

# Roadmap for Coal to Hydrogen production



# Report by Expert Committee



# Contents

1	ACKNOWLEDGEMENT	3
2	EXECUTIVE SUMMARY	5
3	BACKGROUND	8
4	HYDROGEN DEMAND SCENARIO	11
5	COAL AVAILABILITY	13
6	GASIFICATION TECHNOLOGIES	19
7	SYNGAS CLEANUP FOR DOWNSTREAM PROCESSING2	27
8	CARBON CAPTURE, UTILIZATION AND SEQUESTRATION IN COAL-BASED HYDROGEN	
SYS	STEMS: BRIEF OVERVIEW	14
9	HYDROGEN USAGE	47
9	0.1 Refinery & Fertilizer	47
9		
	9.2 Hydrogen based Iron & Steel Making	50
9	9.2 Hydrogen based Iron & Steel Making   9.3 Hydrogen in transport sector	50 65
9 10	9.2 Hydrogen based Iron & Steel Making   9.3 Hydrogen in transport sector	50 65 56
9 10 11	9.2 Hydrogen based Iron & Steel Making	50 65 56 70

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I thank various experts who made presentations to the expert committee and shared their valuable experience in the field of coal gasification, hydrogen production, and CCUS. I thank CSIR – CIMFR for presenting the possible technology options for conversion of Indian Coal to Hydrogen along with CCUS options. I thank Thermax for presenting and sharing its experience on technologies for Coal to Hydrogen production, Coal to chemicals and carbon capture and utilization. I thank Dastur & Co. for organizing a detailed presentation on the entire value chain of Coal to Hydrogen production, and CCUS and bringing together international experts like - GTI for Coal gasification technologies, Honeywell UoP for the technology on CO<sub>2</sub> capture, IIT Bombay and Advance Resources International Inc. for CO<sub>2</sub> utilisation for Enhanced Oil Recovery and Sequestration, Blue Planet for CO<sub>2</sub> to aggregates and Carbon Recycling International for CO<sub>2</sub> to Methanol.

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The committee members met several times over the last 7-8x months and deliberated in detail the possible routes for gasification of Indian Coal, downstream processing of syngas, syngas to Hydrogen, Carbon Capture Utilisation and Storage (CCUS) and usage of hydrogen at demand centres. This report is the successful culmination of the efforts of the expert committee members and hopefully will play an important role in the nation's endeavour to produce Hydrogen from Coal.

Dr. R.K. Malhotra Chairman, Expert Committee New Delhi

27<sup>th</sup> April 2022

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# 2 EXECUTIVE SUMMARY

According to the Hon'ble Prime Minister of India, Shri Narendra Modi, India's energy future has four pillars – Energy Access, energy efficiency, energy sustainability and energy security. Indian government believes in an integrated approach for energy planning to provide affordable energy to cater to its growing economy while reducing carbon emissions.

India is presently dependent for its energy supply primarily on coal and imports of oil and natural gas. In the recent couple of years, there has been significant emphasis on renewable energy derived from solar and wind power with a view not only to reduce the energy import dependence but also to meet the targets for reductions of greenhouse gas emissions.

Gasification is a process that converts carbonaceous materials, such as coal, petroleum coke, and biomass, into carbon monoxide and hydrogen rich gases ,called syn gases. Coal gasification as a power-generation technology, integrated gasification combined cycle (IGCC), is gaining popularity due to the ready global availability of the raw material (coal), as well as positive environmental issues associated with this technology over other combustion technologies. Syngas derived from gasification processes contains a significant amount of hydrogen ( $H_2$ ), which can be increased through water gas shift (WGS) and be readily separated into a pure  $H_2$  product meeting industry product quality standards.

