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Process Flow Sheet Synthesis: Reaching Targets for Idealized Coal Gasification

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The gasification of coal is a process that has been commonly used to produce a mixture of gases containing primarily carbon dioxide and hydrogen, called syngas. This syngas is used as an intermediate in the production of many chemicals such as ammonia, synthetic hydrocarbons, and methanol (to name a few). Coal gasification has a reputation for being "dirty" in terms of its emissions in comparison with other syngas creation technologies, such as methane reforming. However, there is remarkably little information on what the "best case" for coal gasification could actually be and how existing process perform relative to that "best case." The goal of this article is to formulate a preliminary and conceptual flow sheet for the gasification process; this flow sheet is not intended to be a finalized design or a definitive solution. It is intended to illustrate the method of setting and achieving design objectives and provide a basis of comparison for either new or existing processes. Thermodynamics can be used to describe any process, or system of processes. Of particular interest are the properties of enthalpy and Gibbs free energy. Using these two thermodynamic properties together as vectors on a diagram of free energy (ΔG) against enthalpy (ΔH), it becomes possible to develop better process flow sheets that combine the thermodynamics of chemical reactions and the dynamics of physical operations on a single diagram. This article will discuss the selection of the independent mass balances that best describe the process as a whole, then the choosing of design objectives, how these objectives might be achieved, and their implications for the process as a whole. Using these ideas one is able to show how to improve the carbon and operating efficiency of a gasification process, making the process more reversible. It was found that there will always be a price to pay for using coal as the feedstock for creating synthesis gas but there is room for improvement, most notably in how combustion is carried out and how energy is used internally within a process.
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Introduction

The impact of industries on the environment is rapidly becoming a major international concern. Increasing public awareness is evolving the public opinion toward an ever increasing demand for industries to minimize their environmental impact. At the same time, the requirements for industries to maximize their productivity and profitability are also increasing. Meeting these challenges is a complex undertaking that will require new and innovative approaches to process design.

Existing research can be divided into two levels:

1. Placing emphasis on the performance of an overall system and the interactions of individual units within the process with each other. This could be considered a "systems level" or "black box" approach.[1–3] The GH-space itself is on this level of process synthesis. The details of what occurs within each individual unit are the subject of the next level of synthesis. Such techniques include the pinch analysis[4] often used in heat integration, combined exergy and pinch analysis,[5] mixed integer nonlinear programming techniques,[6] and other energy saving techniques.[7] Also, on this level is optimizations of networks within a process, such as heat exchanger network optimization,[8–10] reactor sequence optimization,[11,12] and distillation sequencing.[13]
2. Placing emphasis on the functioning and optimization of individual unit processes. This could be considered "units level" and is far more details oriented than the previous level. The vast majority of research involves the optimizing of reactor performance through the use of specialized catalysts and reactor design.[14–17]

Something that the majority of the existing research has in common is that their techniques are typically applied on existing flow sheets or to meet specific performance targets, they give little information on what the best target actually is. They give a great deal of information of how existing processes might compare with each other but little on how well they compare to the best case.

The flows of heat and work within a process are major contributors to the environmental impact of a process,[18] excess work and heat will be lost as irreversibility in a

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system, if not recovered.[19] This loss can be described in terms of exergy, which is the measure of the departure of the system from equilibrium with its environment. It is, therefore, a measure of a systems ability to affect its environment; it shows the connection of losses in work and heat with environmental impact. By minimizing the loss of heat and work, the environmental impact can be minimized.

Typical procedures for process synthesis[20,21] are first to consider the desired products, then the selection of raw materials and the reactions that will give the desired products, which then lead to the selection of the required unit operations. Each of these units is then individually optimized, usually in sequential order. Only then are the flows of work and heat considered, after the flow sheet has been finalized.

This practice of sequential design may be simple and convenient to manage and will improve the operation of some of the individual units but the effect of that unit's new operation on all the other units in the process is often not considered until the time comes for those units to be optimized. This can lead to difficulties later on, with optimizing requiring modifications that are impossible to perform, further leading to a process that has a few efficient units but is overall inefficient.

Although the optimization of individual units and networks is extremely important, at this stage of a design even small changes in a process translate into significant cost.

It is proposed that an alternative approach to efficient process optimization and design is first to consider how all the process units could integrate with one another from the earliest stages of design, before any significant investment has been made, or even before any flow sheet has even been finalized. This will facilitate the creation of a process that is efficient, in its entirety. Such a process could thus be considered the "best-case" performance target; which could then become the basis for the optimization of the individual units, the design of new facilities, or a guide to show where existing processes can best be improved.

This work is part of series of case studies into processes that have a reputation for poor performance in carbon dioxide emissions. The goal of these case studies is to investigate if it is possible for these processes to perform any better in that regard.

In this article, the thermodynamic properties enthalpy (ΔH) and Gibbs free-energy (ΔG) are applied to the gasification process as well as to other unit operations such as compression and separation, with the goal of creating a gasification system that is as close to reversible as possible, and therefore, having the minimal environmental impact achievable for a gasification process.

The technique described in this article allows processes to be constructed from fundamental thermodynamic principles and basic mass balances alone. Although many of these principles may appear to be simple, this is actually deceptive. The technique herein is actually a very subtle, but elegant, method for designing process layouts.

