

COAL BASED POLYGENERATION WITH CO₂ CAPTURE FOR POWER AND UREA PRODUCTION - POSSIBLE IMPROVED SCHEMES

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ABSTRACT

Polygeneration is the process of combining multiple utility outputs with one or more input(s) in a single integrated unit. The integration is towards a higher energy efficiency as well as environment friendliness. Fossil fuels, specifically coal, will continue as the prime source of energy for a considerable future. However Carbon-dioxide emissions from coal based plants has serious 'climate change' effects. In this paper the Carbon-dioxide produced from the coal is utilized for production of urea in several well integrated and power-dominated polygeneration schemes. Power generation is by combined gas-steam power cycle. Other utilities of the polygeneration are utility heat and in some cases production of sulphuric acid. The utility heat obtained during urea production is proposed to be utilized by either heating the feed water in the steam cycle or by producing process steam. Different alternative schemes with similar utility outputs are discussed for relative advantages and disadvantages. Conceptualized schemes are not only CO₂ neutral though using coal but also compact and efficient with respect to same utility outputs in separate plants. Schematics show that depending upon one upstream process (say, gasification), downstream processes (say, carbon capture or urea production) may be changed according to their compatibility and state points may also vary for individual processes. Different utilities will also vary accordingly.

Keywords: Coal-based polygeneration, carbon-dioxide capture, Power-dominated urea production, utility heat

1. INTRODUCTION

Fossil fuels are the major source of energy worldwide and this trend will continue for significant period in future [1]. Coal is the most widely used fossil fuel for large scale energy applications including power generation [2]. Unfortunately carbon-dioxide (CO₂) emission from the use of fossil fuels is the principal cause of climate change [3]. As the fossil fuels cannot be replaced with renewables in near future [1], development of efficient systems using fossil fuels is very urgent. Polygenerations are such systems with strong potential. These systems integrate multiple utilities in a single unit in an energy-efficient and economical way [4]. Several authors reported conceptualization and estimation of performance of different polygeneration units using fossil fuels [5], [6], [7], [8]. Developing coal based polygeneration matching with local needs is very important presently. In this paper conceptualization and detailed system layout for different possible polygeneration schemes with electric power, utility heat and production of urea with captured CO₂ from the plant are proposed. Sulphuric acid is another utility output for some of these schemes. Relative advantages and disadvantages of these schemes with similar outputs are discussed in this paper.

2. SYSTEM LAYOUT AND DESCRIPTION FOR POLYGENERATION SCHEMES

2.1 Process Description

In these polygeneration schemes coal has been used as the primary source of energy. After gasification of coal (either allotropic or autothermal), produced syngas is made to pass through different units for cleaning (viz., the particulate removal unit, desulphurization unit, etc.) for use in power generation and ammonia production. In these processes the purity requirement of the syngas is relatively high for the production of ammonia and urea [9]. CO₂ removed during this cleaning process is captured and further utilized for urea production. After cleaning, the syngas is utilized in two parts. The first part is used to produce power in gas turbine. The flue gas from the gas turbine is passed through the heat recovery steam generator (HRSG) to heat the feed water of the bottoming steam cycle. The other part of syngas, after cryogenic purification, methanation and in some cases the nitrogen-wash unit becomes suitable for ammonia production [10]. To produce ammonia (NH₃), stoichiometric pure hydrogen is compressed to a pressure and temperature of the order of 270 to 300 atmospheres and 500°C over activated iron oxide catalyst to produce NH₃ [11].

Nitrogen is obtained either from the Air Separating Unit (ASU) or from the nitrogen-wash unit. CO₂ and NH₃ produced in the process are used as the raw materials to produce urea, a useful fertilizer. Liquid NH₃ and gaseous CO₂ and recycled materials (from the previous passes) meet in a heat exchanger reactor at a pressure of 14 MPa and 170° to 190° C to form ammonium carbamate (an intermediate to urea formation) and urea [12].

The general features of the polygeneration schemes are:

- a. Urea production.
- b. Heat produced during formation of ammonium carbamate is utilized in different ways as,
 - i. To heat the feed water in the steam cycle after the condenser. It thus saved the necessity of L.P. Heaters.
 - ii. Heat recovered through quenching during gasification is used to produce steam to generate electricity.
- c. Two stage electricity generation – from the gas turbine and the steam turbine in a combine power plant.

