



Further Insight in the Technology of the Production of Hydrogen from Municipal Solid Waste Components at Considerably Low Temperatures

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Abstract

This study investigates further insight into a technology for the production of Hydrogen from a variety of municipal solid waste components at considerably low temperatures. The technology is a low-cost process that is practical, effective, and environmentally clean. The process is based on the catalytic transfer hydrogenation mechanism. The catalytic production of Hydrogen gas from waste stocks including food waste, cardboard and paper mixture, and textile and wood mixture, has been studied at operating temperatures in the range of 180-340°C. Waste piece size was varied in the range of 0.25-2.00 inches. It was found that the cardboard and paper mixture provided the maximum Hydrogen production yield followed by the textile and wood mixture. The optimum operating temperature was found to be around 300°C with an optimum waste piece size of 0.50 inches. The advantages of the current technology include processing the solid waste feedstock within a considerably shorter process time, instead of conventional pyrolysis which requires temperatures that usually exceed 600°C and require a much longer time duration. The process also participates in addressing planetary concerns such as resolving global warming crises by preventing the emission of detrimental Carbon Dioxide by removing the chemically bound oxygen. In addition, the process is highly cost-effective where municipal solid waste and local catalysts can be used.

Disciplinary: Material Engineering

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1 Nomenclature and Symbols

CTH	Catalytic Transfer Hydrogenation
GC	Gas Chromatography
PET	Poly Ethylene Terephthalate
PID	Proportional Integral Derivative
RPM	Revolution Per Minute
TCD	Thermal Conductivity Detector
ZSM	Zeolite Socony Mobil Reference list

2 Introduction

Hydrogen is an energy carrier, which can help to overcome many energy challenges. Because Hydrogen is Carbon-free, it is considered a green fuel. During burning, Hydrogen produces only pure water, steam, and energy. Hydrogen therefore can help in reducing environmentally hazardous emissions in a variety of sectors such as transportation, chemicals industry and Steel production where it is proven hard to reduce emissions using traditional sources of energy. By

Now, demand for Hydrogen has grown more than threefold since 1980. Production of Hydrogen was estimated to be responsible for emissions of around 830 million tons of Carbon Dioxide CO₂ per year, per the technical report from IEA, 2019.

Current conventional methods for producing Hydrogen include converting Hydrocarbons into Hydrogen and Carbon by reaction over catalysts, electrolysis, photochemical, thermo-chemical and gasification processes at high temperatures to decompose biomass and coal into gas streams that include Hydrogen. Due to the high-temperature requirement, these processes have not been fully economically established, and the need for a cost-effective technology for Hydrogen production in industrial amounts still exists.

The implemented method for Hydrogen production in this study employs a chemical reaction mechanism that releases hydrogen and removes the chemically bound oxygen present in the feed waste to prevent the formation of the greenhouse gas Carbon Dioxide CO₂. While Nitrogen and Sulfur products have in the past posed disposal issues, the current method binds these elements in solid format, such as Ammonium Hydroxide and Sodium Sulfide. The method follows the general concept of Catalytic Transfer Hydrogenation CTH where the solid waste is heated in the presence of a catalyst and a Hydrogen donor in a basic medium at relatively low temperatures. This leads to hydrogenating and then dehydrogenating part of the solid waste to finally produce Hydrogen and Carbon, per Rogers, 2006.

The current method can contribute to the innovation and development of environmental technologies and industries. It contributes to transferring, localizing, and developing high-quality environmental methods to tackle national environmental issues and implementing next-generation waste management practices. In general, the materials that can not be recycled can be passed through for Hydrogen production. In addition, low-temperature catalytic methods may

increase the calorific value of the processed waste material, yielding a value-added Carbonized solid source of energy, per Swiechowski, et al, 2020.

The current method converts municipal solid wastes, which are usually a source of air and water pollution, to a useful source of energy. The world generates 2.01 billion tons of waste annually, per Kaza et al, 2020. Studies show that Saudi Arabia produces around 15 million tons of municipal solid waste each year with an average daily rate per person of 1.4 kg. Figure 1 shows the major ingredients of Saudi Arabian municipal solid wastes, per Miandad, et al, 2016.