Enthalpy and Gibbs Free Energy: Heat and Work

The enthalpy, Gibbs free energy plane: The GH-space

In a previously published article,[22] it was proposed that the ΔG and ΔH of reactions and process units could be drawn on a plot of ΔG and ΔH, hereafter referred to as the GH-space.

ΔG and ΔH are both extensive properties that are calculated independently of one another. This is different from, for instance, pinch diagrams where an extensive property is plotted against intensive property. ΔS could be used instead of ΔG but the latter is used in this article due to its easier relation to work.

Others[23] have done similar work where plots of $T_0\Delta S$ and ΔH were used instead of ΔG. The goal of minimizing the loss of work in that approach would be to minimize the value of the change in entropy. This is likely to provide a very precise measure of process reversibility but does not provide easy access to the practical considerations of operating pressure, separations, mixing, among others. The approach of using the GH-space allows all simple unit operations or reactions, for which ΔG and ΔH can be calculated, to be easily represented on a single set of axes. ΔG and ΔH are also directly related to the work and heat requirements of a process, ΔG represents the work requirements of a process and ΔH represents the heat requirement.

This article will not go into the minutia of the calculations involved in using the GH-space to develop flow sheets. If the reader should desire such details, previous works are available.[24] Instead this article will present a flow sheet developed by the GH-space method and discuss its implications.

Heat, work, and reversibility: The Carnot Temperature

As shown in previous publications,[24] the heat requirements of a process will also meet the work requirements of a process at one specific temperature. This temperature has been termed the "Carnot Temperature."[25]

This temperature is given by Eq. 1

$$W=\Delta G_{\text{process}}=\Delta H_{\text{process}}\left(1-\frac{T_0}{T_{\text{carnot}}}\right) \tag{1}$$

where:

- W is the work requirement of the process
- $\Delta G_{\text{process}}$ is the Gibbs free energy of the process at T_0
- $\Delta H_{\text{process}}$ is the enthalpy of the process at T_0, which is equal to the process heat requirements
- T_0 is the ambient temperature
- T_{carnot} is the reversible Carnot Temperature

The primary result of Eq. 1 is that if the heat requirements of a process were met by operating the process at its Carnot Temperature, it would be reversible. Alternatively, if a process was run at its Carnot Temperature the work required by the process would flow with the heat of the process. This is to say, that the heat supplied to the process would also supply the work.

From a reversibility viewpoint, the Carnot Temperature is the ideal temperature at which a process should be run. The heat automatically carries the correct amount of work: in supplying the needed heat the needed work is also supplied. This is essentially "killing two birds with one stone." It equates with less process equipment being needed. The equipment that supplies the heat, supplies the work at the same time.

However; it is not always desirable, or even possible, to run a process at its Carnot Temperature. When a temperature other than the Carnot Temperature is used there will be a deficit, or excess, between the amount of work carried by the process heat and the amount of work actually required by the process. This difference will need to be covered by additional process equipment and adds to process complexity.

The Carnot Temperature can be used as measure of how reversible a certain process, or "independent mass balance" could be. Independent mass balances with reasonable Carnot Temperature have the potential to be run close to reversibly. Although independent mass balances with impractical Carnot Temperatures are unlikely to ever be reversible, this gives a very quick indication of where most of the inefficiencies of a process are likely to occur and where additional care should be taken.

Application to Gasification

The independent mass balances

In a previously published work, synthesis gas was produced by the methane steam reforming process.[24] In this article, let us consider an alternative feedstock, namely coal.

There are many reasons for using alternative feeds. The most common being: cost and availability. Other reasons include: production efficiency, safety, and environmental impact.

Whenever one attempts to use the GH-space, to synthesize a flow sheet, the first step is to define the "independent mass balances" of the process. The "independent mass balances" could be thought of as the reactions that will be occurring within the process, indeed the term "independent mass balance" and "reaction" can sometimes be used interchangeably. This is not always the case however, often an "independent mass balance" is actually the sum of many individual "reactions," the result of which is the "independent mass balance." It is for this reason that a distinction is being made.

First, let us define the primary feed materials to this coal gasification process:

- Coal. This will be considered to be pure carbon. Not entirely accurate but will suffice for this early design.
- Liquid water. This is water in its natural state. Steam will actually be needed in the process.
- Oxygen. This is actually available as air but will be considered as pure oxygen to begin.

For these simplified feed stocks, it is possible to identify the products produced by the gasification process:

- Hydrogen and carbon monoxide. This is the desired product of syngas. The goal of this design is not only to produce syngas but also to produce it in the desired ratio.
- Carbon dioxide. An undesired by-product that has environmental impact. One of the goals of the design will be to minimize the production of this product.

There will be seven primary species in the process: Coal (as carbon), liquid water, steam, oxygen, hydrogen, carbon monoxide, and carbon dioxide. There are three atomic elements in the process: C, H, and O. Thus, there will be four independent mass balances.

The independent mass balance that will produce the syngas product is

$$C(s)+H_2O(g) \rightarrow CO\,(g)+H_2(g) \quad \text{Gasification}$$

The heat and work requirements for gasification can be determined by performing an energy balance at standard conditions

$$\Delta H_{\text{gasification}} = 131.32\,\text{kW}$$

$$\Delta G_{\text{gasification}} = 91.37\,\text{kW}$$

This reaction is endothermic and work deficient; in other words, both heat and work must be added to the process. This means a source of heat and work is required in order for the gasification to proceed. The Carnot Temperature for gasification can be calculated from Eq. 1 as around 977 K. The 131.32 kW of heat required by gasification would also carry the work requirement of 91.37 kW if the gasification was carried out at 977 K. This Carnot Temperature is not completely unreasonable but could be considered to be somewhat low from an equilibrium viewpoint, as temperatures below approximately 1100 K have poor coal conversion at equilibrium. Although a low single pass conversion can be worked around, it will have significant consequences of potential separation devices and recycles that will come later.