The syngas undergoes the shift reaction and enters the desulphurization unit, as shown in Fig. 1. The desulphurization is done in a rectisol unit due to the high impurity in the syngas caused by the fixed bed gasifier. In the rectisol unit the sulphur compounds in the syngas are removed in the form of Hydrogen sulphide (H₂S) which is then subsequently used as a raw material for sulphuric acid production. The CO₂ in the syngas is also separated in the rectisol unit. After the rectisol unit the syngas is made to enter the N₂ wash unit where the NO_x and the other nitrogenous compounds are removed and subsequently nitrogen is recovered. Moreover purge gas is produced as a byproduct in the nitrogen wash unit which is subsequently used as an additional fuel for combustion. The flue gas is utilized to heat the feed water of the bottoming steam cycle in the HRSG to produce steam for power generation in the steam turbine. The NH₃ produced is subsequently liquefied and reacted with gaseous CO₂ to produce ammonium carbamate and finally urea. The heat produced during ammonium carbamate formation used to produce process steam. This steam is used for water quenching of the syngas after gasification. Resulting superheated steam is utilized to produce power in steam turbine.

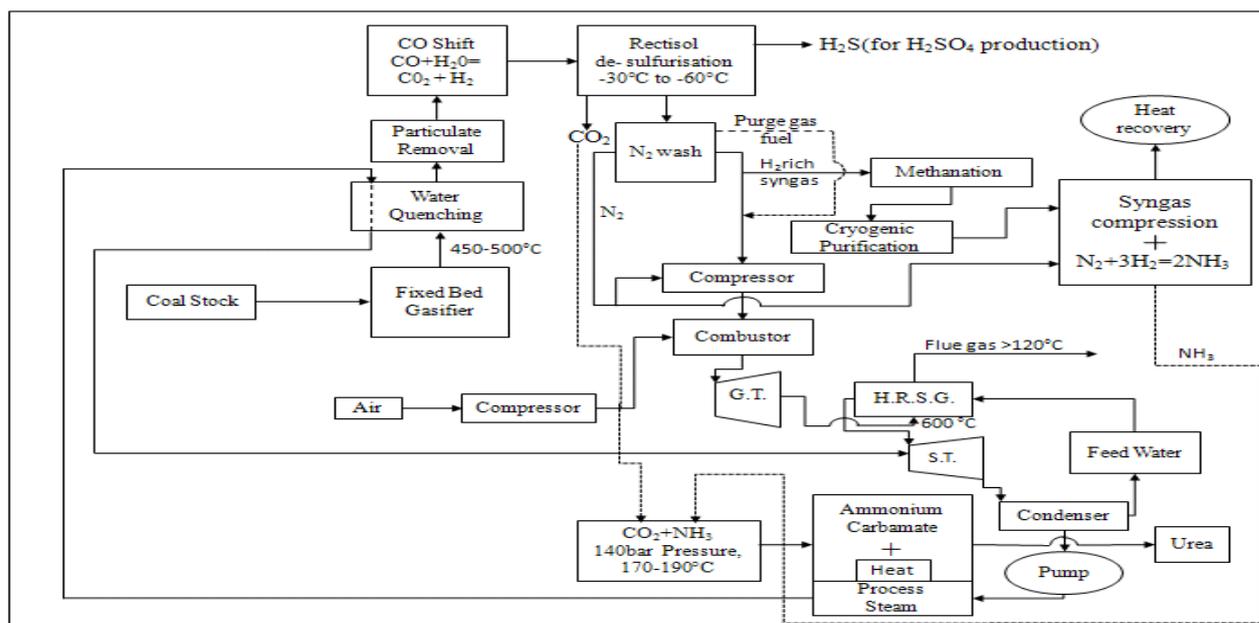


Fig.1: Detail layout of Scheme #1

- d. Utilization of the captured carbon dioxide thus providing a probable pathway for C.C.S.
 - e. Ammonia production.
 - f. Heat recovery during ammonia production.
- Six different schemes with similar outputs are discussed with detail plant layouts with relative advantages and disadvantages.

2.1.1 Scheme #1

In this scheme a fixed bed gasifier is used. The gasification is allotropic. The syngas temperature at the gasifier exit is 450°C [2].

After condensation the same amount of process water that is mixed with the main steam (MS) during power production, is bled out and again used to remove the heat of the ammonium carbamate formation, thus completing a cycle, as indicated in Fig. 1. A pumping arrangement is used to increase the pressure of the process water after being bled from the main condensate in order to raise the pressure to the requisite amount because of direct mixing of the two process steams.

The advantages of this scheme are:

- a. Fixed bed gasifier requires less energy for operation and is also of low cost.

