, which are favorable to hydrogen generation; Furthermore, a lack of clarity about the mechanism by which nanoparticles aid hydrogen recovery has produced grey regions, making the process difficult to comprehend and optimize.

As a result, a novel attempt has been undertaken in the current work to investigate the application of Fe2O3 and NiO nanoparticles for improving bio-hydrogen generation from complex distillery effluent. This work also addresses the possibility of Fe2O3 + NiO nanoparticles co-addition for improving bio-hydrogen recovery. As a result, the current study has four objectives to improve understanding of bio-hydrogen production from complex distillery wastewater: (1) the effect of NiO nanoparticles on the synthesis of bio-hydrogen from complicated distillery effluent. (2) The effect of Fe2O3 nanoparticles on the synthesis of bio-hydrogen from complicated distillery wastewater. (3) The effect of adding Fe2O3 and NiO nanoparticles to complicated distillery wastewater on bio-hydrogen generation. (4) The mechanism of nanoparticle-mediated hydrogen generation processes regulation.

Fe2O3 and NiO Nanoparticles XRD Analysis

The Scherrer formula was used to calculate the average size of nickel oxide and ferrous oxide nanoparticles, as discussed elsewhere [4], [11], [12]. These nanoparticles generally had typical sizes of 23 nm and 33 nm, respectively. The cubic phase of nickel oxide nanoparticles (JCPDS card file number: 4-0835) (Fig. 1A) and the rhombohedral phase of Fe2O3 nanoparticles (JCPDS card No: 01-084-0307), respectively, were revealed by the XRD patterns of the materials [4], [10], [12], [13]. (Fig. 1B).

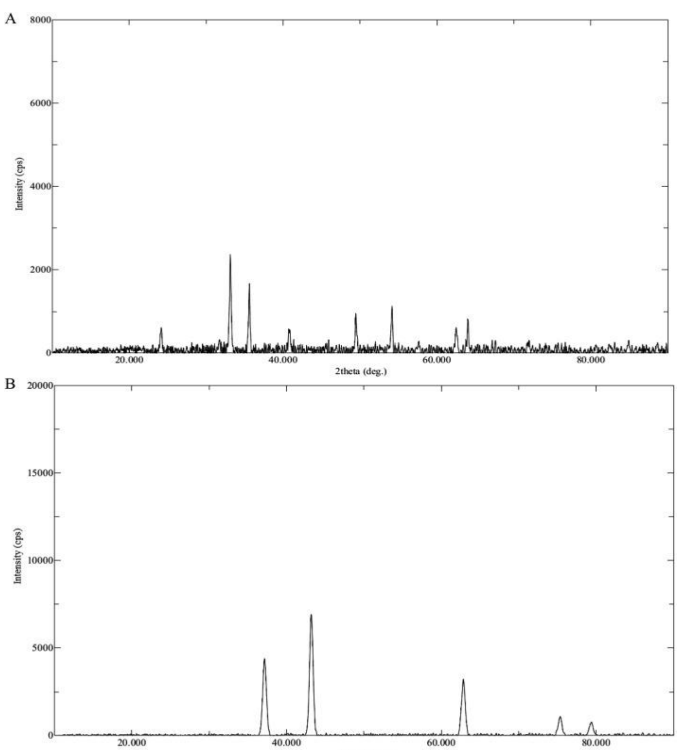

Figure 1: (A) XRD pattern patterns of Nickel nanoparticles (B) XRD patterns of hematite nanoparticles

[Reprinted from Gadhe, A., Sonawane, S. S., & Varma, M. N. (2015). Influence of nickel and hematite nanoparticle powder on the production of biohydrogen from complex distillery wastewater in batch fermentation. International Journal of Hydrogen Energy, 40(34), 10734-10743, with permission from Elsevier]

Mechanism of Hydrogen Production Enhancement by Nickel and Hematite Nanoparticles

Nickel oxide and hematite nanoparticles have proven their usefulness for bio-hydrogen production from complex wastewater. This increase in hydrogen production is due to the increase in the important enzymes for bio-hydrogen production. Figure 2 gives a detailed explanation of the hydrogen production due to the nanoparticle addition.