A heat source is required to provide the required heat to the system. The most convenient source of heat would come from combusting some of the carbon feedstock (coal) in oxygen (or air) to produce the heat. This independent mass balance is

$$C(s)+O_2(g) \rightarrow CO_2(g) \quad \text{Combustion}$$

An energy balance at standard conditions gives

$$\Delta H_{\text{combustion}} = -393.51\,\text{kW}$$

$$\Delta G_{\text{combustion}} = -394.36\,\text{kW}$$

This reaction is exothermic and work producing. This will allow combustion to be used to provide the heat and work to gasification. The Carnot Temperature of combustion can be calculated to be close to −14,000 K. This temperature is clearly impossible to achieve. The heat of combustion could never carry the work of combustion. Combustion is irreversible by its very nature. Combustion is responsible for most of the irreversibility in processes. Burning a unit of coal via combustion will provide too much heat and work for gasifying a unit of coal. Having such an excess will cause the process to become irreversible if the excess work is not recovered or otherwise used. For this reason, no more combustion should be used than is absolutely necessary.

The ratio of carbon monoxide to hydrogen resulting from gasification is 1:1. It may be desired to produce a ratio of carbon monoxide and hydrogen that is not 1:1. Ratios of 1:2 are often favored by coal-to-liquids processes and methanol processes; and ratios of 1:3 are favored by ammonia processes. Higher or lower ratios may be desired depending on the intended application of the syngas.

To modify the ratio of carbon monoxide to hydrogen another reaction is required, namely the water-gas shift (WGS) reaction

$$CO(g)+H_2O(g) \leftrightarrow CO_2(g)+H_2(g) \quad \text{WGS}$$

The energy balance at standard conditions gives the heat and work requirements

$$\Delta H_{\text{WGS}} = -41.19\,\text{kW}$$

$$\Delta G_{\text{WGS}} = -28.59\,\text{kW}$$

This WGS reaction is also an exothermic work producer. Its Carnot Temperature can be determined to be 973 K. This temperature is not unreasonable; if possible the WGS mass balance should be carried out at this temperature. The primary factors that might prevent this are chemical equilibrium, ensuring the WGS mass balance is able to change the ratio of carbon monoxide to hydrogen by a large enough amount, and catalyst activity. It should be noted that consideration was given to the equilibrium of the WGS mass balance but catalyst performance is beyond the scope of this article.

Gasification and WGS use vapor phase water (steam) as a reactant. At ambient conditions liquid water is easily obtained. The phase change needs to be considered

$$H_2O(l) \rightarrow H_2O(g) \quad \text{Phase Change}$$

An energy balance at standard conditions gives

$$\Delta H_{\text{phase change}} = 44.01\,\text{kW}$$
$$\Delta G_{\text{phase change}} = 8.56\,\text{kW}$$

Phase change is an endothermic and work deficient process. The Carnot Temperature for this mass balance can be calculated to be 370 K. It is interesting to note that this temperature is equal to the boiling point of water at atmospheric pressure. When boiling water in a kettle (for instance) the water boils when the kettles element is supplying both the heat and the work required.

Four independent mass balances have been defined and it is clear that all four of them could be integrated to interact with one another. Using the GH-space, it will be possible to investigate how the reactions interact with one another and to make the process more reversible, despite the inherent irreversibility of combustion.

The design objectives

Now that the independent mass balances have been defined, the next step is to decide what the objectives for the flow sheet will be. This is a matter of stating what the purpose of the design is. These objectives can be almost anything; only the designer can decide exactly what the purpose of their flow sheet will be.

For this gasification process design, the objectives will be as follows:

1. Produce synthesis gas with a 1:2 carbon monoxide to hydrogen ratio while ensuring the carbon feed proceeds as close as possible to complete reaction.
2. Use no more combustion than is necessary to minimize the production of carbon dioxide. Depending on the perspective, this could be considered the most important objective.
3. Fully integrate the flows of heat and work within the process
4. Recover, or make use of, as much of any excess heat and work as possible
5. Scale the process mass balance to use 1 mol/s of carbon as feed. This is merely for ease of calculation. The final mass balance can be scaled to any desired flow.

Mathematical formulation of the design objectives

Vector Representation. Although it is desired to keep this article as succinct as possible, some discussion of the mathematical formulations is necessary. If more details are desired, previous works,[24] and the thesis[26] in particular cover the calculations in pedagogical detail.

