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Research Article

Hydrogen Production Via Waste Pyrolysis: A Review of A Study

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Abstract

Hydrogen has great potential for future energy prospects, especially in the potential low-carbon energy system, known as hydrogen economy. It has the highest energy content among all existing fuels [1]. Additionally, as an efficient and clean energy carrier, with only water being released as a byproduct in its conversion into energy. While hydrogen can be derived from various technologies, including both non-renewable (such as fossil fuels) and renewable sources (like biomass) [2], the predominant method still involves fossil fuels, with limited renewable applications due to technological and economic challenges [3]. This study presents a patented model designed by Environmental Power International Ltd (EPi R&D, UK) to produce hydrogen-rich gas from waste while minimising carbon emissions. Model integrates three main sub-systems: High Temperature Pyrolyser, Gas Refinery Unit and Hydrogen Conversion Unit. EPi pyrolysis unit converts the feedstock into syngas and carbon char, and then syngas is refined into methane-rich gas, and ultimately produces high-purity hydrogen and carbon black via thermal plasma electrolysis. Lab scale trials conducted on waste mixtures (Solid Recovered Fuel (SRF) and waste plastic) demonstrated that the model's capability to convert waste into hydrogen with purity exceeding 90%. The design provides great potential for Carbon Capture through its by-products including solid Carbon Black and solid Carbon Char. Carbon Life Cycle Assessment demonstrates that the system leads to net negative emissions. This integrated pyrolysis solution presents a promising avenue for sustainable hydrogen production from waste.

Keywords: Hydrogen, waste, pyrolysis, carbon capture

Atıktan Piroliz ile Hidrojen Üretimi

Özet

Hidrojen, düşük karbonlu enerji sistemleri açısından büyük bir enerji potansiyeline sahiptir. Hidrojen, mevcut tüm yakıtlar arasında en yüksek enerji içeriğine sahiptir [1]. Ayrıca, su dışında herhangi bir yan ürün olmadan enerjiye dönüşümünde etkili ve temiz bir enerji taşıyıcısı olarak tanımlanabilir. Hidrojen çeşitli teknolojilerle elde edilebilir; bunlar arasında yenilenebilir olmayan kaynaklar (fosil yakıtlar gibi) ve yenilenebilir kaynaklar (biyokütle gibi) yer alır [2]. Ancak, mevcut yöntem genellikle fosil yakıtlardan elde edilmekte olup, teknolojik ve ekonomik zorluklar nedeniyle yenilenebilir kaynakların kullanımı sınırlıdır [3]. Bu çalışma, Environmental Power International Ltd., (EPİ R&D, İngiltere) tarafından geliştirilen bir patente sahip modeli sunmaktadır. Bu model, atıktan hidrojence zengin gaz üretirken karbon emisyonlarını minimize etmeyi amaçlamaktadır. Model, üç ana alt sistem olan Yüksek Sıcaklık Pirolizör, Gaz Rafineri Ünitesi ve Hidrojen Dönüşüm Ünitesi'ni entegre eder. EPi piroliz ünitesi, beslenen malzemeyi sentetik gaz ve karbon kömürüne dönüştürür; ardından sentetik gaz metanca zengin gaz haline getirilir ve nihayetinde termal plazma elektrolizi ile yüksek saflıkta hidrojen ve karbon siyahı üretilir. ATY (Atıktan türetilmiş yakıt) ve atık plastik karışımları üzerinde laboratuvar ölçeğinde yapılan denemeler modelin atığı, %90'dan fazla saflıkta hidrojene dönüştürme kapasitesi olduğunu göstermiştir. Tasarım, katı karbon siyahı ve karbon kömürü dâhil yan ürünler aracılığıyla karbon yakalama için büyük potansiyel sunar. Karbon Yaşam Döngüsü Değerlendirmesi, sistemin net negatif emisyonlar elde ettiğini göstermektedir. Bu entegre piroliz çözümü, atıktan sürdürülebilir hidrojen üretimi için umut verici bir yol sunmaktadır.

