

NUCLEAR POWER FOR THE PRODUCTION OF INDUSTRIAL PRODUCTS FROM MIXED PLASTIC WASTE

Mickey Bresnahan^{*1}, Vivek Utgikar², Piyush Sabharwal³

¹Idaho National Laboratory, USA

²University of Idaho, USA

³Idaho National Laboratory

Received: 25/07/2025

Revised: 02/09/2026

Accepted: 01/10/2026

ABSTRACT

A potential solution to the problem of management of ever-increasing plastic waste through chemical recycling using nuclear energy is presented in this paper. The nuclear plant is co-located with the plant decomposing the plastic waste into feedstock chemicals and the one upcycling the feedstock chemicals into high value industrial products such as steel and transportation fuel. Material and energy balances are performed for a number of subsystems producing iron, steel, hydrocarbon fuels, ammonia products, and polycarbonate plastics coupled to a nominal 10 MWt nuclear microreactor. Such a microreactor could facilitate the conversion of 25 metric tons of plastic to valuable industrial products each day and maintain carbon neutrality.

KEYWORDS: Gasification, Haber-Bosch, Fischer-Tropsch, Microreactor, Syngas, plastic

1. INTRODUCTION

Plastics are a miracle material due to their light weight, durability, ease of processing and use, and cost as well as energy advantages compared to materials such as metals and alloys. The global consumption of plastics has increased continuously since the 1950s, leading to a challenging issue of management of the plastic waste. Each year between 800,000 and 2.4 million Mt of plastic will enter the world's oceans [1], at a rate greater than 1.5 Mt per minute. This immense stream of plastic waste only accounts for the portion flowing into the oceans worldwide and does not include the amount of plastic waste disposed of in the landfills. Increasing global demand for plastic products, anticipated to be 884 million Mt by 2050 [2], will only exacerbate the challenges associated with the management of plastic waste. As the consumption of plastic products is not expected to decrease in the near future, a sustainable solution for dealing with the plastic waste stream is required. Plastics, with hydrocarbons (and their derivatives) constituting as building blocks, have potential to serve as a chemical feedstock, suggesting that the ideal method of plastic waste management may be to recycle it into these chemical products.

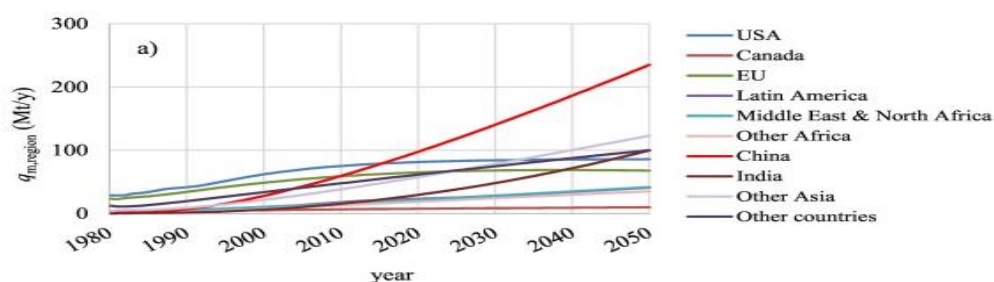


Figure 1: Global plastic consumption as a function of time including projected consumption until 2050 [2]

As of 2019, only 5% of plastic waste in the U.S was recycled [3]. Recycling a greater percentage of our plastic waste may seem like the obvious solution, however not all methods of recycling are created equally. There are four different types of plastic recycling: primary, secondary, tertiary, and quaternary. Each method represents a different level of waste decomposition as summarized in Table 1.

Table 1. Description of the four variations of plastic recycling

Primary	The waste is reused directly without significant alteration
Secondary	The waste is altered from its original form; however, the polymer chains are maintained. This is also called mechanical recycling.
Tertiary	The waste is broken down to its base components by cracking the polymer chains. This is called chemical recycling
Quaternary	In this case the waste is converted into energy (incineration). Most would not consider this to be true recycling.

Primary recycling is essentially reusing what has been thrown away and fails to fully address the problem since not all waste can simply be reused directly. Quaternary recycling has the intention of energy production through waste incineration. Incineration will effectively eliminate plastic waste but will result in significant greenhouse gas (GHG) emissions. In addition, the formation of undesirable and toxic compounds (such as dioxins) from comingled plastic waste stream creates significant technical and regulatory challenges in the management of exhaust flue gases.

Secondary or mechanical recycling is often the implied meaning understood by public when plastic reuse is mentioned. The plastic waste stream is manually sorted, so that each type of plastic can be grouped together, this allows for the removal of the "recyclable" plastics. After sorting, the plastic is ground into small pieces, melted down and reformed into plastic pellets which can be used to produce new plastic products having the same chemical identity as in the original waste stream. This process is labor intensive however and has heavy restrictions on what type of feed it can accept. Plastic products are assigned a number 1-7 known as the resin identification code (RIC). Depending on the RIC, the plastic may not be recyclable through mechanical means, as shown in Table 2 [4].

Table 2. Plastic recyclability based upon RIC

RIC	Plastic Type	Recyclable
1	Polyethylene Terephthalate (PET)	yes
2	High Density Polyethylene (HDPE)	yes
3	Polyvinyl Chloride (PVC)	no
4	Low Density Polyethylene (LDPE)	not always
5	Polypropylene (PP)	not always
6	Polystyrene (PS)	not always
7	Polycarbonate (PC), ABS, PLA, etc.	no

With mechanical recycling, a significant portion of the consumer waste stream is deemed not recyclable. The alternative is to utilize tertiary or chemical recycling. Chemical recycling involves disrupting the polymer chains, usually with energy input, to form base hydrocarbons and intermediate products such as pyrolysis oil, char, syngas, etc. Hard to recycle plastics, such as PVC or PS, can also be amenable to chemical recycling. The specific composition and yield of the products of chemical recycling depend on waste composition as well as the operating conditions of the chosen recycling process [7].

Both the liquid and gaseous outputs of the plastics processing are either valuable products, or feedstock for the production of valuable products such as transportation fuels, fertilizers, plastic products, and much more [11]. In the ideal configuration of the chemical recycling process, the plastics processing plant and the chemical process plant are co-located with the energy source that can provide the needed thermal and electrical energy needed by both the plants. A critically important factor in choosing a centralized energy source for such a system is its capacity factor, which should be as high as possible. Further, this energy source should be preferably carbon-neutral or low-carbon to minimize GHG emissions, leading to the following four options: nuclear, geothermal, wind, and solar. Wind and solar are eliminated from further consideration due to the temperature and process heat requirements that are available only with nuclear and geothermal. Based on the capacity factors shown in Table 3, nuclear is the only logical option for serving as the primary energy source for the two chemical processes.



Table 3. Capacity factors for different energy generation technologies in the U.S (2024) [34]

Source of Power Generation	Capacity Factor in the U.S (2024)
Nuclear	92%
Geothermal	65%
Natural Gas	59.7%
Wind	34.3%
Solar	23.4%
Coal	42.6%

A light water reactor (LWR) representing a typical operational nuclear reactor in the U.S. will have an outlet temperature of 290-320°C. Advanced reactors are capable of producing higher quality process heat (in excess of 500°C) as well as operating at high power density. Even the smallest class of reactors, microreactors, are capable of producing up to 50MW of thermal power while still being portable, modular units. Due to the high capacity factor a nuclear reactor will be able to supply heat and electricity to supply consistently to each process. This, in conjunction with the high-quality process heat, makes nuclear power the ideal candidate for such a design.

The objective of this work is to present a design for a multipurpose co-generation plant for recycling plastic, producing fertilizer, steel, liquid fuel products, as well as electricity. This design was named the high exergy multi-process plant (HEMP). The next section will discuss the finer details of chemical recycling, with the primary focus being on how parameters such as temperature, pressure, and oxygen content of the environment affect the energy input as well as the product distribution. Section 3 will provide a high-level overview of HEMPs architecture as well as description of the many reaction constraints and how they are treated throughout the study. Section 4 will be an overview of the Haber-Bosch process used for fertilizer production. The same will be done in sections 5, 6 and 7 for iron and polycarbonate plastic production as well as Fischer-Tropsch synthesis. Section 8 will describe the energy and mass balance for the entire plant. This will initially be done on a microreactor scale and then generalized to a large-scale facility. After the mass and energy balance has been presented, we will be ready to discuss the specific requirements for the nuclear reactor.