The entire system can be represented by the following vector addition

$$\begin{pmatrix}\Delta H_{\text{process}}\\ \Delta G_{\text{process}}\end{pmatrix} - e_{\text{gasification}}\begin{pmatrix}\Delta H_{\text{gasification}}\\ \Delta G_{\text{gasification}}\end{pmatrix} + e_{\text{combustion}}\begin{pmatrix}\Delta H_{\text{combustion}}\\ \Delta G_{\text{combustion}}\end{pmatrix} + e_{\text{WGS}}\begin{pmatrix}\Delta H_{\text{WGS}}\\ \Delta G_{\text{WGS}}\end{pmatrix} + e_{\text{phase change}}\begin{pmatrix}\Delta H_{\text{phase change}}\\ \Delta G_{\text{phase change}}\end{pmatrix} + \left(\sum_{\Delta G_{\text{seperation}}}^{0}\right)$$

where:

- e_i is the extent of reaction i
- $\sum \Delta G_{\text{separation}}$ is the sum of the work requirements for any required separators. This assumes an ideal separation where enthalpy of separation is zero. Essentially, this is the opposite of an ideal mixing.

Applying Design Objective 1 with mass balances in carbon dioxide and hydrogen gives

$$N_{H_2} = 2N_{\text{CO}}$$
$$3e_{\text{gasification}} - e_{\text{WGS}} = 2(e_{\text{gasification}} + e_{\text{WGS}})$$
$$e_{\text{WGS}} = \frac{1}{3}e_{\text{gasification}}$$

Applying Design Objective 5 and a mass balance in carbon gives

$$e_{\text{combustion}} = 1 - e_{\text{gasification}}$$

A mass balance in water gives:

$$e_{\text{phase change}} = \frac{4}{3}e_{\text{gasification}}$$

Substituting into the vector addition gives

$$\begin{pmatrix}\Delta H_{\text{process}}\\ \Delta G_{\text{process}}\end{pmatrix} = e_{\text{gasification}}\begin{pmatrix}\Delta H_{\text{gasification}}\\ \Delta G_{\text{gasification}}\end{pmatrix} + (1 - e_{\text{gasification}})\begin{pmatrix}\Delta H_{\text{combustion}}\\ \Delta G_{\text{combustion}}\end{pmatrix} + \frac{e_{\text{gasification}}}{3}\begin{pmatrix}\Delta H_{\text{WGS}}\\ \Delta G_{\text{WGS}}\end{pmatrix} + \frac{4e_{\text{gasification}}}{3}\begin{pmatrix}\Delta H_{\text{phase change}}\\ \Delta G_{\text{phase change}}\end{pmatrix} + \left(\sum_{\Delta G_{\text{seperation}}}^{0}\right)$$

Thus, any value for the extent of the gasification mass balance can be chosen, which will allow the overall mass balance, work of separation, and overall energy balance to be determined. The question now becomes: which value of extent of gasification should be chosen?

The implication of Design Objectives 2 and 3 are

$$\Delta H_{\text{process}} = 0 \text{ AND/OR } \Delta G_{\text{process}} = 0$$

The best case scenario would be if both $\Delta H_{\text{process}}$ and $\Delta G_{\text{process}}$ could equal zero at the same time. However, this is a rare occurrence. It is far more typical to have one or the other as zero and attempt to deal with the consequences of the others value. The question then becomes, what is a sensible way to proceed?

It is known from the independent mass balances that combustion is the primary source of energy for the process; this means that any deficit in energy will need to be made up by combustion, which means that any value of $\Delta H_{\text{process}}$ greater than zero can be discarded. This means that a solution to the vector addition where $\Delta G_{\text{process}} = 0$ will only be valid if $\Delta H_{\text{process}} \leq 0$. If this is not the case then seeking a solution where $\Delta H_{\text{process}} = 0$ is more sensible.

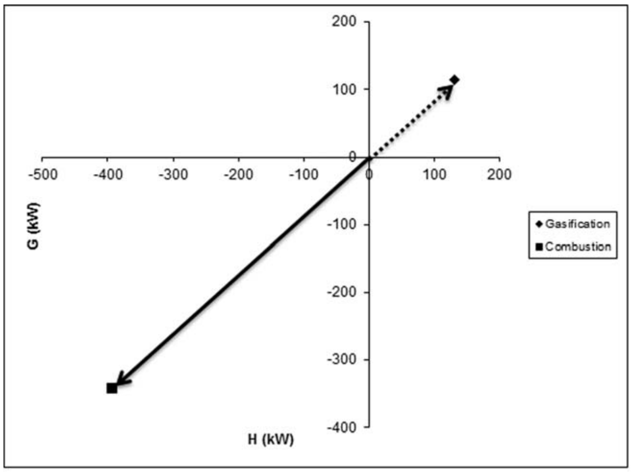

Figure 1. Vectors of the gasification and combustion independent balances on the GH-space.

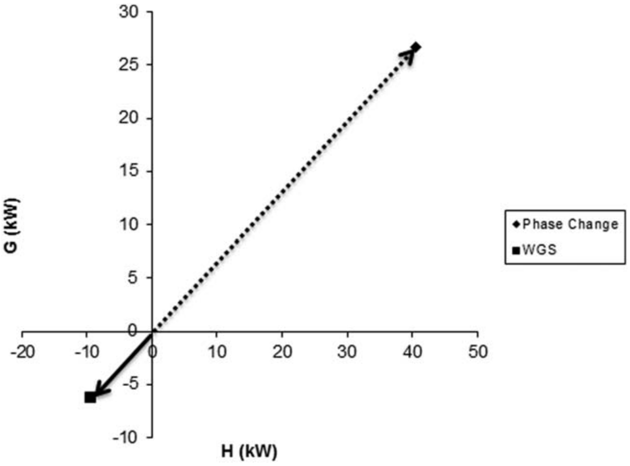

Figure 3. Vectors of the WGS and phase change independent balances on the GH-space.