Anahtar Kelimeler: Hidrojen, atık, piroliz, karbon yakalama

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1. Introduction

Hydrogen holds immense promise for the future of energy, particularly within the framework of the low-carbon energy system known as the hydrogen economy. As the most abundant element in the universe, hydrogen boasts the highest energy content of all fuels. Remarkably, 1 kg of hydrogen equates to the energy found in 2.1 kg of natural gas or 2.8 kg of oil, making it not only the lightest element but also a powerhouse of potential [1]. Hydrogen can be produced through several different methods including non-renewable fossil fuels or renewable sources such as biomass, wind, and solar energy [2]. In this process, it is essential to separate hydrogen from CO_2 and other gases in the resulting gas mixture to obtain pure hydrogen [4]. Hydrogen is regarded as a clean and non-toxic energy source because theoretically, the only by-product released is water. However, many methods of hydrogen production are not environmentally friendly. Factors such as raw materials, energy inputs, and waste emissions throughout the entire lifecycle of hydrogen will determine how clean its production actually is [5][6][7].

Globally, the demand for hydrogen is approximately 70 million metric tons annually, with a significant portion of this demand primarily met by fossil fuels [8]. Approximately 48% of global hydrogen production comes from natural gas, 30% from petroleum, 18% from coal, and 4% from electrolysis [2] [9]. Unfortunately, a significant amount of hydrogen is still produced from fossil fuels rather than renewable energy sources. There are some applications involving biomass for hydrogen production, but they face various technological and economic challenges [3].

The most prevalent technologies used in hydrogen production include steam methane reforming (48%), partial oxidation (30%), coal gasification/pyrolysis (18%), and electrolysis (4%) [10].

Pyrolysis is defined as the direct thermal decomposition of materials in an oxygen-free environment [11]. During pyrolysis, the large molecules in biomass break down into smaller molecules, yielding three primary products [12].

Biomass + Heat → Gas + Liquid + Solid

Liquid products: tar, heavy hydrocarbons, and water

Solid products: charcoal or carbon

Gaseous products: H_2, CH_4, CO_2, H_2O, CO, C_2H_2, C_2H_4, C_2H_6, C_6H_6, etc.

The distribution of solid, liquid, and gaseous products resulting from pyrolysis depends on the material being pyrolysed and the operational conditions. To obtain the desired product, it is crucial to understand the chemical and physical properties of the material, as this knowledge helps determine the optimal operational conditions. The factors influencing pyrolysis can be classified into two main categories: the characteristics of the raw material, such as its type, particle size, and any pre-processing it undergoes, and the reaction (operational) conditions, which include the addition of catalysts, pyrolysis temperature, pressure, heating rate, and retention time [13].

EPi in collaboration with its research and industrial partners has undertaken a pioneering initiative to explore hydrogen production through waste and biomass pyrolysis while achieving net negative CO_2 emissions. This ambitious endeavour, known as the Pure Pyrolysis Refined project, is supported by the Department for Energy Security and Net Zero. The objectives of the project include enhancing the efficiency of hydrogen technologies and developing advanced syngas treatment and upgrading technologies.

Several configurations can enable hydrogen production from waste and biomass. This project utilizes EPi's high-temperature pyrolyser, followed by a five-stage gas-conditioning unit, to produce high-quality fuel gas. EPi's patented pyrolysis process, tested extensively on various waste and biomass streams, showcases its flexibility and efficiency in converting feedstocks such as SRF, waste plastics, sewage sludge, and woodchips into syngas and high-purity hydrogen. Notably, the process generates no airborne emissions and captures carbon in valuable forms like carbon char, supporting carbon capture strategies. The syngas from the pyrolyser is converted into a methane-rich gas in the Gas Refinery Unit, and subsequently supplied to the Hydrogen Rich Gas Conversion Unit, where the methane-rich gas is split into its base components: hydrogen and carbon. This high-speed process operates without emissions in the absence of oxygen, capturing carbon as carbon black.