- b. Maximum desulphurization (purest form of desulphurized syngas) obtained due to the use of Rectisol unit.
- c. ASU is not present, thus less energy is required.
- d. Maximum amount of sulphur is removed in the Rectisol unit as H₂S which can be utilized readily for sulphuric acid manufacture.
- e. CO₂ is obtained directly from the Rectisol unit and thus no separate CO₂ removal unit is required.
- f. No coal mill is required as pulverization is not required.
- g. Medium to high sulphur coal can be utilized.
- h. Due to high purity of syngas the exhaust gas from the HRSG can be at a lower temperature.

The disadvantages of this scheme are:

- a. Rectisol is a highly energy intensive process.
- b. Rectisol process operates at a very low temperature of around -30^o to -60^oC. It requires the carbon dioxide separated to be heated before urea production to bring it to the requisite temperature.
- c. The content of methane is as high as 10 to 15%. If the energy content of the tar is used, the cold gas efficiency may be around 90%. But such waste heat utilization in a raw gas cooler is difficult due to high tar fractions.

2.1.2 Scheme #2

In this scheme the gasifier used is fluidized bed type. So coal milling is not essential. The gasification is allotropic, with the syngas temperature from the gasifier ~ 900^oC [2]. In this scheme the subsequent desulphurization is carried out by MDEA as impurity in the syngas from the fluidized bed gasifier is relatively less. In this process, however, H₂S is not produced for sulphuric acid production. The CO₂ in the syngas could not be separated by MDEA and so a subsequent CO₂ removal unit is required for this purpose, as shown in Fig. 2. After the CO₂ removal unit the syngas enters the N₂ wash unit as before to recover N₂. The purge gas is utilized as before. The heat of formation of ammonium carbamate is used in this scheme for heating the feed water of the bottoming steam cycle in LP1 and LP2 stages. The steam exiting from the HRSG is utilized to quench the hot raw syngas as shown in Fig. 2. This superheated steam further, which then is used for power production in the steam turbine.

The advantages of this scheme are:

- a. Rectisol unit is not used. This saves a lot of energy and capital and operating cost. Moreover the very low temperatures are not required and thus CO₂ need not be heated to higher temperatures.
- b. No ASU means less energy requirement.