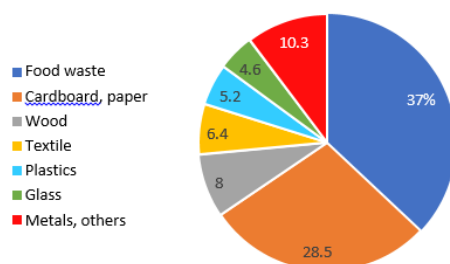


Figure 1: Major ingredients of the 15 million tons of municipal solid waste produced in Saudi Arabia each year, per Miandad, et al, 2016.

3 Literature Review

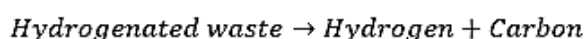
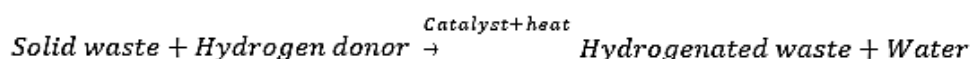
Conventional pyrolysis processes, which require high temperatures, are usually accompanied by the production of high levels of environmentally hazardous gases, to El-Sinawi, et al, 2012. A. El-Sinawi, et al, 2016 conducted a series of laboratory-scale experiments to investigate the production of Hydrogen at low temperature from waste rubber moistened with oil. The process was optimized, and process temperature was varied in the range 150°-350°C, air flow rate in the range 200-300 cc/min, and Zeolite Socony Mobil –5 (ZSM-5) catalyst was used. The process was found to be effective for producing Hydrogen and it was found that in general the higher the temperature and flow rate the more CO and H₂ produced. Retention of essentially all Carbon from rubber could be directly converted to Carbon Monoxide as a Syngas product. It was reported that the recovered residue can be used for additional Hydrogen production in the Water–gas–shift reaction, per El-Sinawi, 2012. In another study,

A. El-Sinawi experimentally investigated the production of Hydrogen from Poly Ethylene Terephthalate (PET) using CT 434 ZSM-5 catalyst. Thermal Conductivity Detector TCD within a Gas Chromatography GC was used to detect the amount of Hydrogen produced at different process temperatures. The spectrum of pure hydrogen, as well as the spectra of process-evolved gases at 250°C, 280°C, 310°C and 340°C, were obtained and compared to validate the presence of Hydrogen. The maximum Hydrogen percentage obtained was 20.11% at a process temperature of only 250°C. It was reported that the process could be more developed to produce Hydrogen from solid wastes, per El-Sinawi, 2016.

N. Miskolczi et al. studied the thermo-catalytic pyrolysis of plastic waste from end-of-life vehicles. Two-step heated batch reactors provided with catalysts were used at temperatures of 425°C and 485°C. The products were further modified in the second step using a catalyst at 380°C. It was found that catalysts in the first reactor significantly affected the product yield and decreased

the pyrolysis time. Catalyst in the second reactor was found to increase the Hydrogen yield in gases but due to coke formation, the catalyst surface was fully covered by coke depositions during the process, per Miskolczi, et al, 2017.

The current method applies the concept of Catalytic Transfer Hydrogenation, per Rogers, 2016, where a small amount of Hydrogen donor is added to the waste in the first step. In a second simultaneous step, the waste releases Hydrogen in relatively very short times, per Rogers, 2016. The focus here is on the use of a wide range of feedstock locally obtained, in addition to the use of locally available materials such as Palm oil fatty acids and Asphalt as will be shown in the next section. This process is also safer than the conventional pyrolysis methods in terms that there is no need to handle the hazardous molecular Hydrogen, which has high infusibility due to its low molecular weight and can easily ignite. Instead, a Hydrogen donor is employed to supply the needed Hydrogen to reduce the organic compounds. The use of Hydrogen donors was proved to have real and potential advantages especially since it eliminates the need of using pressure vessels, Johnstone, et al, 1985. The feedstock is best to be rewetted by the Hydrogen donor which may improve both Hydrogen production rate and feeding rate in continuous production. In general, the process can be summarized as follows, per Rogers, 2006:



However, a proper Hydrogen donor needs to be selected as it can affect the selective adsorption process onto the surface of the catalyst. Experimental investigation can be followed to help in selecting the donor based on the reaction rate and in many reactions with Hydrogen donors; it may not be straightforward to determine the Hydrogen transfer mechanism, per Johnstone, et al, 1985. However, in general, compounds that contain Hydrogen atoms bonded to groups or elements with similar electronegativity to that of Hydrogen are considered the best Hydrogen donors, per Rogers, 2006.