The final design configuration was determined as an outcome of multiple workshops with academic and industry collaborators, 15 simulation studies using gPROMS™, and operations on a test rig at Imperial College. Research and discussions initially focused on hydrogen stripping technologies and production processes to identify complementary technologies for integration with the EPi pyrolyser. The final criteria for selecting the best technologies and configurations included:

- Maximising hydrogen production
- Minimising CO_2 emissions and maximising carbon capture
- Accommodating a wide range of biomass and waste streams
- Maximising conversion of waste to useful products
- Minimising capital and operational costs

All configurations were modelled using gPROMS™ based on gas results from the pyrolysis test rig at Imperial College. The pilot plant operated on various feedstocks with biogenic content ranging from 38% to 84%. Both conventional and novel approaches were simulated using trial data, literature reviews, and performance data from real-world systems.

This article highlights the innovative strides made by EPi and its collaborators, showcasing how their collaborative efforts are advancing sustainable energy production through cutting-edge pyrolysis technology.

2. Material and Method

Extensive detailed literature reviews and scientific research for potential hydrogen production and carbon removal technologies have been undertaken during the project. In addition to that, a series of engineering studies to find the best and optimal design configuration, through assessment of energy generation potential, yields and production requirements have been performed. Since the award of contract, the feasibility assessment has been carried out in four distinctive but correlated steps.

During the first step, the aim was to identify technologies, which could convert and/or strip the pyrolytic syngas into hydrogen and other gases. There are several hydrogen separation/purification technologies each with its own advantages, disadvantages and applications [14] [15] [16] [17] [18] [19] [20] [21]. Technologies reviewed included GTL FT (Gas to Liquid Fischer-Tropsch), water-gas shift reactors, pressure swing adsorption, membranes, chemical absorption, hot potassium carbonate, metal hydrate, catalytic methanation reactors, plasma torch, electrolysis, cryogenic distillation.

Throughout the second step of the project, further literature review and discussions with experts in the field were conducted to identify the factors such as feedstock properties (i.e. composition, moisture content, inert content, particle size) and operational conditions (i.e. temperature, heating rate, retention time), which influenced gas composition and product yield during pyrolysis [13]. The thermochemical behaviour of waste is primarily determined by its chemical composition and structure. The composition of raw materials is a critical parameter for product distribution and, consequently, system efficiency. For instance, products derived from the pyrolysis of plastics are mainly hydrocarbons, whereas those obtained from the pyrolysis of biomass (such as wood, coconut shells, paper, etc.) are highly [13]. Samples rich in biomass yield higher carbon char and lower gaseous products, which is attributed to the high content of cellulose-based materials [22] [23] [24]. Therefore, to observe the impact of feedstock on pyrolysis, two distinct mixtures (SRF + Willow Wood, WW and SRF + waste high-density polyethylene, HDPE) with biogenic content ranging from 38% to 84% were utilized in the pilot-scale trials.

Temperature is one of the most important factors affecting the degradation of the organic content during pyrolysis reactions and it is directly related to product distribution (i.e. gas and char yields) and product properties (i.e. gas composition, carbon content in char etc.). Gas yield increases significantly with rising temperature. Conversely, liquid yield decreases substantially as temperature rises due to secondary cracking reactions that favour gas production. An increase in temperature also results in a reduction in char yield [25] [26]. This study was carried out to ascertain the optimal operating temperature conditions for the pyrolysis unit, which could deliver a gas composition most suited to effective operation in the downstream processes in order to achieve the highest yields of hydrogen whilst reducing CO_2 emissions.

The findings of the literature review and discussions carried out in the first two steps were combined in the third step of the project to determine the parameters to be trialled during the trials conducted at Imperial College and to determine the technologies to be integrated to EPi's pyrolysis system to produce the methane and hydrogen rich gas from pyrolysis gas. The main aim of the laboratory trials is to apply outputs from two previous steps to evaluate the contents of the gas, which is predicted to be produced by the EPi pyrolyser. This is achieved by using a pilot plant, which is specifically created to emulate certain aspects of the EPi pyrolyser.