The bio hydrogen production process can be divided into two categories based on the electron donor available at the terminal: (1) Nicotinamide adenine dinucleotide phosphate (NADPH)-based hydrogen production and (2) ferredoxin-based hydrogen synthesis [14-17]. In NADPH based approach, NADPH is used as an important electron donor by the NADPH: ferredoxin oxidoreductase attached to hydrogenase, and this produce hydrogen. This mechanism is elaborated in Fig. 2 routes 1. And pyruvate: ferredoxin oxidoreductase reduces pyruvate to acetyl coA, resulting in a reduced form of ferredoxin that gives electrons to a hydrogenase for hydrogen generation (Fig. 2 Pathway 2). In comparison to [NiFe] hydrogenase, the [FeFe] hydrogenase enzyme is broadly dispersed throughout the diverse hydrogen-producing microorganisms [18-23]. Furthermore, [FeFe] hydrogenase has been shown to have significantly higher enzyme activity than [NiFe] hydrogenase [24-28]. This results in the increment in ferredoxin oxidoreductase activity due to the simultaneous addition of nickel oxide and hematite nanoparticles. Both the mechanisms are represented in Fig. 2 Pathway 1 and 2. This simultaneous addition of both the nanoparticles increases the synergetic effects of a hydrogenase enzyme, owing to increased expression of route 1, route 2, and 3 (Fig. 2).

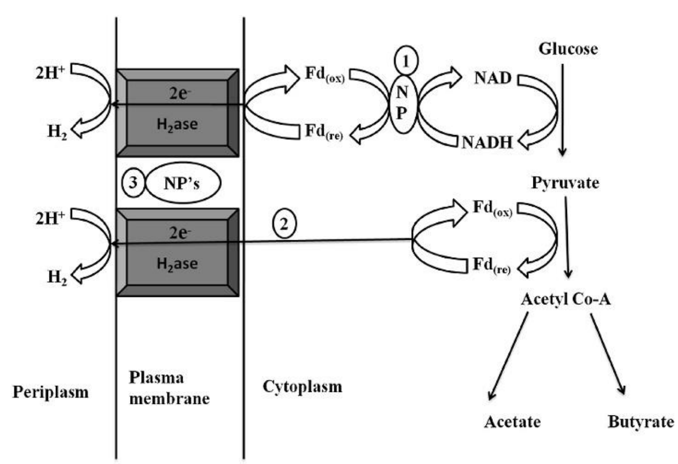

Figure 2: Enhancement mechanism of hydrogen production using nickel and hematite nanoparticles. [Reprinted from Gadhe, A., Sonawane, S. S., & Varma, M. N. (2015).

Influence of nickel and hematite nanoparticle powder on the production of bio hydrogen from complex distillery wastewater in batch fermentation. International Journal of Hydrogen Energy, 40(34), 10734-10743, with permission from Elsevier]

Effect of Hematite Nickel and Hematite Nanoparticles on Hydrogen Production

The effects of iron oxide nanoparticles on hydrogen production from food wastewater were evaluated in the batch hydrogen production potential test, and the results are presented in Fig.3. In a batch hydrogen production potential test, the impacts of iron oxide nanoparticles on hydrogen generation from food wastewater were studied, and the findings are shown in Fig.3. During the batch test, a significant increase in hydrogen output (from 85 to 93 mL/g VS added) was seen at a lower iron oxide content (100 mg/L), reflecting a 9.4% relative improvement over control (Fig. 3A). This demonstrates that the iron oxide nanoparticle has a significant impact on hydrogen generation. From 100 to 400 mg/L of iron oxide nanoparticles, hydrogen generation increases linearly proportional to the iron oxide nanoparticles loading. The complex food wastewater sample with an iron oxide loading of 400 mg/L had the greatest hydrogen output of 133 ml/ g VSadded. It represents a 57 percent increase in hydrogen production. An activation of HPBs due to beneficial changes in nutritional media and higher activity of hydrogenase enzymes under optimum Fe2O3 nanoparticle loading (i.e. 400 mg/L) may be the outcome of an improved electron transfer rate. Furthermore, during the hydrogen generation potential test of food waste with varied loadings of iron oxide nanoparticles, one of the most essential functional parameters to evaluate the microbial reaction is the hydrogen production rate (R). In the current study, the hydrogen production rate increases as the iron oxide nanoparticle concentration increases in every hydrogen production potential tests of food waste samples with iron oxide nanoparticles. (See Figure 3B). Because of an increased hydrogenase activity due to an increased electron adsorption activity mediated by pragmatic surface effect experienced by iron oxide nanoparticles at lower iron oxide concentrations (100 mg/L), a notable increase in the hydrogen production rate from 2.6 to 2.8 mL/h, representing a relative enhancement of 9.2 percent compared to control, was observed. The maximal hydrogen generation rate of roughly 4.2 mL/h, which is a 61 percent increase over the control, is attained with a food waste sample containing 400 mg Fe2O3/L. Because there was no significant increase in the hydrogen production rate when iron oxide nanoparticle concentration was increased from 400 to 500 mg/L, the hydrogen production potential test of food waste with 400 mg Fe2O3/L was the best condition for maximum hydrogen production. The iron oxide nanoparticle boosts NADPH-ferredoxin oxidoreductase activity when used at the right concentration. Increased electron adsorption from NADH and terminal transfer to [FeFe] hydrogenase is the result of this increased activity. The quantum size process causes this transfer, which results in an increase in the ultimate hydrogen production rate during hydrogen production. When comparing a hydrogen production rate of 4.2 mL/h obtained at 400 mg Fe2O3/L to hydrogen production rates of 2.6, 2.8, 3.3, and 3.7 mL/L obtained at 0, 100, 200, and 300 mg Fe2O3/L, it is clear that adding Fe2O3 nanoparticles increase the hydrogen production rates by about 1.1 to 1.6 times during the batch test.