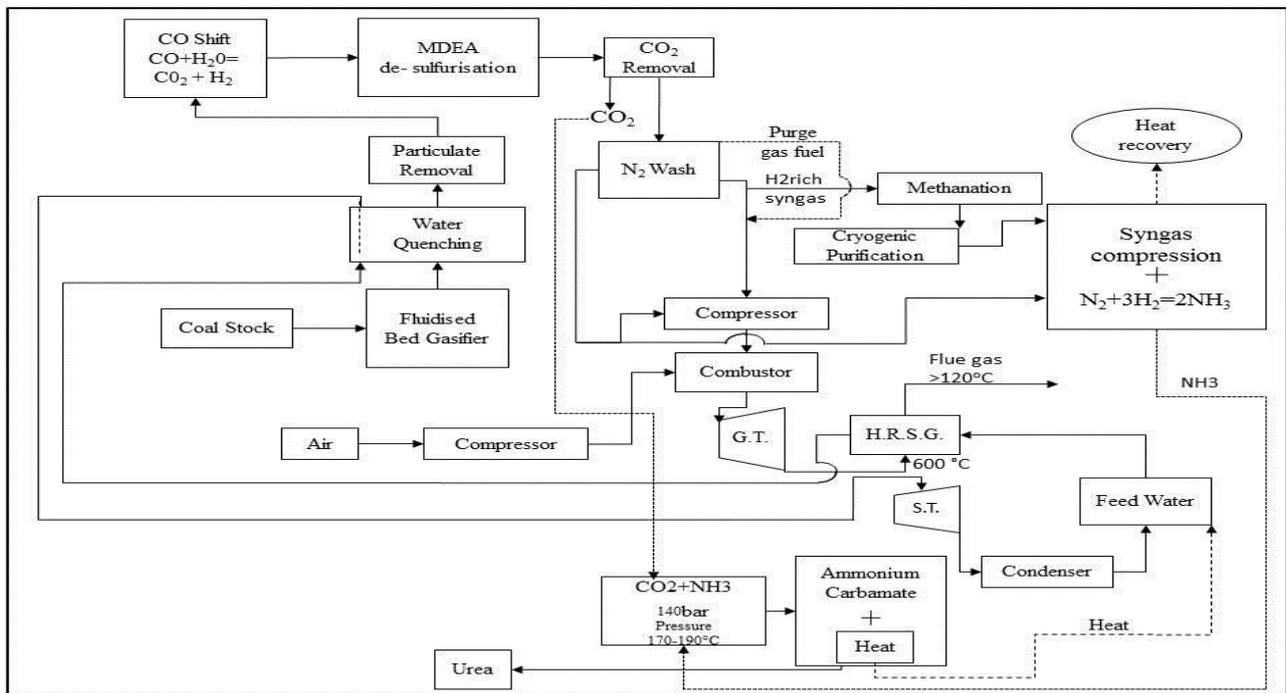


Fig. 2: Detail layout of Scheme #2

- d. High tar content of the product gas – as the product gas cools down, the tar condenses. The fraction of the tar components can amount to 25% with respect to the supplied fuel energy [2].
- e. The requirements of pumping arrangements increase the energy requirement and capital cost.

- c. Coal with higher ash content can be utilized [2].

The disadvantages of this scheme are:

- a. Additional CO₂ removal unit is required.
- b. The sulphur removal efficiency is less for MDEA than rectisol unit.

- c. The purity of syngas for this scheme being less than previous process the temperature of outlet gas from the HRSG cannot be too low. This decreases the efficiency of the system.
 - d. The fluidized bed process is particularly suited to reactive type coals, say lignite.
 - e. No sulphur is removed and sulphuric acid production is not feasible for this scheme.
 - f. Fluidized bed gasifier is costlier than the fixed bed type.
 - g. Due to the high temperature of the steam exiting from the HRSG, the temperature of the syngas after water quench will still be quite high.
- b. The Rectisol unit gives maximum purity (w.r.t. Sulphur) to the syngas for ammonia production.
 - c. The Rectisol unit being present in the ammonialoop the entire process does not become energy intensive.
- The disadvantages of this scheme are:
- a. Though we use a rectisol unit, the amount of H₂S obtained is quite small due to upstream desulphurization, and so sulphuric acid cannot be produced in this process.
 - b. As the syngas of this scheme is not too purified like scheme #1, the temperature of outlet gas from the HRSG cannot be too low. This decreases the efficiency of the system.