2. CHEMICAL RECYCLING OF PLASTIC WASTE

Chemical recycling of plastic involves breaking down the polymer linkages into a plethora of constituent species that are typically simple hydrocarbons, or components of syngas. This process requires heat and typically makes use of a catalyst. Depending on the conditions in which the reaction takes place, the feed stock can be subjected to either pyrolysis or gasification. Pyrolysis takes place at lower temperatures and without the presence of oxygen, whereas gasification takes place at higher temperatures and involves small amounts (sub-stoichiometric quantities) of oxygen to the system. Increasing the oxygen content results in incineration (complete oxidation) of the material treated. The conceptual differences in the processes are shown in Figure 3. Missing Figure 2? All figure numbers will need to be fixed then.

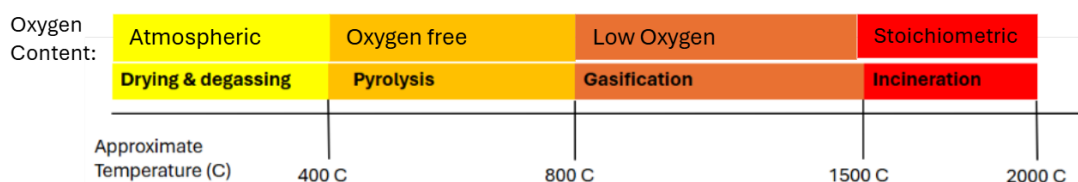


Figure 2: Above is a chart showing the type of chemical recycling process taking place as a function of temperature. The figure was adapted from [6].

Plastic Pyrolysis

The process of pyrolysis takes place in the temperature range of 400 – 800°C, and in an inert, oxygen-free atmosphere. The pyrolysis process can use any plastic as feedstock, however the deviations in the content of the feedstock will have a significant impact on the product distribution [7]. Other factors such as the catalyst used and the heating rate will also have significant effects on the product yield. The

kinetics of the pyrolysis process will be primarily dependent upon the heating rate. Higher heating rates are typically achieved with higher reaction temperatures therefore a process with a high heating rate likely operates near the upper bound of the pyrolysis region. The reaction will have a characteristic time associated with it, typically of the order of seconds or milliseconds. A slow pyrolysis process is one where the time taken to heat the feedstock (t_H) is much greater than the reaction time (t_R). A fast system is one where the heating time is far less than the reaction time. In this case far less or far greater simply means an order of magnitude difference [5].

Slow: $t_H \gg t_R$

Fast: $t_H \ll t_R$

A typical pyrolysis process will yield char, pyrolysis oil, and a gaseous component. The solid component is a char-like substance typically having a high ash content, and a moderate heating value. Pyrolysis oil is the liquid component of the product and is composed of phenols, aromatics, alcohols and several other hydrocarbons. The pyrolysis oil is the most desired product, so it is ideal to maximize its yield. The gaseous component is a syngas composed of H_2 , CO, CO_2 , and CH_4 . A faster pyrolysis process will yield a more gaseous product, whereas a slower process will yield more oil-like products. General estimates of the mass yield and heating value of each product are shown below [7].

- Char: 20-50% of the overall yield, with heating values in the range of 10-35 MJ/kg
- Pyrolysis oil: 30-50% of the overall yield, with heating values in the range of 5-15 MJ/kg
- Gas: 20-50% of the overall yield, with heating values in the range of 3-12 MJ/kg

Figure 4 depicts the overall pyrolysis process. Pyrolysis is a multi-step process, and while the outputs may vary with operating conditions, feedstock, and choice of catalyst, the steps remain the same.

1. Plastic waste is dried and fed into a hopper where it can be shredded into small flakes and fed into an oxygen free reaction chamber
2. Within the reactor chamber the contents are heated rapidly in the presence of a catalyst. This will induce thermal decomposition of the feedstock
3. The solid component will be removed from the stream via cyclone and the remaining stream will enter a condenser which will separate the pyrolysis oil from the gas.
4. The now separated liquid and gaseous products can be further refined into marketable products

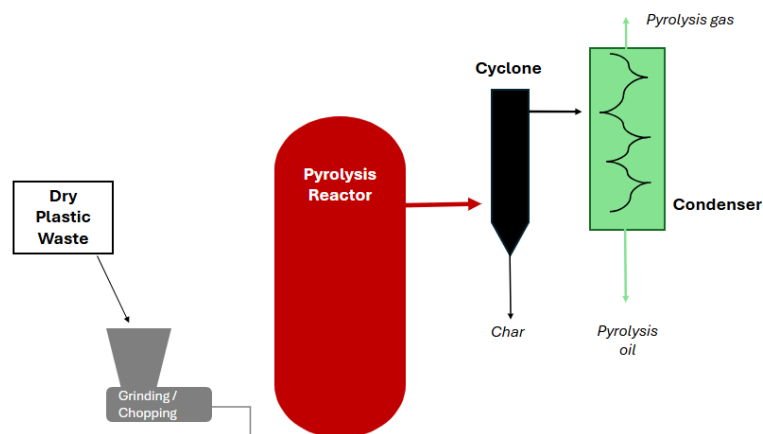


Figure 3: Schematic of the pyrolysis process for biomass feedstock, however the process for plastic is identical

In general, an increase in temperature will yield a greater gas fraction, this is also true for higher pressures. When using mixed plastic from MSW as compared to a pure feedstock the liquid fraction will tend to be lower [7]. Special consideration should also be taken for streams containing PVC due to the inevitable chlorine content in the product stream. The chlorine species will primarily form in the gas phase (HCl) [8]. It will be necessary to include a removal system for acidic species such as a hydroxide scrubber.

Plastic Gasification

The gasification process is similar to pyrolysis except it takes place at higher temperatures (up to 1200° C) and in the presence of a small amount of oxygen. In gasification, the more desirable product is syngas, however, solids and liquids are still produced. The solid component is a char-like particulate similar to what is produced during pyrolysis. The liquid component is called tar and is generally considered to be a contaminant rather than a desirable product. Gasification tar, however, still contains many valuable hydrocarbons, including aromatics such as BTEX and naphthalene. More details regarding the tar product will be discussed in section 8. The gaseous component is a high-quality syngas similar to pyrolysis gas.

- Char: 30-50%
- Tar: 10-20%
- Gas: 30-60%

The gasification process for plastic waste comingled with the municipal solid waste (MSW) is outlined below.

1. The MSW is dried and fed into a hopper with plastic waste to produce shredded waste stream, like the pyrolysis process.
2. The shredded waste, along with the gasification agent, is fed into the reactor where it is rapidly heated.
3. The high heating rate in the presence of a catalyst will generate primarily volatile species which will rise to the top of the reactor to be removed. Some char will be deposited in the bottom of the vessel.
4. Any solid particulates remaining in the stream can be removed via cyclone followed by a condenser for tar removal.
5. After char and tar removal, the syngas is ready to be cleaned and distributed

One of the most important parameters to control in a gasification process is the gasification agent, which is the medium in which the process takes place. Common agents of choice are air, oxygen, steam or a mixture of the three. Air is the cheapest option, while steam is the most expensive due to the high energy input required. CO₂ has also been considered since it will yield a high overall gas fraction, but it will shift the gas towards higher CO content. Table 4 compares the different gasifying agents.

Table 4. The effects that the gasification agent can have on the output stream [8]

Agent	Pros	Cons
Air	low cost	- low hydrogen content -prone to coking reactions -will yield a significant amount of nitrogen
Steam	-high hydrogen yield -high tar yield	-can have a low gas yield- requires high input energy
Oxygen	-less expensive than steam -no nitrogen contamination	-low hydrogen yield -high energy input (for ASU – air separation unit)

Much like pyrolysis, a higher temperature will tend to increase the overall gas yield, and the feedstock composition can alter the composition of the syngas. Gasification is much less sensitive to changes in feedstock than pyrolysis. There are, however, well-defined trends based upon the feedstock composition. Both H₂ and CO production increases with decreasing oxygen in the feed, decreasing pressure, and increasing temperature. Methane production will be favored by low temperatures and high pressures, whereas CO₂ is favored with increasing oxygen feed. CO₂ production is also suppressed by high temperatures and low pressures. [5, 9].

For the purpose of the HEMP design, plastic gasification will be the preferred process due to its superior gas yield and insensitivity to the composition of the feedstock. It will be ideal to maximize the hydrogen yield in the syngas therefore the gasification agent of choice will be steam. The temperature of

the steam will be high (900°C) and be at a relatively low pressure of 5 bar. With this configuration, the gas component is expected to be high in H₂ and CO while being low in CH₄ and CO₂.

3. PLANT OVERVIEW

HEMP is a co-production plant intended to produce, steel, fertilizer, liquid fuels, and plastics using feedstock generated from chemical recycling of MSW. This plant seeks to solve two problems at once. The first being the recycling of plastic waste, and second being reduction in the CO₂ emitted from industrial processes. A successful design will need to satisfy several criteria which are listed below.