A great effort has been made by researchers to find proper inexpensive catalysts suitable for the CTH process. It was early known that some metal-based catalysts such as those of Rh, Ru and Pd metals are among the most active catalysts. In early developments of the CTH process, T. Tatsumi et al. succeeded in preparing a less expensive active catalyst and were the first to observe the capability of Molybdenum Mo-based catalysts in hydrogenation processes. They also experimentally examined several compounds to determine their effectiveness as a Hydrogen donor and concluded that the transfer hydrogenation reactions seem to be selective, per Tatsumi, et al, 1977. Metal-based catalyst deactivation is also common in hydrogenation reactions. Sulfur compounds, Carbon Monoxide, Oxygen, and Hydrogen Halides were reported to act as catalyst poisons, per Sasson and Blum, 1975. Once a catalyst is covered with such poisons, the Hydrogen donor and the acceptor cannot react. The catalyst was even found to become deactivated due to

being locked up in a resulting polymer if the hydrogenation process induces a polymerization reaction, per Dobson et al, 1979. Therefore, one difficulty that is associated with some catalysts of this type is the difficulty of their recovery from reaction products, per Ohkubo, et al, 1979. Another difficulty is the need for careful preparation methods of such catalysts in relation to their particle size, surface area, and pore structure, per Anderson, 1975..

The detailed objectives of the current study are:

- 1) Applying the concept of Catalytic Transfer Hydrogenation CTH mechanism to produce Hydrogen from municipal solid wastes.
- 2) Determining the optimum operating conditions of minimum temperature, and solid piece size required to rapidly convert the solid waste totally to Carbon and Hydrogen.
- 3) Determining Hydrogen yields using a variety of waste feedstock, to identify the waste that provides the highest yield of Hydrogen.

4 Experimental Setup and Methodology

The main components of the experimental setup used in this study are shown in Figure 2. The setup consists of the following main components:

- 1) A reaction vessel where the contents are continuously mixed using a motorized mixer rotating at 30 RPM.
- 2) Heating jacket surrounds the reaction vessel. The jacket set point is controlled based on the gas temperature inside the reaction vessel, which is measured by a thermocouple. The thermocouple is connected to the heating jacket via a Proportional Integral Derivative PID controller such that the reaction temperature is controlled online. The thermocouple tip is located nearly at the center point of the reaction vessel.
- 3) The reaction-produced gases flow to a collecting vessel, from where they are passed through the cotton filter to another upper vessel where samples are taken using tight gas syringes. All vessels are provided with purge valves.
- 4) The setup includes several other equipments such as a feed waste drying furnace, waste automatic shredder, and Nitrogen cylinders to provide an inert vessel environment, and Gas Chromatography GC with Thermal Conductivity Detector TCD for gas sample analysis.

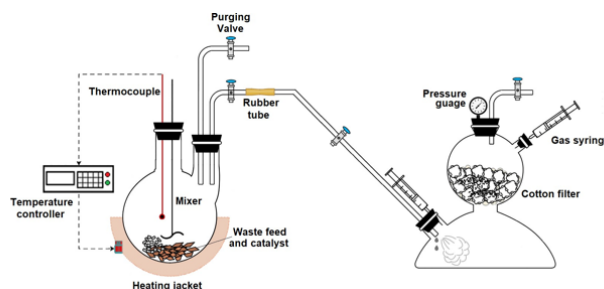


Figure 2: Experimental setup main components.

To prove the process concept, the following methodology was employed to collect the experimental data, for each test, two trials were carried out and the average of the results was taken:

1) The GC system was initially calibrated using pure Hydrogen and the spectrum of pure Hydrogen was obtained for reference. This is shown in detail in El-Sinawi, 2016.

2) Municipal solid waste groups of food waste, cardboard and paper mixture, and textile and wood mixture were collected. Primary samples were dried in an oven at 70°C for 24 hours. This time duration was found long enough such that the sample masses started to not change over 1-hour periods in the furnace.

3) Primary samples were reduced in size using an automatic shredder to around 0.25-2.00- inch squared pieces. The standard ASTM sieves were used to separate the samples of the required size. For example, samples of sizes around 0.25 inches were collected between sieves with mesh openings of 0.265 inches and 0.250 inches. Samples of sizes around 0.5 inches were collected between sieves with mesh openings of 0.530 inches and 0.438 inches. Finally, samples of sizes around 2.0 inches were collected between sieves with mesh openings of 2.120 inches and 2.000 inches.