However, the high ash content of Indian coal is a crucial barrier in the development of suitable technology. Gasification had commercial implementations in India mainly for the production of chemicals/fertilizers. Many of the coal-based gasifiers had ceased operations due to problems related to the quality of coal. Recently, Jindal Steel and Power Ltd. (JSPL) have set up first of its kind a coal gasification based direct reduced iron (DRI) and steel production plant at Angul, Odisha. However, they are also facing problems in their Lurgi Fixed Bed Dry Bottom (FBDB) gasifiers with coals beyond 30% ash. A joint venture of Coal India Ltd. (CIL), Gas Authority of India Ltd. (GAIL), Rashtriya Chemicals and Fertilisers Ltd. (RCF) and Fertiliser Corporation of India Ltd. (FCIL) is in the process to establish coal gasification plants for the production of ammonia and urea at Talcher, Odisha. However, addressing of various operational issues arising due to complex gasification behaviour with high ash Indian coal is the focal challenge for the successful implementation of the project.

The Committee had the benefit of presentations made by CSIR – CIMFR, Thermax & GTI on various technologies to produce the syngas. In addition, the committee had the benefit of learnings in the recent R&D gasification projects executed in the country for conversion of coal to methanol which were awarded by DST to IIT Delhi -Thermax, CIMFR and BHEL R&D. The EIL R&D also has experience on the gasification technology based on a pilot plant built up by them. In India, RIL had set up huge gasifiers for pet-coke gasification in which they are also producing hydrogen. IOCL R&D also has a patented technology based on which a pilot plant had been set up for co-gasification of coal, pet-coke and bio-mass. Such co-gasification of coal with bio-mass could possibly be also examined to introduce the green factor in production of hydrogen from coal.

Among the three types of the gasifier, the Entrained flow gasifier is a matured technology but yet to be tested for high ash coal. Fixed bed gasification technology is also a developed one but restricted up to the coal of 35% ash. Fluidized bed gasifier seems to be suitable for high ash coal but not widely commercialised at this stage. Though, coal ash content is a crucial parameter, other properties such as cold and hot crushing strength, gasification reactivity & surface area, Ash Fusion Temperature (AFT), slag viscosity and behaviour, ash composition, caking nature, rank, and petrographic characteristics, etc. are also imperative towards selection of type of gasifier. Based on the presentations made by various organizations, placed as Annexures of this report, the Expert Committee members are of the view that the fluidized bed gasifier appears to be more promising for Indian type of coal.

Raw syngas produced from the gasifier contains fine ash, char, slag and acid gases that need to be removed prior to the downstream processing. The various kind of technologies that are available have been included in this report. Depending upon the configuration of coal gasifier used and its operating conditions, typically 3 to 10% of the sulphur present in coal is converted to carbonyl sulphide which is converted to H<sub>2</sub>S by catalytic hydrolysis process and then H<sub>2</sub>S is removed by downstream acid gas removal unit. Further, we also need to remove mercury and other impurities through various processes which are commercially available and documented in this report.

In the commercial gasifiers, the syngas contains high amount of CO which can be converted to hydrogen by water gas shift reaction. This is an important reaction for extra hydrogen production from syngas. In this catalysed reaction, steam and CO react to produce H<sub>2</sub> and CO<sub>2</sub>. The CO<sub>2</sub> produced in this reaction will have to be captured and either stored underground or used in the enhanced oil recovery (EOR) or used for conversion to chemicals or used in beverages or fixed in the form of aggregates etc. Subsequently, after the removal of acid gas components from the shifted syngas it contains mainly hydrogen in which some impurities like CO, CO<sub>2</sub>, H<sub>2</sub>S/SO<sub>2</sub> may be present in minute amount which can be removed through the pressure swing adsorption as well as membrane technology which are commercially available.

Carbon capture utilization and storage (CCUS) technologies integration will enable conversion of brown hydrogen to blue hydrogen which may be more acceptable in the current scenario, though it may add to the cost to some extent. The blue hydrogen thus produced will still be cheaper than the green hydrogen in the current state of technologies for producing green hydrogen as has been reported in various studies.