1. The plant must be able to consume large amounts of plastic waste without requiring significant sorting or pre-conditioning
2. The chemical recycling process must be able to always deliver a steady supply of feedstock to all processes
3. All carbon species produced must be used to maintain carbon neutrality
4. The overall design must be scalable and easy to implement
5. Each industrial process used must produce a meaningful quantity of valuable resources

Several processes will be needed to consume the entire gasification syngas product. The syngas vector produced will be a H₂-CO-CO₂-CH₄ mixture. The methane component is a valuable product on its own, however a steam methane reforming process could be used to convert it into a mix of H₂ and CO. In order to consume the rest of the gas, processes that rely on hydrogen and either CO or CO₂ will be utilized. When dividing the gas stream among the processes the hydrogen component will be the limiting reagent. Table 5. is a summary of the processes that will be used.

Table 5. Overview of the inputs and outputs from each process in within the HEMP design

Production Process	Input	Output
Haber-Bosch	- H ₂ - N ₂ - Heat & electricity	- NH ₃ - Urea - Ammonium Nitrate - Nitric Acid
Iron & Steel	- Iron ore - Limestone - H ₂ - O ₂ - electricity	- Iron - Steel
Fischer-Tropsch	- H ₂ - O ₂	- LPG - Naphtha - Kerosene - FT wax
Polycarbonate Plastics	- CO ₂ - Propylene Oxide - heat	- Polycarbonate - Polypropylene - Carbonate

Reaction constraints

For this study, mass flow rates of reactants and produced were primarily scaled from work done by McMillan *et al.* [11]; which detailed information on the production rates of nominal sized systems. The product and reactant flows were given in Mt/day. This means that the kinetics of the reaction is already accounted for in the scaling of the processes and no explicit kinetics consideration is required. Throughout the plant, most processes will be endothermic, requiring energy as part of the feed. The energy input required for endothermic reactions in conventional industrial processes often requires a portion of the feedstock to be diverted from the main flow and combusted. This is done to satisfy the thermal constraint of the reaction. However, since the heat is externally supplied by the nuclear reactor this thermal constraint becomes irrelevant. The only remaining constraint is the stoichiometric constraint which will be the primary constraint for the design of the system. Many proposals have been made for the coupling of nuclear reactors to industrial processes; however, it is the coupling of multiple industrial processes in a carbon-free manner that makes HEMP unique.

4. AMMONIA AND FERTILIZER PRODUCTION

In 1908 Fritz Haber and Carl Bosch developed a process for producing fertilizer from hydrogen and nitrogen, later named the Haber-Bosch process [10]. In 2023, 14 million Mt of Ammonia was produced, 88% of which is used to produce fertilizer [11], the other 12% is used for explosives, synthetic fibers and resins. Figure 4 below describes the energy and mass balance for ammonia production.

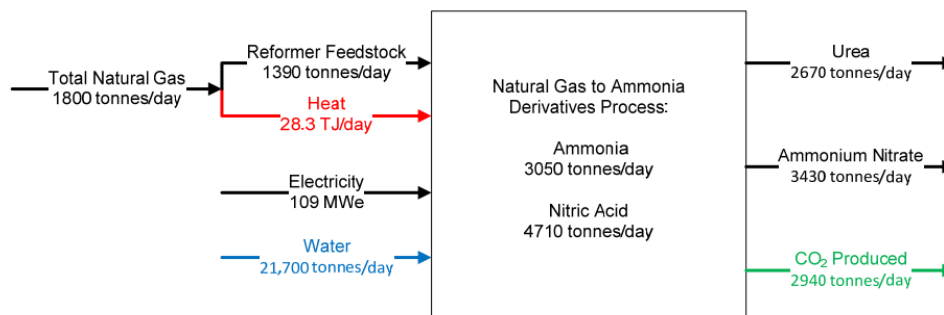
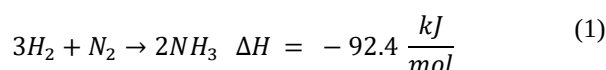


Figure 4: Mass and energy balance for a nominal full size ammonia production plant taken from [11]

Haber-Bosch Process

Producing ammonia is always the first step since all other products cannot be synthesized without ammonia. This reaction is exothermic, reversible and requires a hydrogen to nitrogen molar ratio of 3:1.



The feed is kept at a pressure of 13.8 MPa, and a max temperature of 500°C [11]. This reaction typically takes place in the presence of an iron-based catalyst. The product is then cooled below its boiling point (99°C at atmospheric pressure) and stored in the liquid phase. Cooling the product is often done by recuperation in a steam generator. This entire process is shown in Figure 5.

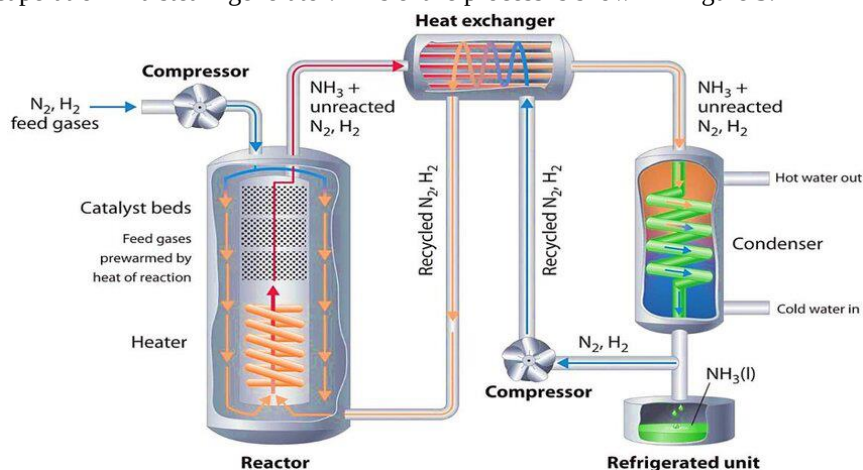
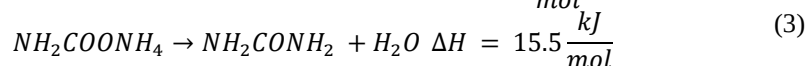
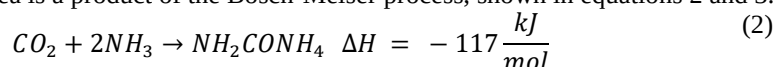


Figure 5: Diagram showing the ammonia synthesis step as well as the recuperation of heat with a steam generator

Bosch-Meiser Process

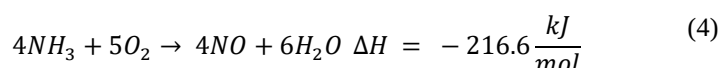
Ammonia is an intermediate product used to produce two final products: Urea and Ammonium Nitrate. Urea is a product of the Bosch-Meiser process, shown in equations 2 and 3.



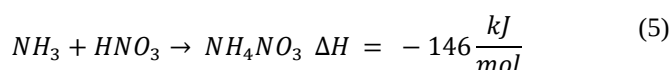
The two reactants (CO₂ and NH₃) are combined at high temperature and pressure (110 atm and 160° C) to form ammonium carbamate which will then slowly convert into urea and water [12]. The liquid product is stripped of excess CO₂ and fed back into the first reaction. Because of the CO₂ stripping process some trace amounts of CO₂ may be released from this process. These trace amounts can be mixed with the H₂ – CO syngas and sent off to the Fischer-Tropsch process.

Oswald Process & Ammonium Nitrate Production

Nitric acid is produced through the Oswald process which combusts ammonia with air under high pressure and temperature (800°C and 5 atm). The combustion yields water and nitric oxide which is cooled and oxidized to form nitric acid [13].



This reaction is extremely exothermic and can be used to recuperate heat and improve process efficiency. After oxidizing with oxygen and water, nitric oxide yields nitric acid (HNO₃). This nitric acid is then reacted with ammonia to produce ammonium nitrate.



As seen above, ammonia and nitric acid are typically intermediate products whereas urea and ammonium nitrate are the main products. This is not always the case; ammonia and nitric acid are valuable products on their own.

Nuclear Energy for Ammonia Production

A full-size ammonia production plant will produce several thousand metric tonnes of ammonia and emit just as much CO₂ every day. The primary source of this CO₂ is from the combustion and reforming of natural gas. In a traditional ammonia plant, each Mt of ammonia produced will require over 590 kg of natural gas and release 963 kg of CO₂ into the atmosphere [11]. The combustion of natural gas can be replaced with process heat from the nuclear plant whereas the steam reforming can be replaced by the H₂ and CO₂ components of the syngas generated from the gasification of plastic waste. Nitrogen can be acquired from an air separation unit (ASU). Using these methods for obtaining feedstock eliminates the need for natural gas and therefore eliminates the carbon emissions.