In the laboratory-scale experiments, pyrolysis trials were conducted using SRF, by mixing them with waste HDPE and WW to obtain the necessary material blend to meet the target of producing methane-rich gas. Figure1displays sample photographs of SRF, HDPE and WW. In the pilot tests, SRF was obtained from a London-based company specializing in the residual waste left after the separation of household or industrial waste. The composition of this waste includes mixed paper, cardboard, wood, and other cellulosic materials, with a biogenic carbon content exceeding 85%. These raw materials were mixed and used in predetermined ratios (70% SRF:30% WW and 30% SRF:70% HDPE) to achieve the minimum 25% biogenic content in the feedstock according to the cone and quartering method as presented in Figure 2.

Drawing from the literature, two distinct temperature regimes, low and high, have been selected for experimentation to evaluate the impact of temperature on pyrolysis products and gas composition.

The plan has been developed for laboratory tests to analyse the inputs and outputs (pyrolysis gas and carbon char) generated during the pyrolysis experiment. These tests will determine moisture content, inert material content, calorific value, elemental composition (C, H, S, N, O), and gas composition (H_2, CO, CO_2, CH_4, etc.). Additionally, mass and energy balances have been calculated to determine the product distribution and the energy content of the resulting products.

Figure 1. Photos of SRF (left), waste HDPE (middle), and Willow Wood (right) samples

Figure 2. Photos of mixing samples by cone and quartering

The fourth stage involved laboratory trials, to assess the impacts of the factors identified in the first three steps and to collect the required data for the simulation trials. Mass and energy balance data obtained from the experiments were simulated on gPROMS™ Process 2.2 advanced mathematical modelling package. This a high-fidelity model constructed for processes which convert syngas to hydrogen. The model was designed to maximise hydrogen production and carbon capture while minimising the energy requirements. It provides the final design of plant and performs the necessary calculations to identify the product yield and composition at every individual process step, whilst demonstrating optimum operational conditions. This high-fidelity model predicts the conversion, selectivity, reactor types and dimensions, and operational conditions of each sub system. Fifteen different configurations have been designed to determine the best possible configuration that can produce highest yields of hydrogen while minimising the carbon emissions.

After finalising of the high-fidelity model, the data utilised in modelling the reactors in Gas Refinery Unit has been validated through experimental testing carried out at another lab-scale facility. The findings of the test results were used to confirm the vessel dimensions, flow rates and performance of the catalyst obtained from simulation and modelling. The rationale behind the necessity for experimental validation of reactors is to confirm the performance of catalysts and flow rates determined by the high-fidelity model to ensure that the data used is still applicable to the system designed on the specific gas content received from pilot pyrolysis plant. The test rig is designed to reproduce the exact operational conditions that the actual scale plant will be operated at.

3. Results and Discussion

Simulation studies and pilot plant trials added significant value to the project, to define, lay out and mature the concept. Various exercises were performed combined with a series of simulation studies to reduce the complexity of the system, resulting in the creation of the final version of the design, that meets the project requirements effectively.

The model was built upon primary data collected from lab scale pyrolysis plant and secondary data from industry. The lab scale trials conducted demonstrated that up to 80% of the feedstock (25% biomass content by CV) can be converted into synthetic gas. Gas yield determined during these trials was used as input gas data to model the gas flow throughout the downstream processes. This data is the first point of entry to the model, which determines the amount of gas to be supplied to the first set of reactors. The data gathered from the trials has been incorporated into decision matrix as presented in Table 1. Evaluations focused on achieving the following objectives:

- Enhancing methane and hydrogen content in the syngas
- Lowering CO and CO_2 levels in the syngas
- Optimizing pyrolysis operations to promote gas formation by increasing the conversion of waste into lighter gaseous products