The procedure could thus be summarized as follows: choose a value for extent of gasification such that $\Delta G_{\text{process}} = 0$. Check that $\Delta H_{\text{process}} \leq 0$, if it does then the solution could be useful. If it does not then choose the value for extent of gasification such that $\Delta H_{\text{process}} = 0$, the value of $\Delta G_{\text{process}}$ will then have to be dealt with.

The above vector addition can also be solved graphically by drawing the independent mass balances and separators on a plot of ΔH against ΔG. This is called the "GH-space." In this specific design, the vectors are very close to each other, making the GH-space difficult to read. For this reason, the vectors will be drawn onto separate figures.

To begin, the gasification and combustion balances can be considered part of a single process unit (or reactor), thus occurring at the same temperature. Thus, the gasification and combustion balances can be drawn onto the GH-space of Figure 1.

Figure 1 one shows the vector for the gasification balances and the combustion balances. These two balances lie on the same straight line as both of these balances are considered to be at the same temperature. These vectors can then be multiplied by their relationship to the extent of gasification and added together, the resultant vector of this operation can be called the "gasifier" as both of these independent balances can be considered as a single unit operation. This results in a resultant vector which is shown in Figure 2. Figure 2 also shows a vector which takes into account the effect of mixing; this mixing is assumed to be ideal and thus has no enthalpy.

The same exercise can then be repeated for the WGS and phase change balances. Figure 3 shows the vectors for the WGS and phase change balances. Just like in Figures 1 and 2, the vectors in Figure 3 can be added together and mixing can be accounted for. This yields Figure 4.

The two final resultant vectors in Figures 2 and 4 can then be drawn on Figure 5.

Figure 5 shows the two previously determined resultants and the final resultant of the entire process.

Figures 1–5 show that all the vectors lie nearly on top of one another; if they were all drawn onto a single set of axes, it would be difficult to glean meaning from the graphical solution. Fortunately, the previously derived mathematical formulation can be used to gain an analytical solution as well.

The vector addition can be solved with the value for the extent of gasification as 0.69; this allows the following results to be summarized.

Each independent mass balance for this gasification process is

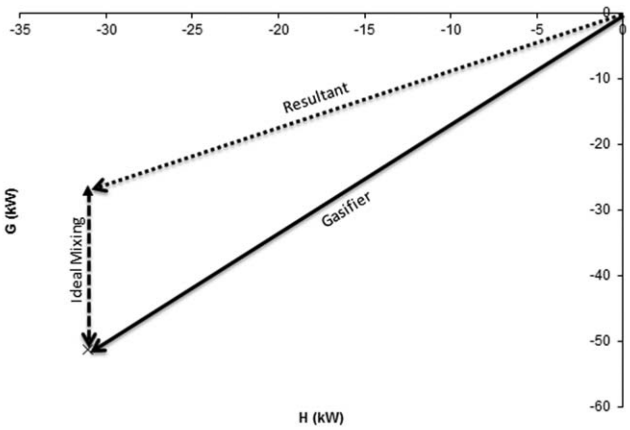

Figure 2. Resultant vector for the "gasifier" allowing for ideal mixing.

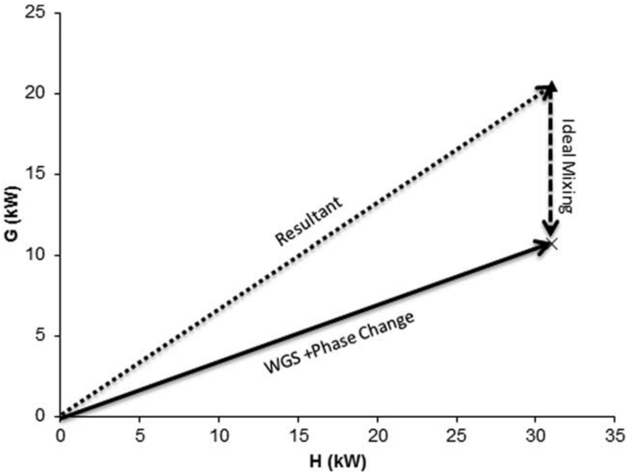

Figure 4. Resultant vector for the WGS and phase change balances with allowance for ideal mixing.

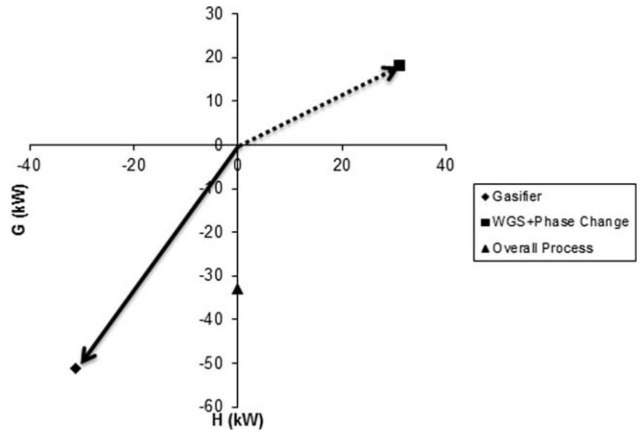

Figure 5. Resultant vectors from Figures 2 and 4 showing the process resultant.