5. IRON & STEEL MANUFACTURING

Iron and steel production accounts for 7% of the world's CO₂ emissions [14], making it one of the largest CO₂ emitting industries. Aside from raw iron ore, coal and natural gas are the primary feed stocks for iron & steel plants. A traditional steel mill will require 7 Mt of feedstock to produce a single Mt of steel. For each Mt of steel produced nearly twice as much CO₂ is released. The mass and energy balance for the process is shown in Figure 6.

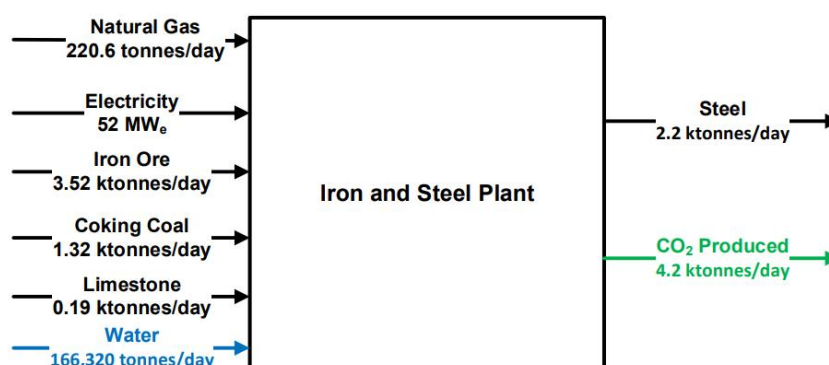


Figure 6: Mass and energy balance for a full-scale iron/steel plant taken from [11]

Iron Production

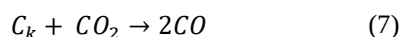
Steel is produced from iron which is produced by melting and reducing iron ore. The first step is the production of coke. Coke is produced by carbonization of coal at high temperatures (1100° C) [11]. The

next step is to introduce the coke, iron ore and limestone into the blast furnace. Coke enters from the top of the furnace and is ignited by the introduction of air, producing heat and CO₂. The carbon dioxide will reduce to CO which will reduce the iron oxide ore to pure iron. The heat released will melt ore producing molten iron. The limestone will react with impurities to produce slag; this slag will float to the top of the pool, separating it from the pure liquid product [11].

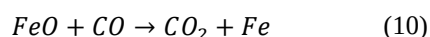
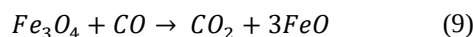
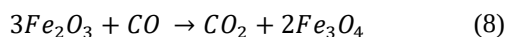
As the coke descends it will undergo various chemical reactions with the blast air. Initially the coke will react with the air.



The CO₂ is then reduced by excess carbon.



As the CO rises to the top of the blast furnace it will reduce the iron ore. The ore will undergo several reductions producing CO₂ in the process.



Fully reduced hot metal, which is just molten iron, reaches the bottom of the furnace. A visual representation of this process can be seen in appendix A.1.

Steel Production

Steel can be thought of as high carbon iron. From molten iron there are two primary methods of steel production; basic oxygen furnace (BOF) and the electric arc furnace (EAF). The BOF introduces the molten iron to pure oxygen producing iron oxide and evolving both CO and CO₂. The EAF uses high current and graphite electrodes heated to over 2200°C to remove impurities from the molten iron, The electrodes will burn and produce another molten metal. These impurities form a slag and are removed from the melt, yielding molten steel [11]. The EAF is a batch-wise process where a new batch must begin each time the electrodes have been completely burnt. Both methods are capable of using scrap steel as input. This helps build a circular steel economy. A diagram of an EAF can be found in the appendix A.2.

Nuclear Energy for Iron & Steel Production

The primary source of carbon emission for iron production is the blast furnace. The iron ore must be melted and reduced meaning that whatever method is used to replace the blast furnace must be able to accomplish both of these tasks. Hydrogen direct-reduction will be used (H-DR) to accomplish this. In this process hydrogen is used as both the heat source and the reducing agent. Among the various methods being proposed to reduce carbon emissions of steel manufacturing H-DR is the most promising, with the most significant challenge being the high cost of producing green hydrogen [15]. HEMP produces the hydrogen from plastic waste and therefore avoids this issue. Directly reduced iron (DRI) can then be used in the same manner as before in order to produce steel. If a BOF is used hydrogen combustion can act as the heat source; if an EAF is used, the on-site reactor will supply the electricity. Regardless of which furnace is used, the process remains carbon neutral.

6. POLYCARBONATE PRODUCTION

Polycarbonates are high temperature plastics known for their impact resistance, good durability and transparency. Because of its favorable properties, PC is a common construction material and is used in the lenses of eyeglasses, cameras, and automotive headlights. Although useful for certain applications PC is not nearly as popular as PE, PP, or PET. With a resin number of 7, PC can be tough to recycle. In 2019 only 1% of all plastics with a resin number of 7 were recycled [3].

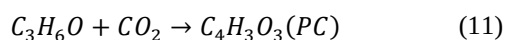
Several methods exist for the production of PC, however the primary method is through transesterification. This process has two reactants, bisphenol A (BPA) and dimethyl carbonate, and requires temperatures between 200°C and 250°C. Methane is produced as a byproduct and is removed to drive the reaction towards polymer production [15]. Several other methods exist, however they each have their own drawbacks that make them less popular than transesterification.

- Direct polymerization method (by-product formation is significant and reaction is energy intensive)
- Carbonate polymerization method (promising method with low energy requirements and high yield, but has little production experience)
- Solution polymerization (easy to control process but has low production efficiency and is not suitable for large scale applications)

Nuclear Energy for Producing Polycarbonates

As with most industrial processes, the main source of carbon emissions is from the production of process heat. Emission of CO₂ however is not the only problem with plastic production. BPA has been shown to have adverse effects on reproductive organs and endocrine function [17]. Ideally a new PC production process would be both carbon and BPA free.

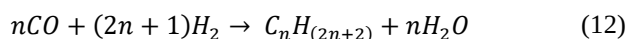
Due to advances in catalysts, a new production process involving CO₂ and propylene oxide can be considered. This process eliminates the need for BPA and makes use of CO₂. A report from The University of Nebraska Lincoln (UNL) outlines such a process through simulation of an industrial scale plant [16]. In the presence of a catalyst, under high pressure (114.7 psi) and temperature (212°F), the reactants will be converted to PC.



By changing to a chromium chloride-based catalyst and higher pressure (850psi), polypropylene carbonate (PPC) can be produced with a smaller amount of PC forming as a by-product. As long as a carbon free heat source is used, such as nuclear, this process can produce PC and PPC plastics free of carbon emissions.

7. FISCHER-TROPSCH SYNTHESIS

Invented by Franz Fischer and Hans Tropsch in the 1920s, the Fischer-Tropsch process converts syngas (H₂ – CO) into liquid fuels [18]. The process was originally developed for biomass conversion but will work equally well with the gas stream from our plastic waste. Under high temperatures the syngas is polymerized into hydrocarbon chains of various lengths, with the generalized case shown below [18].



This polymerization takes place on the surface of metal catalyst, typically iron or cobalt. The chain length, also known as the carbon number, is determined by the number of monomers in the polymer chain denoted by C_n. The length of the chain will determine what type of products are formed. The length of the chain determines the weight of the product, and its volatility; the lighter the product, the lower the boiling point.

The primary product of a Fischer-Tropsch process is called Syncrude. Syncrude is refined using fractional distillation is essentially identical to that of conventional crude oil, where the lighter hydrocarbons such as methane are collected at the top of the fractionating column, with the products having higher boiling points obtained from progressively lower positions as shown in figure 7 [20]. These products are typically divided into four major fractions:



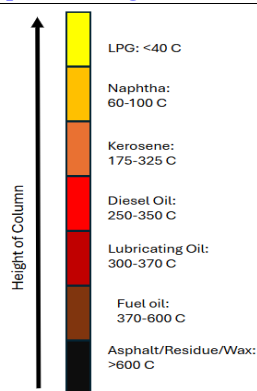


Figure 7: Conventional distillation tower used to fraction crude oil

1. Liquefied Petroleum Gas (LPG) ($C_1 - C_4$): gaseous product, typically less desirable than their heavier counterparts
2. Naphtha ($C_5 - C_{10}$): less volatile than LPG, precursor to light hydrocarbon fuels such as gasoline
3. Middle Distillate (Diesel/Kerosene) ($C_{11} - C_{22}$): These are the heavier liquid hydrocarbons with a higher boiling point than the naphtha fraction. The middle distillate can be upgraded into jet fuel and diesel products.
4. Fischer-Tropsch Wax (C_{23+}): Waxes are the heaviest fraction of the product, they form a solid and are typically the least valuable.