Table 1. Decision matrix for the identification of the region of the optimal conditions to reach the target

DECISION MATRIX						
			Option-1	Option-2	Option-3	Option-4
		Target	SRF+WW at high T	SRF+WW at low T	SRF+HDPE at low T	SRF+HDPE at high T
Criteria-1	Gas Yield, %	Max.	74.00%	62.00%	72.00%	83.00%
Criteria-2	CH4 yield, %	Max.	15.90%	18.60%	27.87%	34.20%
Criteria-3	CO2 yield, %	Min.	13.40%	19.20%	9.09%	1.20%
Criteria-4	CO yield, %	Min.	26.70%	27.00%	12.57%	3.70%
Criteria-5	H2 Yield, %	Max.	33.40%	27.40%	19.59%	42.30%
Other Hydrocarbos Yield, %		-	10.60%	7.80%	30.89%	18.60%

The SRF+HDPE mixed feedstock at elevated temperatures (Option-4 in Figure 3) yielded the highest production of H_2 and CH_4, along with minimized emissions of CO and CO_2, and maximized overall gaseous production as demonstrated in Table 1 and Figure 3.

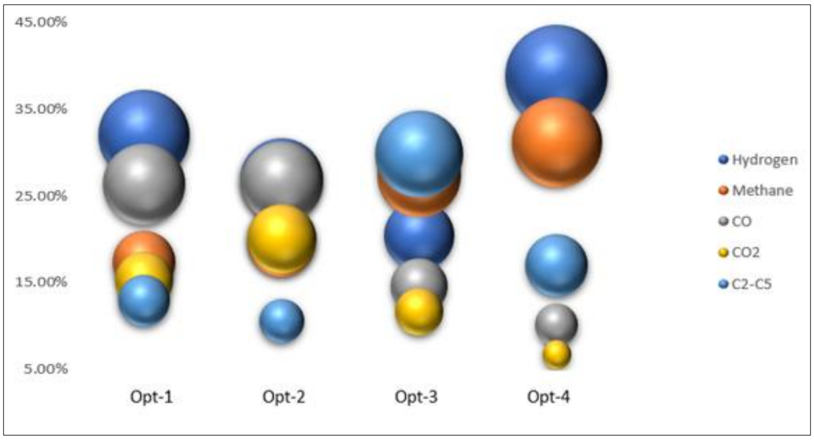

Figure 3. Decision matrix data; Gas composition at different temperature with different feedstock types

Gas composition is another important parameter that directly effects the quality, content and volume of the output products including hydrogen. Input gas composition, obtained from test rigs was provided to the computerised model to calculate the amount of Hydrogen and C Black produced from the configuration.

The gas quantity and composition data obtained from the pilot-scale trials have been utilized in the computerized model to identify the optimal process design for hydrogen production from waste pyrolysis. The proposed configuration for the Pure Pyrolysis Refined project is designed to operate on syngas produced from various waste streams in EPi's high temperature pure pyrolysis system. Simulation studies concluded that EPi model entails the following complementary technologies which are integrated to the high temperature pyrolysis unit to obtain the highest H_2 yield with the minimum CO_2 emissions: The plant design modelled integrates three main sub-systems as demonstrated in Figure 4:

- High temperature pyrolyser
- Gas refinery unit which includes conversion of syngas into methane rich gas
- Hydrogen rich gas conversion unit by thermal plasma electrolysis technology