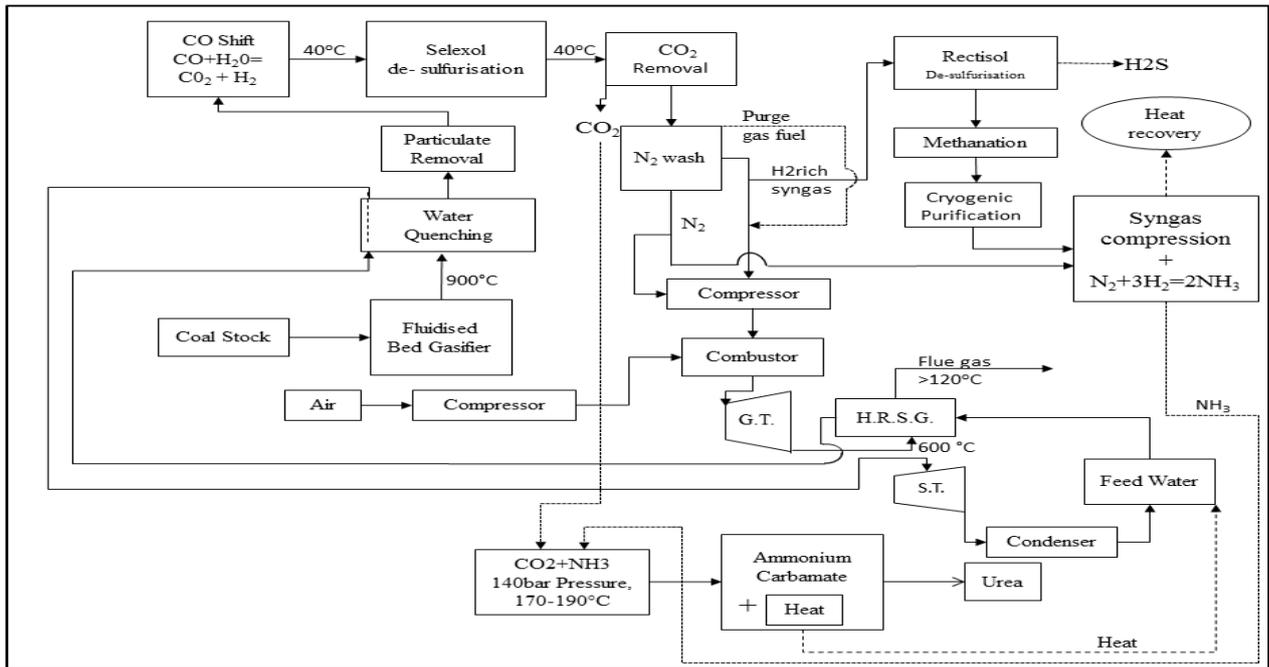


Fig. 3: Detail layout of Scheme #3

2.1.3 Scheme #3

In this scheme fluidized bed gasifier is used. The allotropic gasification produces syngas at ~ 900°C [2] at the gasifier exit. Selexol process is used for desulphurization which meets the syngas purity requirements of the gas turbine, but not up to the specifications for ammonia production. Downstream of the N₂ wash unit, the syngas for ammonia production is passed through a rectisol unit and subsequently underwent methanation and cryogenic purification to purify the syngas for the requirements for NH₃ production, as shown in Fig. 3. The N₂ from the N₂ wash unit is divided and utilized in combustion of syngas and NH₃ production. The heat from ammonium carbamate formation is also proposed to be utilized as in Scheme #2.

The advantages of this scheme are:

- a. The power requirement of the auxiliary units is lower in the power generating cycle. This reduces the cost of power.

- c. The fluidized bed process is particularly suited to reactive type coals like lignite.
- d. Extra CO₂ removal unit is required.
- e. Due to the high temperature of the steam exiting from the HRSG, the temperature of the syngas after water quench will still be quite high.

2.1.4 Scheme #4

In this scheme the gasifier used is of the entrained flow type. So coal mill is introduced in this scheme. Besides, an ASU is also used in this process. The O₂ from the ASU is used for the gasification. This makes the gasification an autothermal one, producing syngas at a temperature of about 1500°C [2]. This syngas is cooled by water quench. In this scheme the cleaning process used is the hot gas cleaning process [2].

When the temperature of the syngas is 550-600°C it is passed through the HCl, HF removal units and NH₃, HCN removal units, H₂S removal unit respectively, as shown in Fig. 4. Due to the high temperature of gasification, SO_x is essentially absent in the syngas[2]. So the H₂S present in the syngas is removed efficiently by using zinc titanate. The syngas is then further made to undergo CO shift and subsequently CO₂ is removed in a CO₂ removal unit. Now the N₂ separation from air in the ASU causes very low amount of N₂, mainly in the form of NH₃ and HCN to be present in the syngas and are removed during the cleaning process. The division of the syngas in two parts takes place right after the clean syngas comes out of the CO₂ removal unit. The N₂ obtained from the ASU is utilized for combustion of the syngas and a part is separated out for NH₃ production. The heat of ammonium carbamate formation is utilized to produce process steam. The subsequent processes are exactly same as in Scheme#1.

- f. The temperature of the syngas is never too low and it increases the efficiency of the cycle.
- g. Due to high purity of the syngas the temperature of the syngas at the HRSG exit can be quite low. This increases the efficiency of the system.
- h. Some heat can be removed by water quench.
- i. There is the potential to reduce capital and operating costs [14], [15].
- j. The power output increases with higher mass flow of steam.
- k. As the temperature of the syngas at the gasifier exit is much higher in entrained flow gasifier than the fixed bed gasifier the degree of superheat of the steam after quenching is much higher in this scheme. This increases the power output of the cycle. The relative disadvantages of this scheme are:

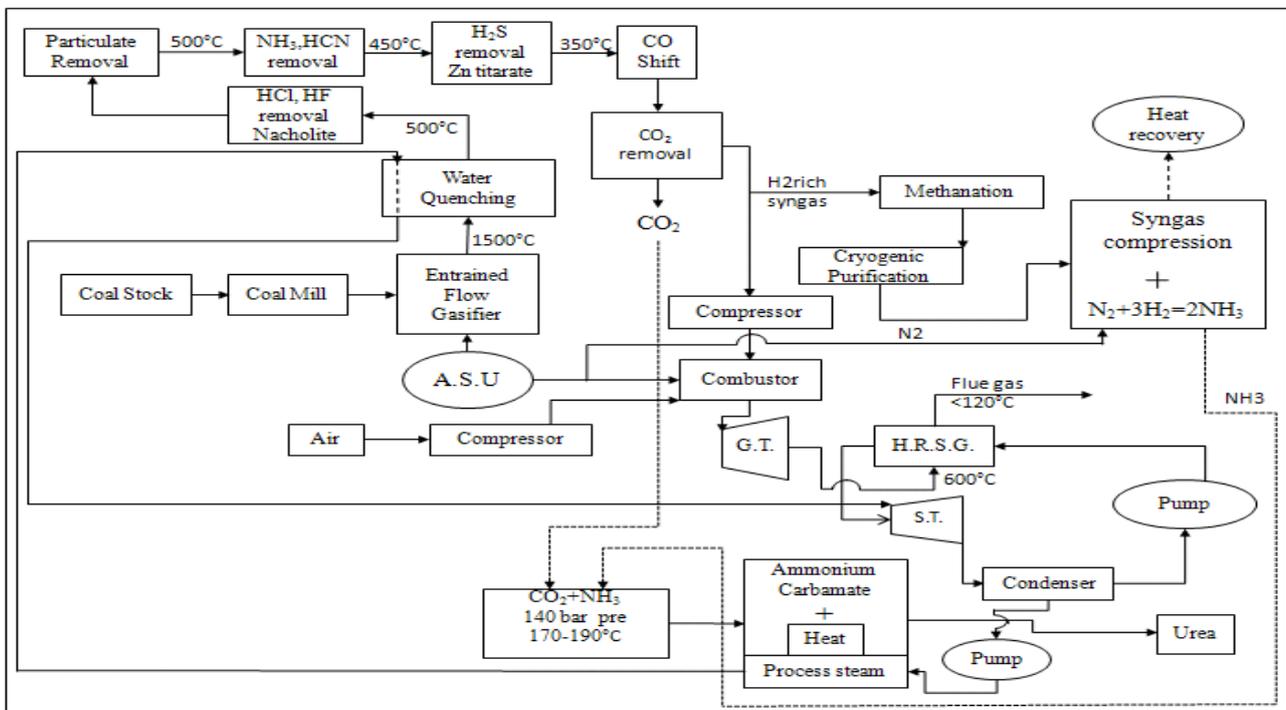


Fig. 4: Detail layout of Scheme #4

The relative advantages of this scheme are:

- a. Due to O₂ from ASU, very high temperature of the syngas from the entrained flow gasification is obtained.
- b. The ASU also provides pure and substantial amount of nitrogen.
- c. Due to the absence of Rectisol unit this scheme is a less energy intensive process.
- d. The sulphur removal rate is relatively high by zinc titanate [13].
- e. The residual nitrogenous compounds in the syngas are easily removed and so separate nitrogen wash unit is not required.

- a. It is an energy intensive process due to presence of the ASU.
- b. There is the need of extra pulverization for entrained flow gasifier.
- c. The exergy loss in water quench is significant due to the lower temperature of process steam.
- d. The requirements of pumping arrangements increase the energy requirement and capital cost.
- e. Sulphur removed is not as H₂S and not useful for sulphuric acid production.
- f. The efficiency of sulphur removal is less than the Rectisol units.
- g. Due to the lower temperature of the steam for water quenching, the temperature of the syngas after water quench will still be quite high.

2.1.5 Scheme# 5

In this scheme entrained flow gasifier is used and coal milling is essential. An ASU is also used. The produced syngas is at a temperature of about 1500°C. As shown in Fig. 5, the syngas is water quenched. The main difference of this process from the Scheme#4 is in syngas cooling, as shown in Fig. 5. CO₂ is removed in the rectisol unit. Besides, H₂S is obtained in substantial amount from the rectisol unit and so could be used for sulphuric acid production.

e. The process steam being directly mixed with the main steam greater mass flow rate in the steam cycle results greater power from steam cycle.

The relative disadvantages of this scheme are:

- a. It is a highly energy intensive process due to the rectisol, ASU units and the entrained flow gasifier.
- b. It increases the cost of both the power generated and the urea produced.