The Anderson-Shults-Flory Distribution (ASF)

The Anderson-Shultz-Flory distribution (ASF) is a function that predicts the product distribution of a Fischer-Tropsch synthesis process. The ASF has two input parameters; the being the chain length (n) and the second being α , which is a variable related to the chain growth probability. The parameter W is the weight fraction of the product having a chain length n .

$$W = \alpha^{n-1}(1 - \alpha^2)n \quad (13)$$

Below is an empirical relation for α developed by (Song, et al.,2004) [19]. T is the temperature in Celsius, and the square brackets denote the mol fraction of a species in the syngas.

$$\alpha = \left(0.2332 \frac{[CO]}{([H_2] + [CO])} + 0.6330 \right) (1 - 0.0039(T - 260)) \quad (14)$$

As easily seen above, the value of α depends on both the temperature and the composition of syngas fed into the process. As the value of α changes, the product yield distribution will change. A higher value of α will favor a heavier product yield with greater wax and middle distillate fraction whereas a lower α will favor the lighter naphtha and LPG products.

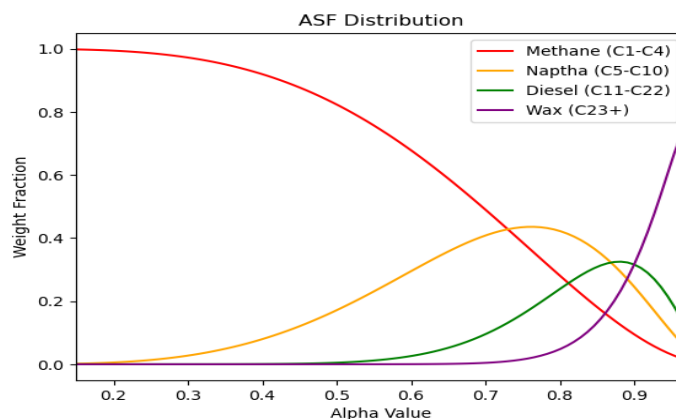


Figure 8: ASF distribution as a function of alpha

Figure 8 shows that α is a weak function of syngas composition and that process temperature is a much more significant variable. The two more desirable products (naphtha and diesel) reach peak yield under different conditions. This means that a choice must be made with respect to which products should be prioritize; this choice will determine the process temperature.

8. MASS & ENERGY BALANCE

The products manufactured from the processes described in the previous sections all rely on similar inputs: heat, electricity, and syngas components. All three of these demands can be met through the use of nuclear micro-reactors. Making use of these modular portable units can reduce the cost and complexity of both construction and operation. In this section the total energy and mass balance of the plant will be presented on a "per 10 MWt microreactor basis". All calculations were done in the interest of stoichiometric constraints for the given reaction. The kinetics of each reaction is considered to be beyond the scope of this work.

Our goal is to maximize the hydrogen yield of our syngas, since it is going to be the limiting reagent for our coupled processes. For our gasification reaction, the medium of choice will be high temperature steam at 900°C and 5 bar. Steam was chosen as the gasification agent for its high hydrogen and low carbon syngas yield. Steam has also been associated with a lower overall gas yield, however this can be partially made up for with higher temperatures. The increase tar yield from using steam as the gasification agent since the liquid component still contains valuable species.

Gasification Products

The molar ratio of hydrogen to carbon monoxide in the syngas produced in the gasifier determines the quantity of final product that can be produced from each process. However, as previously mentioned the composition of the syngas will be dependent upon the feedstock used. The goal of this design is to consume plastic waste without the need of labor-intensive separation of undesirable feed. The data reported by Mojaver, et al. 2021[22], that have been selected for this design.

The dataset applies to high temperature steam gasification of 4 different types of plastics: PET, PE, PC, and PP. Together these four resins make up 87% of the U.S MSW stream as shown in Table 6 [3], and are a reasonable approximation of the overall U.S plastic waste stream. Table 7. Shows the vector of plastic components used for this design.

Table 6. Breakdown of MSW within the U.S in 2019 [3]

Plastic Type	Mass (kt)	% by mass
PET	5,986	14%
HDPE	7,910	18%
PP	8,189	19%
LEPE/LLDPE	15,139	34%
PVC	3,094	2%
PS/EPS	3,094	7%
Other(PC/PLA/ABS)	3,115	7%

Each type of plastic yields syngas whose composition is dependent upon processing conditions, specifically, temperature, pressure, and the mass ratio of steam to plastic (SPR). High temperature, high SPR, and low pressure are all indicative of a greater hydrogen yield in the syngas, although diminishing returns are seen with temperatures above 850°C and an SPR of 2 [21]. The syngas composition of each plastic feed was taken at an SPR of 2 and a temperature of 900° C; these were then weighted using the values in table 6 to produce the final composition presented in Table .7.

Table 7. Weighting scheme and final syngas composition, all are given in mol fraction. The "Final Composition" column will be the representative syngas vector for MSW [21].

Gas Component	PP	PE	PET	PC	Final Composition
H_2	67.5%	66%	56.5%	59%	64.638%
CO	28%	29.5%	27%	29.3%	28.765%
CO_2	3.5%	3.5%	16%	11%	5.684%
CH_4	TRACE	TRACE	TRACE	TRACE	0.913%

The other component of determining the syngas production term is the overall mass yield with respect to the feedstock. As previously stated, gasification can produce up to 60% syngas, however, a conservative value of 50% is employed in this analysis. The balance of product consists of 20% tar and 30% char. The mass of syngas produced each day is determined by the daily feed of plastic waste. For the scale of a single micro-reactor, the plastic waste feed rate is set to 25 Mt/day; making the steam input 50 Mt/day and bringing the total mass flow to 75 Mt/day. Using the 50/20/30 product yield the product distribution was found to be 4,075 kg of hydrogen, 25,146 kg of carbon monoxide, 7,807 kg of carbon dioxide, and around 455 kg of methane. In addition, it is predicted that roughly 15 Mt of tar and 22.5 Mt of char will form.

The Carbon Dioxide Problem

Our predicted output stream has a little under 8 Mt of CO₂ each day, all of which needs to be used as feedstock so that no carbon is released to the environment. The Bosch-Meiser process utilizes CO₂, however, it also demands ammonia. If all of the CO₂ is used as feedstock for urea production, then the corresponding amount of ammonia required would consume too much hydrogen. As previously stated, hydrogen will be the limiting reagent for most processes. This means we need an additional process that can make use of CO₂ without consuming more hydrogen. For this the polycarbonate production process is used. Thus the CO₂ stream will need to be divided between the Haber-Bosch, and the polycarbonate production processes. It was determined that a desirable distribution of products was achieved when 33% of the stream was sent to the Haber-Bosch process and 67% was left for PC production. This split allows for all of the CO₂ to be consumed while leaving enough hydrogen to produce meaningful amounts of the other products.

Local Mass & Energy Balance for Each Industrial Process

Below are flow diagrams representing the mass and energy balance for each industrial process based upon a single micronuclear reactor and a MSW feed-rate of 25Mt/day.

Ammonia: As previously mentioned, 33% of the CO₂ produced during gasification will be used for production of urea via the Bosch-Meiser process and will be the parameter used to scale the rest of the ammonia production stream. Using the output stream of urea, the rest of the outputs and intermediate products were scaled in accordance with figure 5.

Iron & Steel: For the iron and steel production process the hydrogen and oxygen feed-rates were scaled according to a report describing the potential coupling of the Japanese GTHTR300C to a hydrogen shaft furnace for direct iron reduction [23]. Nitrogen is required for the Haber-Bosch process which is supplied from an ASU. Roughly 20% of the air that enters the ASU yields pure oxygen. This oxygen stream is the basis upon which the other parameters are scaled. The ASU is considered to function as a separate unit from the ammonia plant and will therefore require its own energy consumption calculation.

Polycarbonate: Both the feedstock and energy input for the plastic production process were linearly scaled in accordance with the UNL study. The CO₂ stream was the limiting reagent based upon the 67% branching ratio from the gasifiers CO₂ output. A small component of CO₂ is also included from the combustion of methane, used for stream heating this will be in greater detail in section 8.3.3.