$$0.69C(s)+0.69H_2O(g) \rightarrow 0.69H_2(g)+0.69CO(g)$$
$$0.31C(s)+0.31O_2(g) \rightarrow 0.31CO_2(g)$$
$$0.23CO(g)+0.23H_2O(g) \rightarrow 0.23H_2(g)+0.23CO_2(g)$$
$$0.92H_2O(l) \rightarrow 0.92H_2O(g)$$

The overall mass balance will then be the sum of all the independent mass balances

$$\begin{aligned}&1C(s)+0.31O_2(g)+0.92H_2O(l)\\&\quad\rightarrow 0.92H_2(g)+0.46CO(g)+0.54CO_2(g)\end{aligned}$$

The overall energy balance for the process is

$$\Delta H_{\text{process}}=0$$
$$\Delta G_{\text{process}}=-33.05\,kW$$

It is immediately clear that, in the case of this gasification process and a previously published work[24] using natural gas, there will always be a price to pay for using coal. The overall mass balance above shows that a coal gasification process will always emit carbon dioxide, even under idealized conditions.

In terms of process performance and environmental impact natural gas will always be a preferred feedstock, at least where a "hydrogen rich" syngas is desired. Where coal is the only option, or a "hydrogen lean" syngas can be used, this synthesis shows how a process might be designed to make it is as efficient as possible, minimizing what is lost and making best use of what is available.

Almost all of the irreversibility of the process comes from the combustion of the carbon feed to provide energy to process. Although the use of combustion to provide energy to such a process will remain necessary for some time it does show the importance of making the best use of what is being combusted and not burning any more than is absolutely necessary. It also shows that there may be significant opportunity in investigating alternative energy sources for processes, such as: alternative reactions, combustion of another feed materials or even nuclear reactions, or renewable energy sources.

Work Recovery: Dealing with ΔG. As can be seen; for this specific design, with these specific design objectives, when $\Delta H_{\text{process}}=0$ $\Delta G_{\text{process}}$ has a negative value. In other process designs, or even a similar design with different design objectives, it is possible to instead have a positive value for $\Delta G_{\text{process}}$. For the sake of brevity, this section will deal with how to deal with this negative value.

The implication of a negative value for $\Delta G_{\text{process}}$ is that there is work available within the process that could potentially be recovered as real work. If the work is not recovered it will simply be lost to the surroundings. Once this work enters the surroundings it will do something to the surroundings, which has environmental impact implications.

The first method to deal with a negative value of $\Delta G_{\text{process}}$ is to do nothing, just let the work be lost. This is typically done when the cost of recovering the work is too prohibitive. For instance, recovering the work might require running and maintaining turbines and electrical generators. This could potentially only make financial sense at very large scales.

Consider, however, that in this design any number of turbines and compressors can be used to recover this work. Another method to recover this work is by using a compressor-turbine system: if more gas leaves the process than enters the process; then the amount of work recovered by the turbines, at the exit of the process, will be greater than the amount of work used by the compressors, at the entrance of the process. The net effect will be work recovery. If the opposite were true then the net effect would be work input.

Inspection of the above results shows that more gas does leave the process than enter it. Thus, a compressor-turbine system could be used to potentially recover the work. The next step would then be to determine what pressure would allow recovery of the process work.

Complete recovery of all the excess work would require almost 4000 bar, this is clearly unworkable. However, it is still possible to operate at a more reasonable pressure and recover at least some of the excess work. At 75 bar, 52% of the total available work can be recovered. This is shown on the GH-space as Figure 6.

Figure 6 shows the recovery of work by the compressor-turbine system as a vector that pushes the resultant of the overall process toward $\Delta G=0$. Ideally, the goal would be for this "work recovery vector" to take the overall process all the way to the origin of the GH-space. As already stated, this would be impossible from a practicality viewpoint.

By taking the pressure higher, more of the available work could be recovered. A lower pressure would recover less of the available work. The operating pressure used has implications on the mass balance, as pressure will affect the maximum equilibrium conversion. Operating pressure also affects

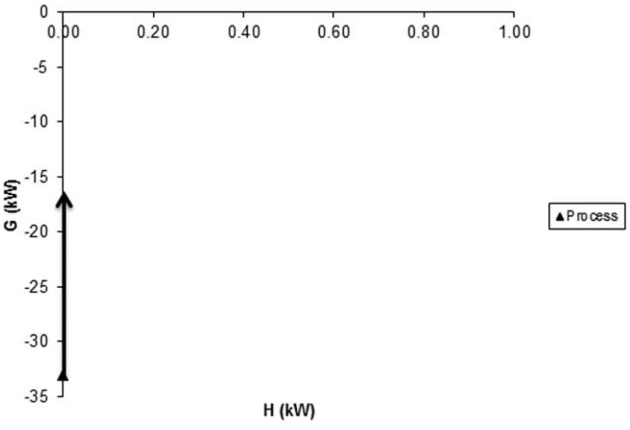

Figure 6. Final process resultant after work recovery.