Currently, India has hydrogen demand of 6.7 million tonnes per year of which the refineries and fertilizer plants are the largest consumers of hydrogen which is primarily being produced from natural gas as grey hydrogen. , though the refineries have plans to use the CO2 for conversion to chemicals and becoming net zero refineries. The demand is likely to increase to 11.7 million tonnes by 2030 and 28 million tonnes by 2050 as per the plans put in place particularly for Green Hydrogen. With plans for decarbonisation of various sectors, demand of hydrogen will increase. In case we could produce hydrogen from coal along with CCUS, we will be able to produce blue hydrogen (without carbon emissions) from indigenous sources rather than imported natural gas. It is felt by the Committee that we have ample opportunities for utilisation of hydrogen produced from coal in the refineries, fertilizer units, steel plants and transport sector etc. and coal to hydrogen may be one of the cheapest option and can produce hydrogen in the targeted price of USD 1-1.5 per kg without

CCUS. CCS costs are estimated to be of the order of 0.5/Kg, but the technologies are still to mature and this number would be refined in times to come.

It is suggested that the coal gasification units be established near the hydrogen demand centres and/or near the coal mines. The cost of transportation of hydrogen may be more at present and hence we need to examine the economics before deciding upon the locations of gasification plants. Alternatively, we could establish the plants closer to the natural gas grid so that hydrogen thus produced could be injected to some extent i.e. up to 18-20% into the natural gas pipelines, as to that extent of hydrogen injection in the natural gas pipelines may not need the modification of the gas pipelines. This will facilitate the utilization of hydrogen produced from coal in the industries currently using imported natural gas.

The Committee is of the view that India has an opportunity to produce hydrogen from domestic coal and we may aggressively pursue this option in our overall hydrogen ecosystem. We could set-up a couple of semi-commercial/demonstration gasification units for conversion of coal to hydrogen. We may also go for integrating the CCUS units along with gasification so that the blue hydrogen thus produced is more acceptable and should help in reducing overall CO2 emission. Further, to assess overall impact, a detailed LCA study can be undertaken. The gasification technologies may be selected based on assessment of the potential for eventual commercial upscaling and keeping the option of bio-mass co-gasification along with coal subject to availability of bio-mass in the close vicinity of such gasification units. The involvement of agency/agencies which have proven track record and are in a position to offer complete solution for delivering blue hydrogen from coal along with technologies preferably for conversion of CO2 to fuel/ chemicals/ aggregates which could add value to the plant rather than simply putting CO2 underground may be beneficial.

#### **3 BACKGROUND**

#### 3.1 Hydrogen as a future fuel

Hydrogen is a clean fuel. It is an energy carrier that can be used for a broad range of applications. Also, it could serve as a possible substitute to liquid and fossil fuels. At standard temperature and pressure, hydrogen is a nontoxic, non-metallic, odourless, tasteless, colourless, and highly combustible diatomic gas. It is also the most abundant element in the universe, making up more than 90% of all known matter. The abundance of hydrogen on earth, minimal environmental consequences of its use and the need to replace fossil fuels, makes it the ideal fuel of the future. Hydrogen energy involves the use of hydrogen and/or hydrogen-containing compounds to generate energy to be supplied to all practical uses needed with high energy efficiency, overwhelming environmental and social benefits, as well as economic competitiveness.

The production of hydrogen requires utilizing one of the primary energy sources – fossil fuels, nuclear, solar, wind, biomass, hydro, geothermal and urban waste resources. Once hydrogen is produced, it can be reacted with oxygen in a manner similar to gasoline combustion in an engine or used in a fuel cell to generate electric power. The electricity produced by a fuel cell can then be used to power electrical devices such as computers or an electric car. An important benefit is that, using hydrogen does not produce carbon monoxide or carbon dioxide. This makes it attractive because no greenhouse gases are produced

To produce hydrogen, it must be separated from the other elements in the molecules where it occurs. Hydrogen can be produced via thermochemical pathway, which uses a fossil fuel feedstock to produce hydrogen. This process must be paired with carbon capture utilization and storage (CCUS) to produce clean hydrogen. Steam methane reforming (SMR), which relies on natural gas as an input, and coal gasification are the mature technologies of this pathway. Gasification is a process that converts carbonaceous materials, such as coal, petroleum coke, or biomass, into carbon monoxide and hydrogen. Coal gasification as a power-generation technology is gaining popularity due to the ready global availability of the raw material (coal), as well as positive environmental issues associated with this technology over other combustion technologies. Syngas derived from gasification processes contains a significant amount of hydrogen ( $H_2$ ), which can be increased through water gas shift (WGS) and be readily separated into a pure  $H_2$  product meeting industry product quality standards.