Fischer-Tropsch Synthesis: The ideal input feed for a Fischer-Tropsch process is a 2:1 mol ratio of H₂:CO, however ratios as low as 1.8:1 are deemed acceptable, particularly with the use of an iron catalyst. At the Argonne National Laboratory (ANL) a Fischer-Tropsch synthesis process coupled to a nuclear reactor was modeled using Aspen Plus [24]. During this study it was determined that 1.574 kg of Syncrude will be produced from each kg of H₂ input. This is the value used to estimate the total product yield. This total mass of Syncrude was then fractionated according to the ASF distribution for a feed ratio slightly above 1.94:1 and a process temperature of 238° C. The mol ratio of the syngas was chosen to be in the middle of the acceptable range so that the hydrogen content can be varied without upsetting the mass balance of the system. The hydrogen feed from the gasification will not remain completely constant since the composition of the plastic waste will not be constant. Another option to account for a hydrogen deficit is to increase the mass flow rate of steam and methane entering the SMR.

As previously discussed, the Fischer-Tropsch process is exothermic therefore it will require cooling rather than heat input. The fractionation of the product which takes place at 600° C, however, will require energy input. The input is calculated to be the heat required to raise the temperature of the product to the distillation temperature.

$$Q = m_{crude} C_p \Delta T \quad (15)$$

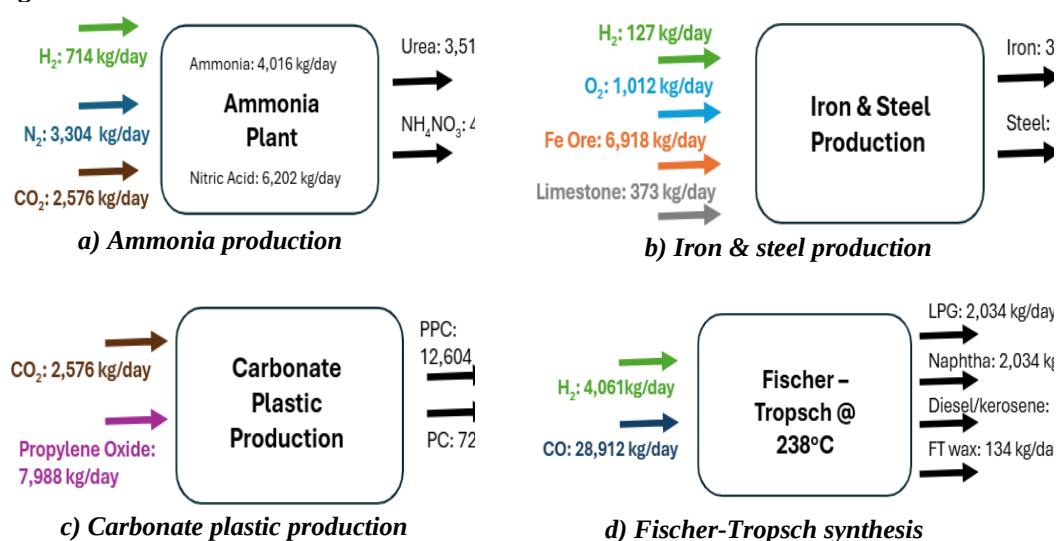
If the temperature of the Syncrude exiting the reaction vessel is assumed to be 238°C then the required temperature rise is 362° C. To determine the specific heat of the crude oil each fraction was approximated as a single product and a weights sum was taken using the properties in table 9.

Table 8. Values used to approximate the specific heat of Syncrude determined to be 2.14 kJ/ (kg K)

Fraction	Representative Product	C _p (kJ /kg K)
LPG	Methane	2.22
Naphtha	Gasoline	2.22
Middle Distillate	Diesel	2.05
Wax	Fuel oil	1.88

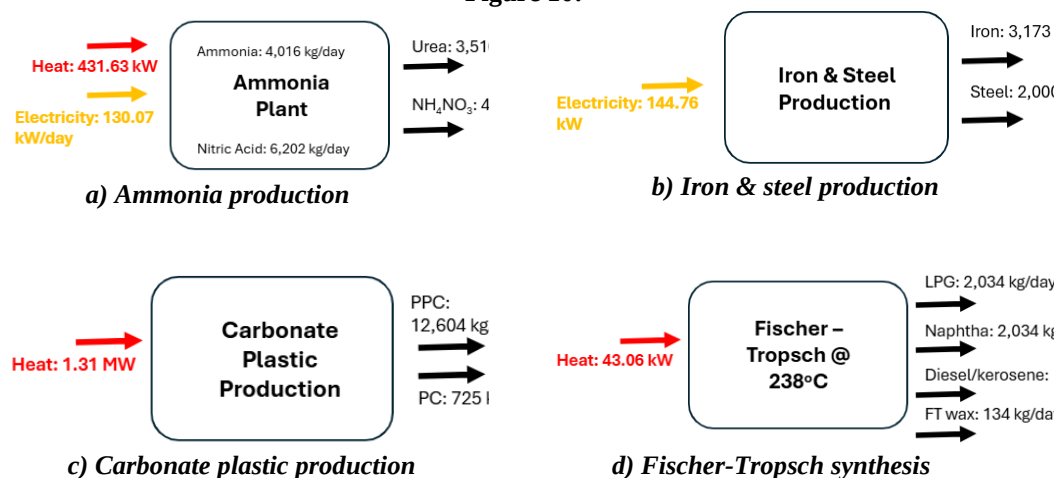
For clarity, the mass and energy balances for each of the nuclear coupled processes described above are shown separately (figure 9 for the mass balance and figure 10 for the energy balance).

Figure 9:



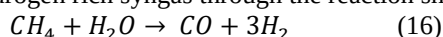
Mass balance for all nuclear coupled processes

Figure 10:



Energy balance for all nuclear coupled processes

Given that the output of methane from the Fischer-Tropsch process will be significant and our hydrogen availability is lower than we would like it to be, a portion of the methane can be siphoned off from the fractionation tower and used in a steam methane reforming (SMR) process to produce more hydrogen rich syngas through the reaction shown in equation 16.



This process requires steam at 700° – 1000°C and 3-35 bar. This means that our 900° C, 5 bar steam supply will be sufficient. To supply this process, the initial quantity of methane and steam input will be taken from the gasification stream at a flow rate of 157.114 kg/day, leaving 300kg/day for combustion. however, as the SMR generates syngas it will increase the yield of the Fischer-Tropsch reactor which will in turn increase the amount of methane that the SMR process can consume. This repeats until the mass flow rate of methane is 2,157.144 kg/day. At this point, combustion of methane at a rate of 300 kg/day can support no additional steam.

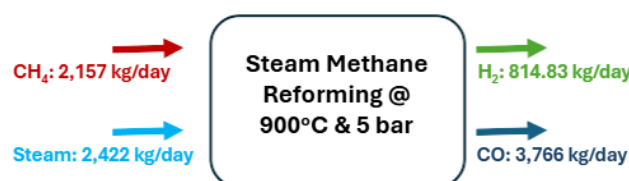


Figure 11: Mass balance for the steam methane reformer

Energy Balance for Supporting Systems

The nuclear reactor serves as the sole energy source for not only the chemical processes but also supporting systems such as the ASU and the cryogenic distillation unit.

Cryogenic Distillation: The cryogenic distillation, aside from the reactor, may be the most important component in the entire plant since it is responsible for separating the syngas into its individual species. The process however is extremely energy intensive with a specific energy consumption (SEC) of 8.85 kWh/kg H₂ [25]. Other processes that consume less energy also exist such as pressure swing adsorption; however, it may have a low recovery rate with respect to cryogenic distillation.

$$P_{CD} = \frac{8.85m_{hydrogen}}{24} \quad (17)$$

Using equation 18 the energy required for the cryogenic distillation unit can be found where the mass flow rate of syngas is in kg/day and P_{CD} is in kW. For our feed-rate of 25Mt/day the energy required will be 1.5 MWe which is by far the largest energy consumption of any process within the plant.

Air Separation Unit (ASU): As previously mentioned, the ASU is required for obtaining nitrogen for the Haber-Bosch process and oxygen to the iron and steel plant. When compared to cryogenic distillation, air separation has a significantly SEC (0.191 kWh/kg air) [26]. Replacing both the mass flow and SEC values we find that the ASU will consume 33.3253 kWe. This is still in kW (power) not kWh (energy).

$$P_{ASU} = \frac{0.191m_{air}}{24} \quad (18)$$

Feedwater Heating & Methane Combustion: Another system that can be considered a secondary process is feedwater heating. Due to the use of steam as the gasification agent, a continuous supply of fresh feedwater will be required. Which means significant power will be needed to supply 900°C steam to both the SMR and the gasification unit. This feedwater can be preheated by other processes that require cooling such as the Fischer-Tropsch reactor as well as the condenser for pre-cooling the syngas before it enters cryogenic distillation.