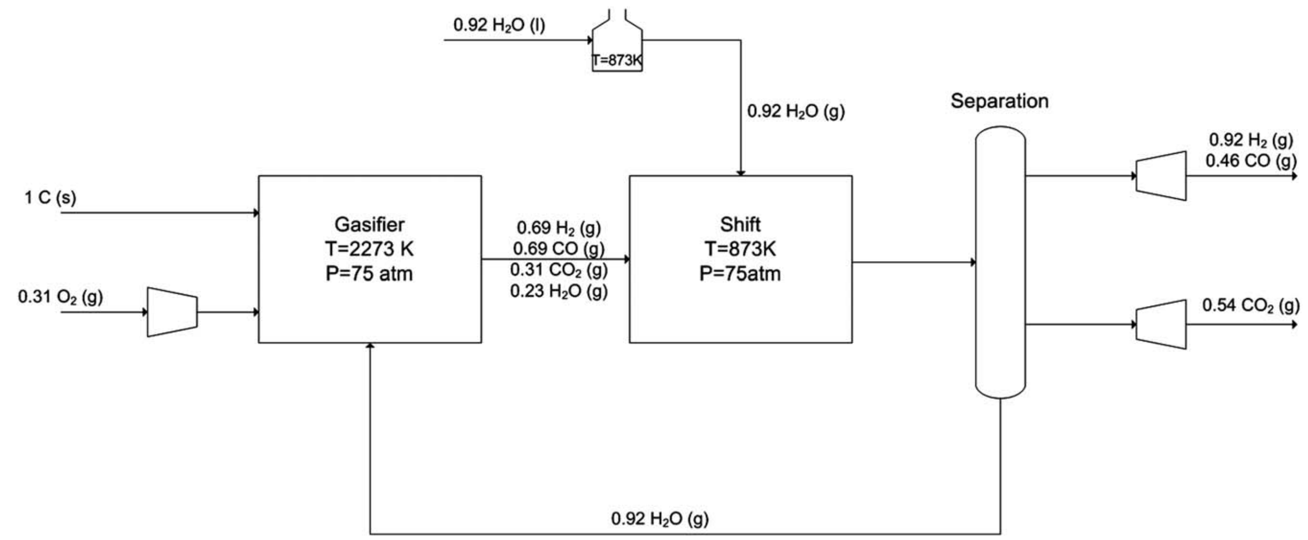

Figure 7. Mass flows for a target gasification process.

the materials and cost of construction. Thus, for this design, choosing an operating pressure would be a balance between how much of the work is worth recovering (if any), conversion, and cost.

It should be noted that by recovering the $\Delta G_{\text{process}}$, the $\Delta H_{\text{process}}$ will no longer be zero. $\Delta H_{\text{process}}$ will instead be equal to the amount of real work recovered. This is essentially transforming "virtual" work into "real" work.

An idealized coal gasification process

With the results discussed in the previous sections, it is possible to construct an ideal flow sheet, Figure 7.

Figure 7 shows the integrated compressor-turbine system connected to the processes gas streams. Because the gasification and combustion independent mass balances both use carbon as a feed, they were combined into a single unit which has been called the "gasifier." The oxygen and recycled water were then cofed into the gasifier. The WGS independent mass balance was then carried out in a separate unit, called "shift." The conditions of this unit were chosen to ensure that the ratio of carbon monoxide to hydrogen is the desired 1 to 2. The separator then recovers the syngas product while removing carbon dioxide and separating the water for recycling. It should be noted that, it may not be necessary or practical to remove the carbon dioxide from the syngas.

The carbon efficiency for this gasification process is 46%. This is the highest carbon efficiency attainable by the process with these design objectives. This is significantly lower than what is potentially attainable by natural gas processes. It also shows that more carbon dioxide is produced by coal gasification than syngas product. A noticeable amount of the carbon dioxide comes from increasing the hydrogen content of the syngas, if the intended application for the syngas can accept a hydrogen lean syngas the carbon efficiency could potentially go as high as 69%. An improvement but when compared to the 104%[24] that could be achieved using methane steam reforming is still low.

Design Objective 1. Inspection of Figure 7 and the overall mass balance shows that the process produces the syngas product in the desired ratio of 1:2 carbon monoxide to hydrogen.

The choice of the gasifier temperature, 2300 K, was chosen to ensure that the equilibrium of the gasfication mass balance favored the products and reacted the carbon feed to near completion. Alternatively, additional water could have been fed to the process to push the gasifier equilibrium further to the products. Doing that has implications for Objective 3 but may be worth considering, as excess water also suppresses carbon deposition.

This very high temperature was also chosen to assist with Objective 4.

Design Objective 2. It was known when the independent mass balances were defined that combustion was inherently the most irreversible part of the entire process. To minimize any loses to irreversibility and minimize carbon dioxide emissions combustion had to be carefully controlled.

This is accomplished by carefully specifying the amount of oxygen being fed into the "gasifier" unit, 0.31 mol/s. By precisely specifying this oxygen flow, no more combustion than is necessary will be allowed to occur. This corresponds to making the overall process adiabatic. In addition, the specification of the oxygen flow also has impact on design Objectives 3 and 4. By fully integrating the flows of heat and work and ensuring that the process is overall adiabatic, no more combustion will be needed than what is necessary and there will be no excess heat that needs to be recovered.

Design Objective 3. As discussed, specifying the flow of oxygen into the process had a significant contribution to achieving Objective 3. However, combustion was not the only producer of heat and work and gasification was not the only consumer of heat and work. Figure 8 (note that dashed lines are heat flows and dotted lines are work flows) shows how all the process units were integrated to assist with Objective 2.