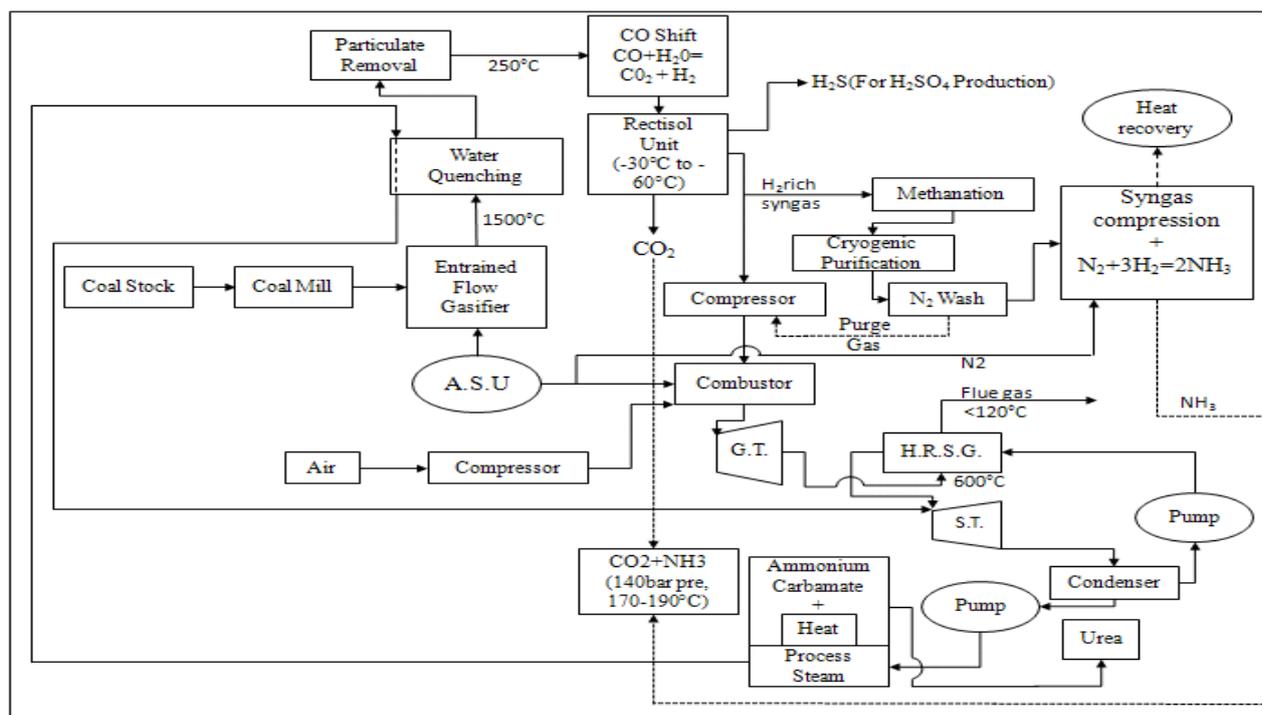


Fig. 5: Detail layout of Scheme #5

The N₂ from the ASU is used similar to the above scheme for combustion and NH₃ production. The syngas downstream of the rectisol unit is separated as before for power and NH₃ production, with the amount for NH₃ production being in correct proportion to N₂ as mentioned before. The syngas for ammonia production is further purified and is passed through an additional nitrogen wash unit in order to remove trace amounts of NH₃ and HCN in the syngas that causes poisoning of the catalyst during NH₃ production. The purge gas from the N₂ wash unit is used as before. The downstream processes are exactly same as Scheme#4.

The relative advantages of this scheme are:

- a. Due to high purity of the syngas the temperature of the syngas at the HRSG exit can be quite low.
- b. This scheme produces the cleanest syngas and thus is highly efficient.
- c. Sulphuric acid can be produced in this process.
- d. The carbon gets converted almost completely, the conversion rates amounting to more than 99%. Owing to the high reaction temperature, the entrained-flow process is also suitable for less reactive fuels [2].

- c. The exergy loss in water quench is more due to the lower temperature of process steam.
- d. The requirements of extra pumping arrangements increase the energy requirement and capital cost and reduces reliability of operation.

2.1.6 Scheme# 6

This scheme is almost similar to Scheme#5, as indicated by Fig. 6. The difference is that the steam exiting from the HRSG is utilized to quench the hot raw syngas. This superheated steam is further, which is then used for power production in the steam turbine. The difference also is in the utilization of the heat of ammonium carbamate formation during urea manufacture. Here, similar to Scheme#2 and Scheme#3, the heat is utilized to preheat the main condensate exiting from the condenser in the feed water cycle.

The relative advantages of this scheme are:

- a. The exergy destruction is much less due to the higher temperature of the steam used for quenching.
- b. Due to higher temperature of the steam, power production increases.

c. Due to the high temperature of the syngas in the entrained flow gasifier than the fluidized bed gasifier, the degree of superheat is much higher in this case.

The relative disadvantage of this scheme is: Due to the high temperature of the steam exiting from the HRSG, the temperature of the syngas after water quench will still be quite high.