The quantities of both acids are directly related to the hydrogen yield, according to the metabolic profile of a liquid mixture (Fig. 1C). The maximum accumulation values of both acids, 29.29 mM for butyric acid and 63.28 mM for acetic acid, were found to be linked to the maximum hydrogen output of 133 mL/g VS added. For hydrogen generation, butyrate routes are critical. These routes are the primary cause of the increased HBu/HAc ratio, which is in the 1.5-2.1 range (Fig. 3D). Similarly, the increase in hydrogen production due to the addition of nickel nanoparticles is represented in figure 4. An increase in hydrogen production is represented in figure 5.

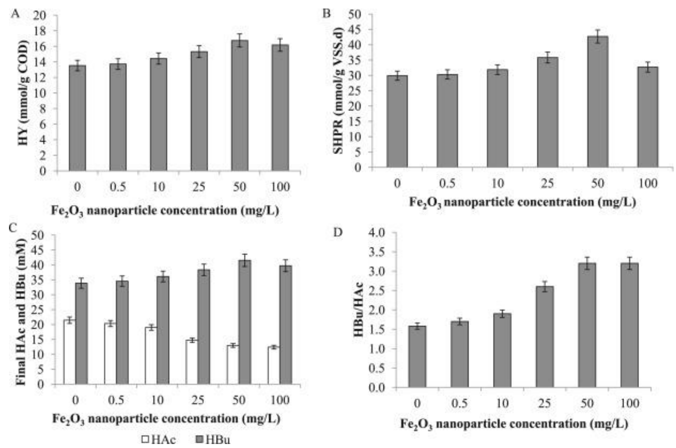

Figure 3: Effect of hematite nanoparticles on the production of hydrogen. (A) Hydrogen yield (mmol/g COD) (B) Hydrogen production rate (C) Final HAc and HBu (mM) (D) Hbu/HAc [Reprinted from Gadhe, A., Sonawane, S. S., & Varma, M. N. (2015). Enhancement effect of hematite and nickel nanoparticles on biohydrogen production from dairy wastewater. International Journal of Hydrogen Energy, 40(13), 4502-4511., with permission from Elsevier]

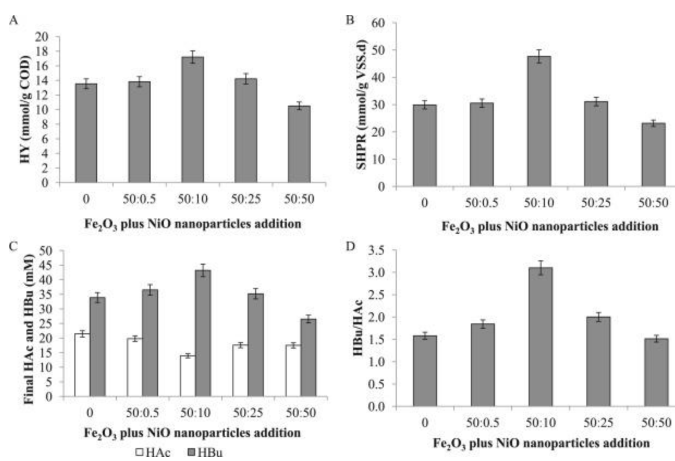

Figure 4: Effect of nickel oxide nanoparticles on the production of hydrogen. (A) Hydrogen yield (mmol/g COD) (B) Hydrogen production rate (C) Final HAc and HBu (mM)

(D) Hbu/HAc. [Reprinted from Gadhe, A., Sonawane, S. S., & Varma, M. N. (2015). Enhancement effect of hematite and nickel nanoparticles on biohydrogen production from dairy wastewater. International Journal of Hydrogen Energy, 40(13), 4502-4511., with permission from Elsevier]