4) A Hydrogen donor (asphalt) was added in a concentration of 7% of the weight of the feedstock planned to be placed in the reactor (Each batch was operated with 50 g of feed waste). A base (Calcium Hydroxide) was also added in a concentration of 15% of the Hydrogen donor. Composition analysis of the used Asphalt hydrocarbon showed that it contains about 76 wt.% Carbon, 13 wt.% Hydrogen, 4 wt.% Sulfur, and the rest consist of Nitrogen, Oxygen, and traces of other elements.

5) The vessel contents were heated to temperatures in the range of 180-340°C with continuous mixing.

6) A catalyst composed of a mixture of 50 wt.% Isopropanol Alcohol Isopropanol and 50 wt.% Palm oil fatty acid has been used. This fatty acid has been selected due to its availability in the country and in the gulf region. Alcohol can also act as a Hydrogen donor, per Tabanelli, et al, 2020. The catalyst was added in a concentration of 5% by weight of the feedstock. The shredded waste was then added to the bottom of the heated vessel with mixing continued.

7) Inert gas Nitrogen was added to the vessel. This is advantageous to replace Oxygen at the headspace during Hydrogen production.

8) Heating of the contents was continued with mixing. At this step, active Hydrogen provided by Hydrogen donor reacts with Hydroxyl groups present in Oxygenated feed stocks to produce Water, which is collected in the second vessel.

9) Heating was continued to around 340°C to decompose the Carbon structure and release Hydrogen into the gas stream. The decomposition of feedstocks is completed in less than 30 minutes.

10) Some of the Hydrogen donors could be cracked to produce lower molecular weight Hydrocarbons which could be collected in the second vessel. This collected Hydrogen donor might be reused to process other waste materials.

11) Data was collected and analyzed to determine Hydrogen yield from each feedstock under consideration. The weight of the second vessel of the experimental setup was measured before and after each test and the difference in weight was taken. This difference represents the mass of the gases produced. The GC system was also used to measure the percentage of Hydrogen in this stream. Dividing the obtained Hydrogen amount by the initial feedstock gives the Hydrogen yield.

12) The process parameters were optimized by finding the optimum waste piece size, and process temperature. Based on the gas analysis device and the deviation between the repeated tests, the error in measuring the Hydrogen yield is estimated to be around 7%.

Table 1 shows the materials of municipal waste groups used in this study.

Table 1: Materials of municipal wastes used in this study.

Waste group	Description
Food waste	47% Rice 12% Chicken, meat 41% Peel, fruit, vegetables
Cardboard and paper (mixture)	50% Cardboard boxes 50% office paper
Textile and wood (mixture)	50% Cotton shirts and fabrics 25% Wood 25% Tree fronds

5 Results & Discussion

5.1 Effect of Processing Temperature

The processing temperature was varied to determine the minimum temperature required to convert the waste rapidly and totally to Carbon and Hydrogen with the maximum possible amount of Hydrogen produced. The temperature varied in the range of 180-340°C.

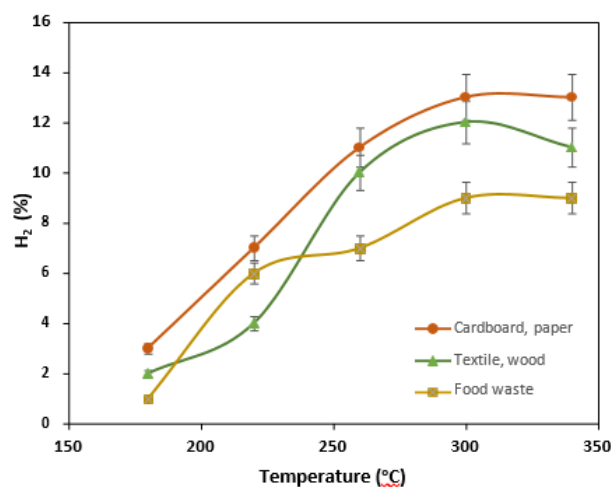


Figure 3: Effect of processing temperature on amount of Hydrogen production.

The amount of Hydrogen produced versus the operating temperature is shown in Figure 3. The figure shows that in general the amount of Hydrogen increases by increasing the temperature

within the range studied. This result agrees with several studies in literature even for other types of catalysts where in general it was found that the production rate increases almost linearly by temperature with a decrease at the higher temperatures due to side or reverse reactions, per Imai, et al, 1974. However, there is not much change in Hydrogen production beyond the temperature of 300°C. Therefore, this temperature is selected as the optimum process temperature. It was observed that the reaction was completed in less than 30 minutes, which is a very short time in comparison with the conventional decomposition methods, which usually takes hours of operation, per El-Sinawi, 2012.