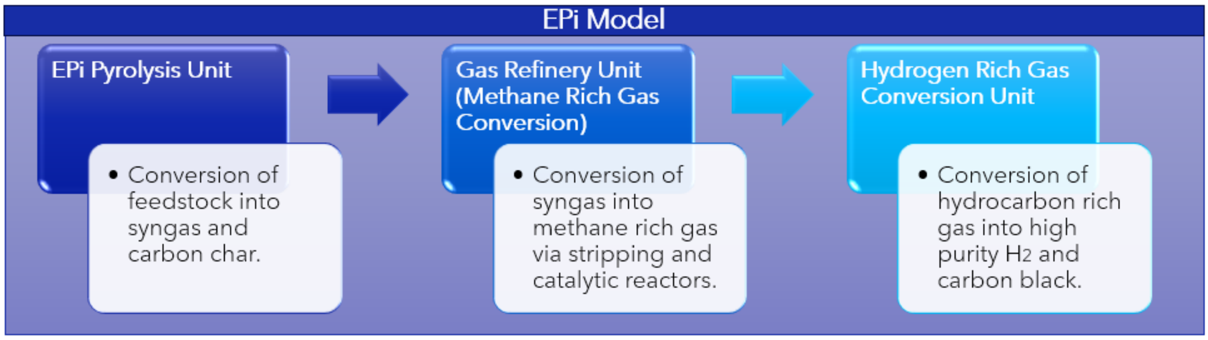

Figure 4. EPi model for hydrogen production via waste pyrolysis

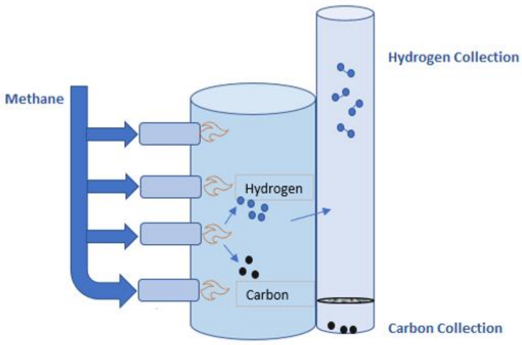

Figure 5. Hydrogen rich gas conversion unit

In this model, EPi pyrolysis unit converts the feedstock into syngas and carbon char. Syngas produced by pyrolysis contains mainly methane, hydrogen, CO and CO_2. The gas refining unit is configured to convert syngas into methane rich gas via catalytic reactors. Final production of hydrogen takes place in the last unit, thermal plasma electrolysis, which convert the hydrocarbon rich gas into high purity H_2 and carbon black as presented in Figure 5.

The high-fidelity model demonstrates that hydrogen with 99% purity) is produced, with 25 kg/h CO_2 emissions.

The primary objective of the project is to produce hydrogen, as mentioned earlier. According to the model, it predicts that 19% of waste can be converted into hydrogen. It is important to note that alongside hydrogen, two other significant by-products are produced in the process: char and carbon black. These by-products not only capture and sequester carbon but also have diverse applications. There are numerous opportunities to generate additional income by utilizing the carbon char derived from the feedstock, especially since the char is predominantly produced from the biogenic fraction of the feed material. Potential uses include soil improvement, remediation of contaminated solids, and construction materials for roads and other applications. Carbon black is a valuable by-product, which provides a good carbon capture option. It is stable and fine powder that is an amorphous form of carbon, possessing a structure akin to disordered graphite [27]. Carbon black stands as a cutting-edge material with many uses. It serves as a black pigment in printing inks, plastics, coatings, and paints. Furthermore, carbon black is crucially employed as a filler in tires, reinforcements, electric conductive agents [28][29].

The experimental validation tests have been conducted to confirm the performance of catalysts and flow rates determined by the high-fidelity model. The outcome of the experiments confirmed the vessel dimensions, flow rates, and performance of the catalysts as initially determined by the high-fidelity model. The gas output of each reactor was confirmed as being in line with the model. The trials have demonstrated the exact operation conditions for the reactors in Gas Refinery Unit to match the products composition set in the whole plant model.

Experimental studies signified that the design parameters and specifications of the reactors, including vessel dimensions, flow rates, and catalyst performance, have been verified and can be confidently implemented in the final design of the commercial-scale plant.

Following the finalization of the design configuration, a carbon life cycle assessment of the Pure Pyrolysis Refined Project has been performed by an external body, to evaluate the environmental impacts of the products produced and each of the processes in the proposed configuration.