This process accomplished the integration of its heat and work flows by the use of "heat engine" units. What these units might actually be, or look like, is a matter for further research. Heat engines and heat pumps do exist in reality but they contain their own inefficiencies and have never been applied in the manner suggested by this article. Further work is required to investigate how these integrations of heat and work could be carried out in a more practical manner and the costs involved. Such an investigation would constitute a publication on its own. Although this would be a worthwhile

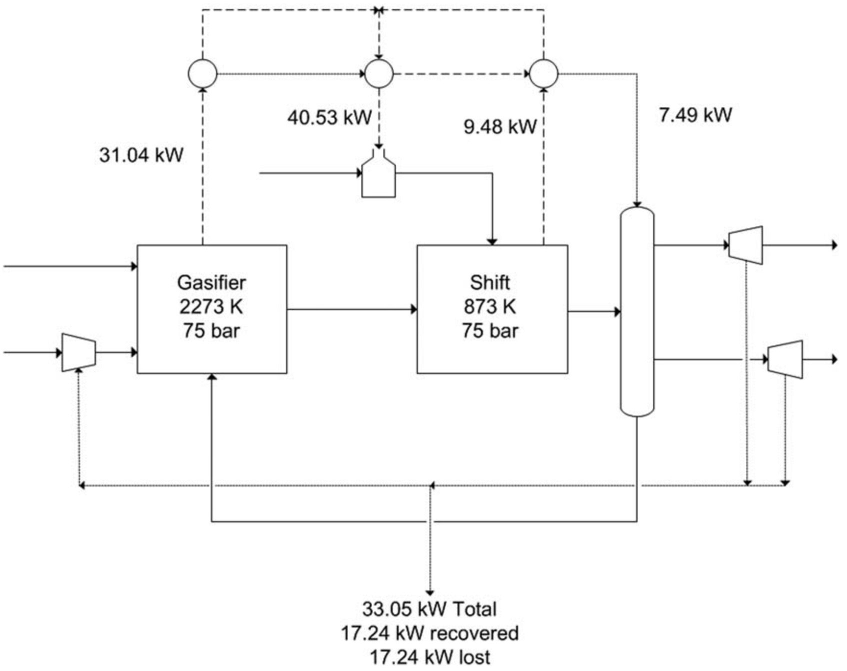

Figure 8. Heat (dashed lines) and Work (dotted lines) flows for a target gasification process.

exercise, it is well outside the intended scope of this research.

Design Objective 4. Once again Objective 2 had a noticeable impact on achieving Objective 4. By making the process overall adiabatic, it was ensured that all the heat of the process was used internally by the process and thus there was no excess heat to be recovered.

However, by ensuring that the heat of the process was used internally it was not possible to also ensure that all the work of the process would be used internally. Thus, there was an excess work within the process that needed to be recovered.

This recovery was accomplished by the use of a compression and turbine system. The pressure used by the process was chosen such that the work generated by the turbines could both be used to run the compressors of the process and recover the excess work of the process itself. However, it should be noted that it was not practical to recover all the excess work due to very high pressures being required.

Only very high operating temperatures and pressures could potentially allow complete recovery of the excess work but these conditions could very well prove impractical. Indeed 2300 K could already be considered impractical and is on the bounds of plasma processes, which do not operate a very high pressure.

The proportion of excess work that is not recovered will be lost to the surroundings as irreversibilities and has implications on environmental impact. It is for the designers to decide how much loss of excess work is acceptable given their unique requirements and objectives.

Conclusions

In this article, a process flow sheet was presented which attempted to accomplish a specific set of goals, using the "GH-space" technique presented in previous works. It should be noted that the purpose of this technique is not to present a finalized process flow sheet but to present an ideal or a "target." The gasification process presented in this article is not what every gasification process should be by any means. It is merely the thermodynamic ideal for the objectives it was designed to achieve.

For the objectives described in the article, the flow sheet presented represents the "best-case" scenario. No real process could ever hope to match its performance. Knowing what the ideal actually is, however, creates a valuable basis for comparison. It seems likely that the actual implementation of the processes presented using the GH-space technique will not perform well from a capital expenditure point of view, requiring the construction of possibly unique heat engines and heat pumps along with an associated utility system, in addition to all the other units and utilities typically required by processes.

One of the strengths of the GH-space is that it allows the construction of process flow sheets from basic thermodynamic principles. Even comparatively inexperienced designers can use the simple, yet elegant, principles of the GH-space to formulate flow sheets that are as thermodynamically efficient and environmentally friendly as possible while also allowing any previous knowledge or experience to be integrated into the technique almost seamlessly.

In the discussion of the "Design Objectives," it became clear that the Objectives are not entirely independent of each other. Decisions taken to achieve one objective have implications on achieving the other objectives. This is one of the biggest problems facing typical sequential design methodologies. However, sequential design methodologies are comparatively simple to manage. Managing a team of designers using this GH-space method would require either an uncommonly skilled manager or the development of an entirely new management method. Although this is well outside the ambit of this article it is a matter worth giving some thought to.

All the design objectives can be achieved simultaneously rather than consecutively. This is perhaps the greatest strength of the GH-space, the capability to allow all the interactions in a design to be taken into account simultaneously and presented in an analytical (or graphical) manner at the very earliest stages of a design. It is at the earliest stages of a design, before much investment in time and money has been made, that the greatest opportunity for change and innovation is present.

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