The Fischer-Tropsch reaction will generate heat at a rate of 165 kJ /mol CO. For the total CO mass flow rate of 28,912.5 Kg/day this equates to a heat generation term of 1.97125 MWh. Feedwater pressurized to 5 bar will be used to cool the vessel. Assuming that the water will be a saturated vapor upon exiting the heat exchanger the required mass flow is found using equation 20 where Q represents the heat generated in the vessel and Δh_g represents the enthalpy rise from ambient temperature (assumed to be 20° C) to the saturation temperature of the feed-water when the quality is 1.

$$m_{water} = \frac{Q}{\Delta h_{og}} \quad (19)$$

The required mass flow is greater than the mass flow required by both the gasification unit and the SMR process. Therefore, the excess portion of the steam can be diverted for supplementary power generation.

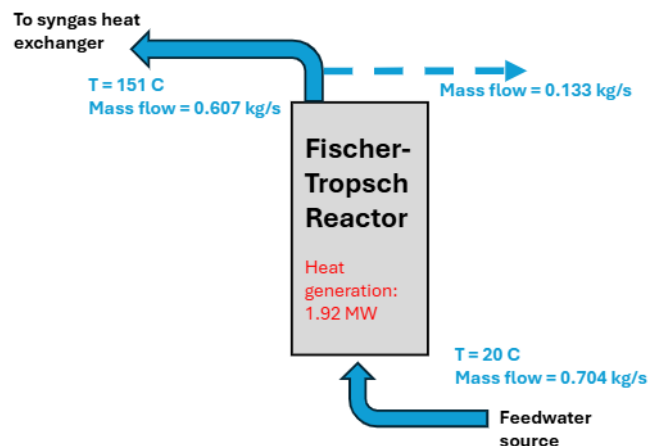


Figure 12: Schematic of the FT cooling in steady state

After cooling the Fischer-Tropsch reactor the saturated steam will be passed on to syngas cooler where the syngas will be cooled from 900°C down to 200°C. Equation 16 was used to determine the heat transfer. The C_p of the syngas was determined through a weighted sum of specific heats obtained at 550°C (the average of the gas inlet and outlet temperature). NIST databases are sparse at high temperatures therefore the properties were obtained from Engineering Toolbox.com [27]. Equation 20 is then used again to determine the enthalpy rise of the steam.

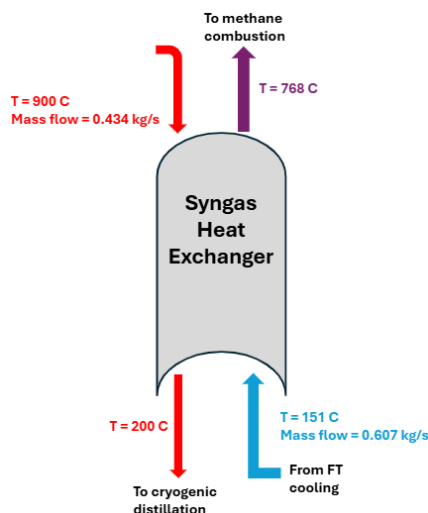


Figure 13: Schematic of the syngas heat exchanger in steady state

Once the steam has exited the syngas heat exchanger it will enter the final heat exchanger where it will be heated through combustion of methane to the SMR and gasification inlet temperature of 900°C. To accomplish this, 300kg of the gasification methane stream is diverted for combustion while the remainder is used to start off the SMR process. Below is the Methane combustion process



This reaction will liberate 55.5 MJ/kg and release significant amounts of CO₂. To ensure that HEMP remains emissions free the CO₂ component is piped off to the PPC/PC production process.

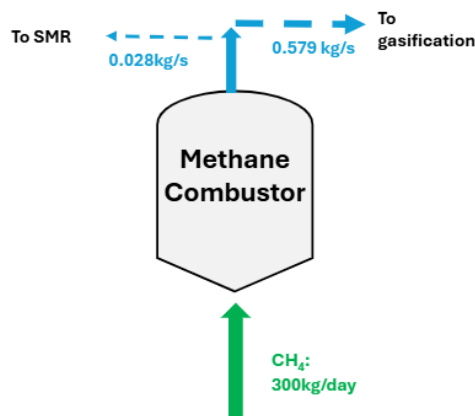


Figure 14: Schematic of the methane-steam heating unit in steady state

Condensed State Products of Gasification

According to the 50/20/30 rule used in 8.1, half of the gasification units' output will not be part of the syngas stream. Between the tar and char components over 37 Mt of product needs to be accounted for each day.

The tar stream produced during gasification is a complex mixture of primarily polycyclic aromatic hydrocarbons (PAH) and BTEX (benzene, toluene, ethylbenzene, and xylene) compounds. BTEX will make up roughly 65% of the tar stream by mass [28], or 9,750 kg/day. BTEX products are precursors for the production of many valuable products such as pharmaceuticals, nylon, and polyurethane as shown in table xx. Additional stream will be produced during the refinement of naphtha products; these two streams can be combined to increase total mass flow of the valuable BTEX precursors [29]. BTEX processing for these products is beyond the scope of this paper.

Table 9. Common products produced from BTEX chemicals [26]

BTEX Species	End Products
Benzene	-Germicidal Paint -Nylon -Adhesives
Toluene	-Polyurethane -Octane Booster -TNT -Paint Solvents
Ethylbenzene	-Polystyrene
o-Xylene p-Xylene m-Xylene	-Apparel -Upholstery -Pharmaceuticals

The remaining fraction of tar is dominated by naphthalene, phenol, pyridine, acenaphthene, and acenaphthylene [27]. The compounds on their own are toxic, posing a potential health risk. Much like the BTEX products however, these PAH species are useful components in the production of plastics, dyes and pesticides [30].

It is expected that over 22Mt of char will be produced each day from the gasification unit. The solid product is a carbonaceous, nonporous char, similar to charcoal used for cooking. This char component can be activated to produce high quality adsorbents. (Solis et al , 2022) explored the use of char produced from plastic waste for lead adsorption in water [31]. This study showed a maximum adsorption of 700 mg(Pb) /g(Char), indicating the potential of application of produced char for environmental cleanup of polluted streams

Overall Mass & Energy Balance

Figure 16, shown below, is a schematic representing the mass and energy flow of the entire plant. Red arrows denote heat inputs while yellow arrows denote electricity inputs. Dotted lines signify streams that are fractions of a predict gas stream. All flow rates are given on a per day, per microreactor basis. All final products are shown in non-bolded black text. The feedwater is shown in the upper right corner, entering the Fischer –Tropsch process to provide cooling, before being sent off to the syngas cooler. The blue heat exchanger between the gasifier and the cryogenic distillation unit is the syngas-feedwater heat exchanger.