The carbon emissions from the process are:

1. Emissions from utilisation of the carbon char (assumed zero if not combusted)
2. Emissions from the release of CO_2 to atmosphere
3. Any scope 2 emissions caused by plant parasitic energy requirements
4. Emissions captured and sequestered in the form of carbon char and carbon black generated from the pyrolyser and the hydrogen rich gas conversion unit.

The results of the carbon assessment show that the process is found to release 0.057 tonnes/hr of carbon dioxide emissions to atmosphere, of which 0.013 tonnes/hour is biogenic and can be treated as neutral, resulting in 0.043 tonnes/hr carbon dioxide emissions to atmosphere. The process captures a total 2.093 tonnes/hr of carbon dioxide in carbon char and carbon black, of which 0.795 tonnes/hr is biogenic and therefore can be treated as negative. Therefore, by this rationale, the process results in 0.752 tonnes/hour net negative emissions as shown in Figure 6.

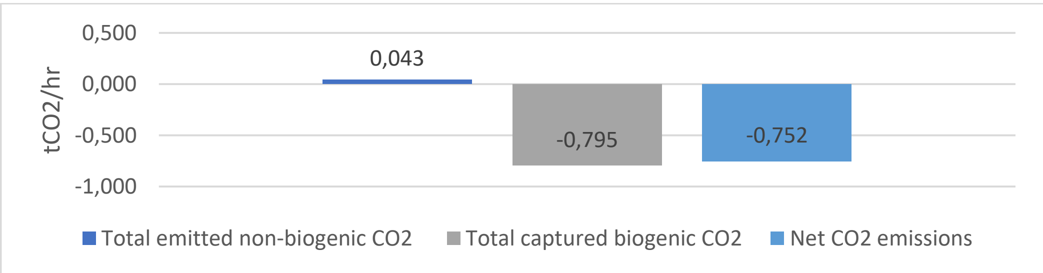

Figure 6. Net emissions from pure pyrolysis process

4. Conclusion

EPi Ltd., in collaboration with its partners, offers an integrated system designed to convert waste into high-calorific-value fuel gas, hydrogen and carbon char and carbon black, aligning with the objectives of the H2BECCS programme. This article summarizes the comprehensive studies conducted throughout the project to meet the goal of hydrogen production from biogenic feedstocks (minimum 25% by calorific value) with minimal carbon emissions.

The final design proposal emerges from an extensive literature review, incorporating findings from two-stage laboratory-scale pyrolysis trials and 15 simulations conducted using the gPROMS™ Process 2.2 computerized model.

The proposed design integrates several complementary technologies into the high-temperature pyrolysis unit to maximise hydrogen yield while minimizing CO_2 emissions. This configuration is engineered to produce hydrogen efficiently from various organic feedstocks, accommodating fluctuations in input waste materials. Laboratory trials conducted on a blend of SRF and waste HDPE, coupled with simulation results, demonstrate the capability of this configuration to convert 19% of waste into hydrogen with 99% purity.

Furthermore, the proposed design offers significant potential for carbon capture through its by-products, including 541 kg/hr of carbon black, 100 kg/hr of carbon char, and 100 kg/hr of oil derived from pyrolysis and subsequent processing of 1 tonne/hr of waste.

The carbon assessment results underscore the environmental benefits of EPi's pure pyrolysis process, revealing a net negative emission of 0.752 tonnes per hour. This achievement underscores the project's success in meeting the stringent requirements of the programme.

In conclusion, EPi and its collaborators have advanced hydrogen production technologies while making substantial strides towards sustainable waste management and carbon capture. This integrated approach not only supports the goals of the H2BECCS programme but also sets a benchmark for future innovations in clean energy production.

Ethics in Publishing

There are no ethical issues regarding the publication of this study.

Author Contributions

Sinem Kurt Yuzbasioglu contributed to conceptualization, methodology, investigation, writing - original draft preparation. Hayati Olgun provided supervision, reviewed and edited the article.

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