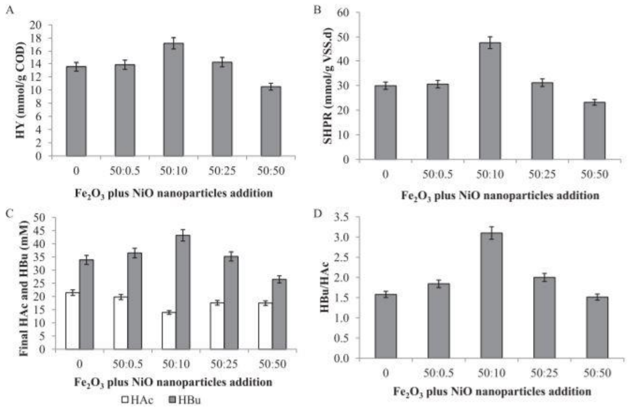

Figure 5: Effect of simultaneous addition of nickel and hematite nanoparticles on the production of hydrogen. (A) Hydrogen yield (mmol/g COD) (B) Hydrogen production rate (C) Final HAc and HBu (mM) (D) Hbu/HAc. [Reprinted from Gadhe, A., Sonawane, S. S., & Varma, M. N. (2015). Enhancement effect of hematite and nickel nanoparticles on biohydrogen production from dairy wastewater. International Journal of Hydrogen Energy, 40(13), 4502-4511., with permission from Elsevier]

Parameters Influencing the Bio-hydrogen Production

The synthesis of bio-hydrogen is influenced by various parameters like the types of feedstock, bioreactor design, feed hydrolysis, retention period, medium pH, pre-treatment procedures, and catalyst are some of the most critical elements. The generation of hydrogen can be considerably boosted by manipulating certain operating conditions. The following is a quick rundown of these factors.

Feed Content Composition

In the creation of bio-hydrogen, the type of substrate employed in the fermentation process is critical. The production of bio-hydrogen varies depending on the properties of food wastes generated from various sources. Carbohydrates, protein, starch, fat, cellulose, lipids, and other substances are common components of food waste [29-32]. Microorganisms thrive in food wastes that include 95 percent volatile substances and 85 percent moisture. The synthesis of bio-hydrogen from these constituents or combined compositions has been thoroughly explored and scaled-up operations [33-38].

Han et al. used an enzymatic hydrolysis method to conduct fermentation processes of high carbohydrate content food waste and discovered that the process can yield 8 mmol h-1L-1 bio-hydrogen with a hydraulic retention duration of 6 h [39-42].

Metal Ions

Metal ions are required for bacteria to create enzymes such as hydrogenases for bio-hydrogen generation via fermentation. These metal ions are vital in the creation of microorganism enzymes [43-48]. Metal ion concentration has been shown to affect the kinetics and enzymatic activities of microorganisms in several investigations [49-53]. Iron is the most important nutrient for bacteria to manufacture enzymes out of all of these metal ions. When combined with sulfur, it also acts as a protein carrier. Excess metal ion concentrations, on the other hand, may stifle bio-hydrogen synthesis. Excess ions influence microorganism development, resulting in greater feed substrate consumption. The inhibitory rate is reduced when a high amount of feed is consumed, which enhances microbe production and metabolic activity [54-58]. The microbial population is affected by iron ions, which affect the fermentation routes. As a result, the Fermentation process is heavily reliant on Fe-ions for bio-hydrogen productivity [27]. When the hydrogenase enzyme is involved in the bio-hydrogen synthesis, nickel (Ni) is another active element that aids in electron transport. [59-63]

Magnesium (Mg) is a cofactor in enzyme synthesis and is found in the cell membranes of most bacteria. It has a considerable impact on the microorganisms' enzymatic fermentation activities.

Mg has been found to serve an activator role in enzymes that induce fermentative action to carry out feed substrate bioconversion metabolism [64-66]. As a result, a significant amount of Mg is required to produce the correct amount of hydrogen [67-70].

II. Conclusion

In this article, we have discussed the recent advances in nanoparticle addition in wastewater streams and the recovery of bio-hydrogen production. Nanoparticles are useful to improve the mass transfer rate. In this article, we have explored the potential of the nickel and hematite particles for the bio-hydrogen production increment. Batch fermentation test is used in the experimentations, and distillery wastewater is used as a feed for the process. During the batch fermentation test, the complicated distillery wastewater served as a substrate. The current study's findings strongly suggest that adding Fe2O3 and NiO NP together improves hydrogen recovery from complicated distillery effluent by 1.2–4.5 orders of magnitude as compared to using only nanoparticles. Ferredoxin, hydrogenase enzymes, oxidoreductase and ferredoxin are important for the increase in hydrogen production.

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