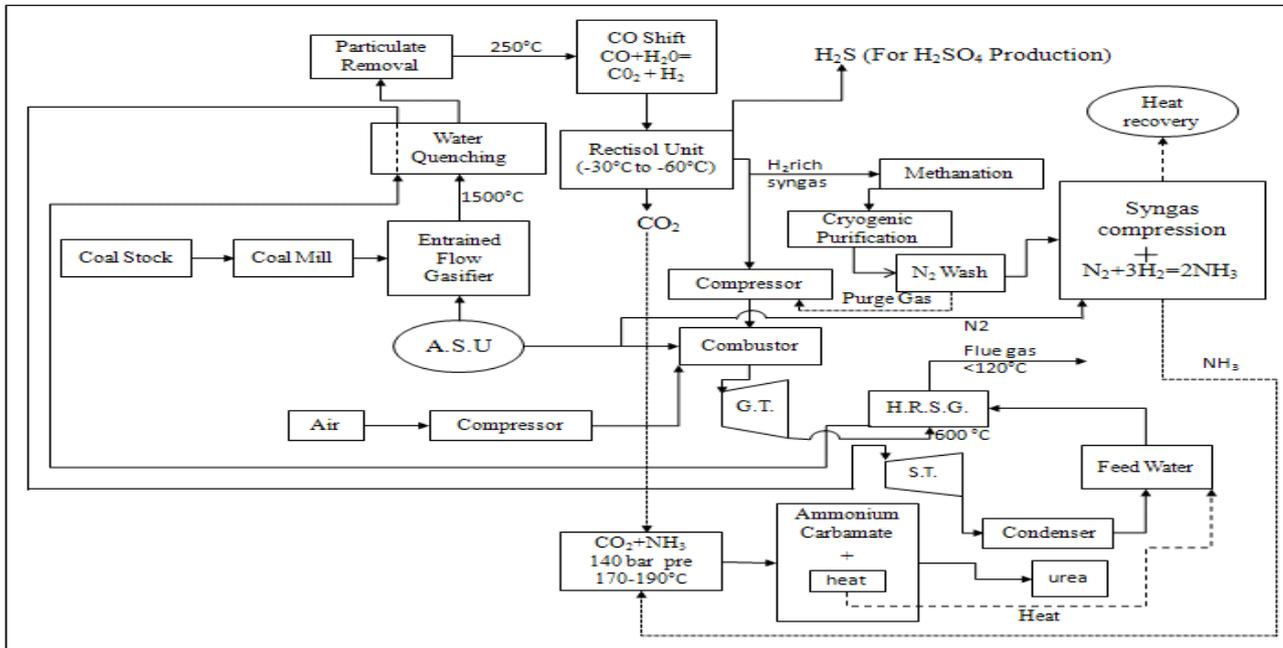


Fig. 6: Detail layout of Scheme #6

3. COMPARATIVE EVALUATION OF THE PROPOSED SCHEMES

Table 1: Comparison between schemes

Comparison No.	Comparison Type	Scheme1	Scheme2	Scheme3	Scheme4	Scheme5	Scheme6
1.	Gasification	Fixed Bed	Fluidised Bed	Fluidised Bed	Entrained Flow	Entrained Flow	Entrained Flow
2.	Coal Mill	No	No	No	Yes	Yes	Yes
3.	ASU	No	No	No	Yes	Yes	Yes
4.	FGD	Rectisol	MDEA	Selexol,Rectisol	Zinc Titanate	Rectisol	Rectisol
5.	Carbon Capture	Rectisol	MEA	MEA	MEA	Rectisol	Rectisol
6.	Power	Yes	Yes	Yes	Yes	Yes	Yes
7.	Urea	Yes	Yes	Yes	Yes	Yes	Yes
8.	Sulphuric Acid	Yes	No	No	No	Yes	Yes
9.	Water Quench at low/high temperature	Low	High	High	Low	Low	High
10.	FeedWater Heating	No	Yes	Yes	No	No	Yes

4.CONCLUSION

Polygeneration integrates output of multiple utilities from a single unit with one or more inputs for better energy efficiency and environment friendliness. Deciding optimum output utilities based on available input resources depends on local needs and available resources and involves creativity. In this paper six different polygeneration schemes have been conceptualized for electric power, urea production, utility heat as well as sulfuric acid for some schemes. Power is the main output and urea as well as other utilities are auxiliary outputs of the polygeneration system. So in these schemes power should not be sacrificed for urea and other utilities. The requirement of different auxiliary units and their working temperatures and pressures vary considerably for different schemes. Relative advantages and disadvantages are also discussed. Schemes are improved ones progressively however, generally with greater complexity.

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