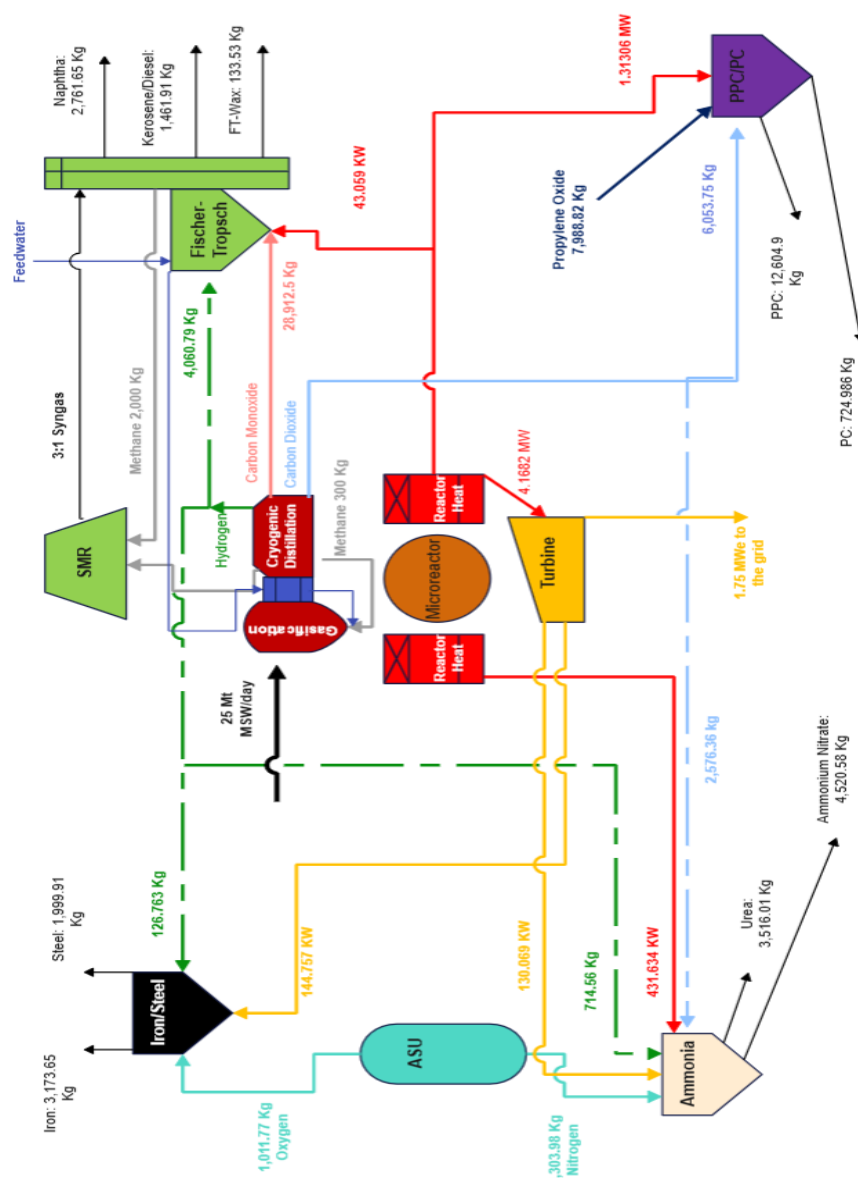


Figure 16: Diagram of the HEMP system in its entirety

9. REQUIREMENTS FOR THE NUCLEAR REACTOR

The total duty on the nuclear reactor will be given in terms of thermal power, with the electrical power requirement for the processes converted into thermal power assuming an efficiency of 45% , which representative of the anticipated efficiency of an HTGR. A summary of the thermal demands from each process is presented in table 10.

Table 10. Thermal power requirements for the microreactor scale plant

Process	Total Thermal Power Required
Ammonia	720 kW
Iron & Steel	321 kW
PC & PPC	1.313 MW
Fischer-Tropsch	43 kW
Cryogenic Distillation	3.34 MW
ASU	94 KW
Total	5.8318 MW

According to table 10 just under 6 MW will be needed to operate all the processes discussed in the previous sections. This means a microreactor rated for 10MWt should be able to comfortably support this processing scheme. Over 4MW (4.1682) is left over for electricity production, which translates to approximately 1.87 MWe. The microreactor should be able to supply ~1.75 MWe consistently to the electric grid with the remainder being available for the energy demands that haven't yet been accounted for. This includes but is not limited to pumps or compressors used to move feedstock between the different processes as well as the reactors house load.

Many of the currently proposed microreactors can meet the power demand; however, the temperature demand is more difficult. To heat the steam to 900°C the reactor outlet would need to be in excess of that. Requiring the reactor to generate steam at these temperatures is simply not practical for near term deployment. It is worth noting however that the Japanese high temperature test reactor (HTTR) sustains a core outlet temperature of 950°C and has done this for several years [32]. The HTTR is a test reactor however, and is not commercially available. If a microreactor comparable to the HTTR was ever produced then the methane combustion system would become obsolete, and the final portion of the steam heating could be performed by the primary coolant of the reactor.

The outlet temperature of the microreactor will still need to be as high as possible (in excess of 500°C). These temperatures can be reasonably achieved by both high temperature gas reactors (HTGR), and molten-salt reactor (MSR). Utilizing an MSR will have the added benefit of online refueling which means that the production of industrial products does not need to stop periodically for refueling, as would be required for an HTGR. This reduction in downtime can improve the profitability of the plant.

Scalability

One of the import design parameters previously stated pertains to scalability, which is one of the most desirable aspects of microreactors. Microreactors are designed to be cheap and portable requiring much lower up front capital investment as compared to traditional large-scale plants. If the demands of the plant change for any reason the power can be uprated simply by adding more microreactors.

To demonstrate the scalability of the system, a plant utilizing a 'fleet' of microreactors can be considered. Many start-up companies are proposing large-scale power plants comprising of several much smaller reactors. One such company is Nuscale (add website) whose primary product is the Nuscale power module (NPM), a small modular reactor rated for 77 MWe. Nuscale has also conceptualized a full-size power plant consisting of 12 NPMs [33]. Although SMRs are significantly larger than microreactors the same concept applies. Below, in table 11 the total product yield is considered for a plant utilizing 12 microreactors, each rated at 10MWt.

Table 11. Estimated product yield for a 12-microreactor plant. This plant would consume 300 Mt of MSW/day and output 21 MWe to the grid

Process	Ammonia	Iron & Steel	PPC/PC	Fischer-Tropsch
Products (Mt/day)	Urea: 42.19 NH ₄ NO ₃ : 54.24	Iron: 38.08 Steel: 23.99	PPC: 151.26 PC: 8.70	LPG: 24.41 Naphtha: 33.14 Diesel: 17.54 FT-Wax: 1.60

10. CONCLUSION

With global consumption of plastic and other petroleum products rising each year, there is an urgent need for a solution to both plastic waste and carbon emissions problems. Due to the capabilities of chemical recycling, and advancements in microreactor technology, a new opportunity has presented itself, for simultaneous solution to both problems.. The HEMP design allows for the conversion of mixed plastic waste into valuable industrial products using nuclear power, making the entire system carbon neutral. After outlining the requirements for running several industrial processes at once, a simple mass and energy balance was used to demonstrate the feasibility of this system. This low-level conceptual design is an example of how nuclear technology can act as a centralized source of power for applications requiring high quality process heat and having high electrical demands.

APPENDIX

A.1.

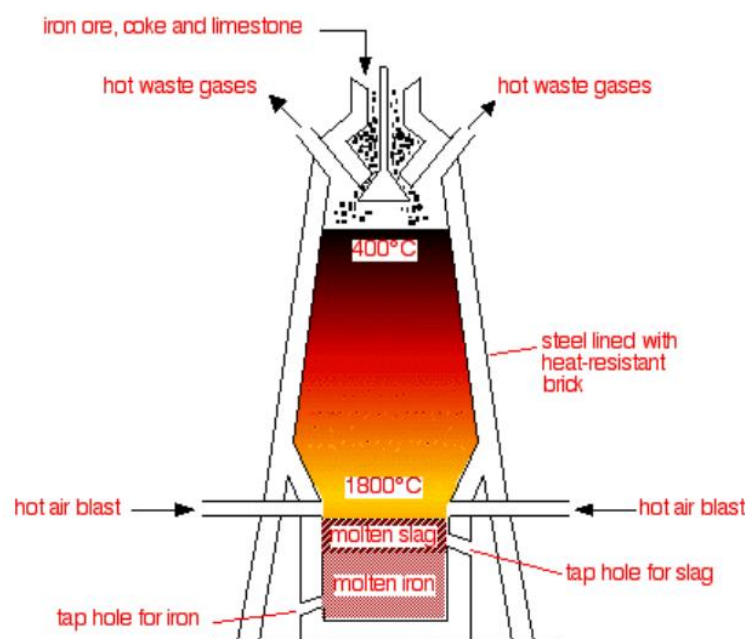
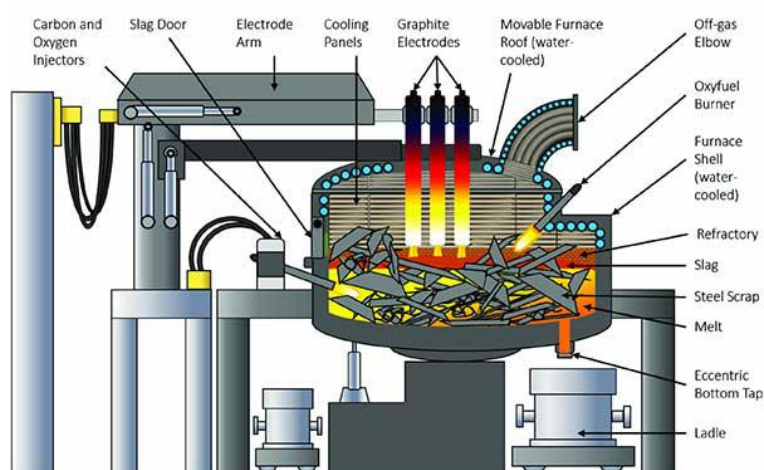


Diagram of a typical blast furnace used in an iron production process [15]

A.2.



Electric arc furnace (EAF) used to convert metal scrap and molten iron into fresh steel [15]

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CITE AN ARTICLE

Bresnahan M., Utgikar V., Piyush Sabharwall P. (2026). Nuclear Power for The Production of Industrial Products From Mixed Plastic Waste. *International Journal of Engineering Sciences & Research Technology*, 15(6), 14-35
<https://doi.org/10.64149/j.ijesrt.15.6.14-35>