5.2 Effect of Waste Material

Different types of solid waste materials can be used as feedstock in the current process. This can solve one of the major solid waste problems and turn the waste into a good source of energy. The amount of Hydrogen produced from the various feed wastes used in this study was determined in order to identify the most efficient waste. In general, the order of waste selection for use was found to be in order: Cardboard and paper mixture, Textile and wood mixture, and Food waste. This result is shown in Figure 2 above for the range of operating temperatures. As an example, the results at the operating temperature of 300°C are shown in Figure 4.

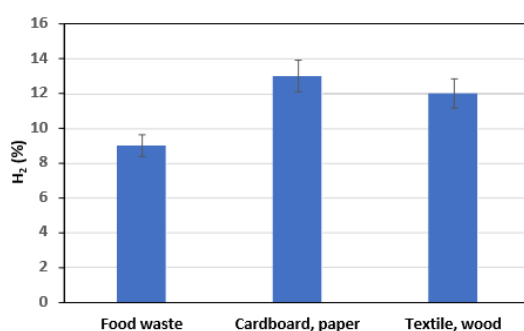


Figure 4: Effect of waste type on amount of Hydrogen produced.

5.3 Effect of Shredded Waste Piece Size

The size of the shredded feedstock pieces plays an important role in controlling the processing time and temperature, so it is important to determine the optimum size required for the waste to be rapidly converted to the desired products. It is expected that in general as the piece size decreases, the contact surface increases, and thus a Hydrogen higher production is obtained. The way sample size was determined as described in the Experimental Setup and Methodology Section above. The results are shown in Figure 5. The figure shows that as the piece size got smaller, the production rate increased. However, there was not much change in the production rate when the piece size was reduced from 0.50 to 0.25 inches. Therefore, to save shredding energy and time, a size of 0.50 inches is considered the optimum piece size.

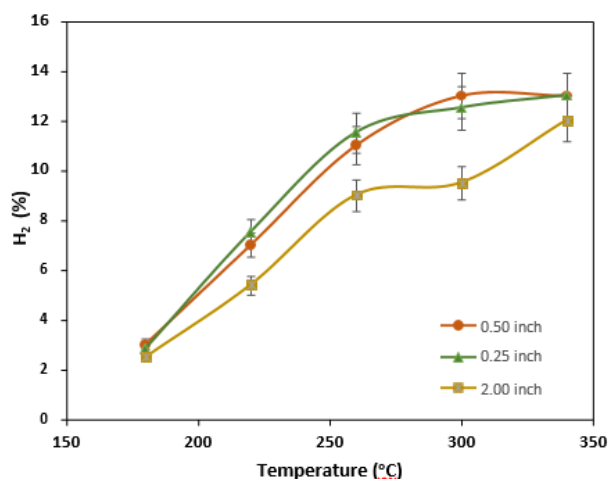


Figure 5: Effect of shredded waste piece size on amount of Hydrogen produced.

6 Conclusion

The current proposed technology of Hydrogen production from municipal solid waste materials at considerably low temperatures has been found to be an effective way that requires minimum energy input. The optimum conditions were examined by varying process temperature, waste type, and waste piece size. The feasibility of the results is promising. The process is flameless; there is no combustion but a catalytic decomposition at low temperatures. In addition, there is no Carbon Dioxide CO₂ formation or emission, which complies with the global warming precautions to protect the environment. The feedstock consists of municipal solid waste. Food waste, cardboard, paper, textile, and wood, which are examples of the challenges that face researchers and environmental specialists, can be included in the feedstock turning them into a good source of Carbon and Hydrogen. The optimum operating temperature was found to be 300°C and the optimum waste piece size was found to be 0.50 inches. One recommendation for future research is the use of Saudi Arabia's natural Zeolite in the process as it is available in abundance in the country and has the potential to be used as a catalyst in different waste-to-energy fields after some treatment, per Miandad, et al, 2016. The use of the country's natural catalysts will make the process highly economical due to local abundance.

7 Availability of Data and Material

Data can be made available by contacting the corresponding author.

8 Acknowledgment

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