There are several conventional  $H_2$  separation processes, but modern installations preferentially choose pressure swing adsorption (PSA), which is a well-proven technology offering high availability and low cost. PSA has the ability to produce high purity (99.9%) hydrogen at near feed pressure; however, relatively high  $H_2$  concentration in feed gases is required for its economics to remain favourable. New technologies are being developed to increase the efficiency and reduce the costs associated with  $H_2$  production from coal gasification.

Although most hydrogen is produced by steam reforming of natural gas (85% of world total), hydrogen production or co-production from syngas generated by liquid or solid gasification is commercially practiced as well. This is mainly based on gasification of residuals, heavy oil or petcoke from refinery operations, with the hydrogen helping to satisfy the in-house demand for hydrotreating, hydrodesulfurization, hydrogenation, and hydrocracking. In our country coal

gasification pilot plants for producing syn-gas have been completed, but there is currently no commercial coal gasification plant producing H<sub>2</sub> as a final product, though RIL has large units based on Pet-coke. Indian scenario in which the high cost of natural gas including import dependence occur concurrently with the low cost of domestic coal provide the impetus for the production of hydrogen from coal feedstock. Moreover, H<sub>2</sub> production technologies are gaining attention because hydrogen is predicted by some to be the energy carrier of the future, as it is extremely clean when reacted with oxygen (producing water) and has a high energy density by mass. Hydrogen can be used to feed fuel cells or combusted in a hydrogen turbine to generate electricity. Hydrogen could also power fuel cell vehicles. Although there are technical challenges to overcome, a clean coal gasifier to produce H<sub>2</sub> would be a key component of a hydrogen economy and hydrogen-based power generation as envisioned.

#### **ROLE OF HYDROGEN IN THE ENERGY TRANSITION**

Hydrogen is an excellent energy carrier, and produces zero emissions when used as a fuel. It can play a major role in the energy transformation, which span from the backbone of the energy system to the decarbonization of end-use applications:



Figure-3.1: Hydrogen for decarbonization of end use applications

#### Source: Hydrogen Council

- Enabling the renewable-energy system (1–3). By providing a means of long-term energy storage, hydrogen can enable a large-scale integration of renewable electricity into the energy system. It allows for the distribution of energy across regions and seasons and can serve as a buffer to increase energy-system resilience.
- Decarbonizing transportation (4). Today's transportation sector depends almost entirely on fossil fuels and creates more than 20 percent of all CO<sub>2</sub> emissions. Hydrogen-powered vehicles, with their high performance and the convenience offered by fast refuelling times,

can complement battery electric vehicles to achieve a broad decarbonization of transport segments.

- Decarbonizing industrial energy uses (5). In heavy industry, hydrogen can help decarbonize processes that are hard to electrify, in particular those requiring high-grade heat. Hydrogen can also be used in cogeneration units to generate heat and power for industrial uses.
- Decarbonizing building heat and power (6). In regions with existing natural-gas networks, hydrogen could piggyback on existing infrastructure and provide a cost-effective means of heating decarbonization.
- Providing clean feedstock for industry (7). Current uses of hydrogen as industry feedstock amounting to more than 55 million tons per year—could be fully decarbonized. Hydrogen could also be employed to produce cleaner chemicals and steel, by being used as a chemical feedstock in combination with captured carbon and by being used as a reducing agent for iron

# 4 HYDROGEN DEMAND SCENARIO

Currently, hydrogen is used mainly in refining (to remove sulphur from crude oils) and fertilizer production except small usages in other sectors like chemicals, textiles, and electronics. Total demand of hydrogen in 2021 was around 6.7 million tonnes, of which around 54% or 3.6 million tonnes was in petroleum refining, 3.0 million tonnes in fertilizer production and 0.1 million tonnes in Gas based DRI. This is, 'grey' hydrogen produced from fossil fuels, mainly from natural gas and partly from naphtha. It is expected that the demand of Hydrogen in India may increase to 11.7 million tonnes by 2030 with 6.8 million tonnes in Refinery, 4.6 million tonnes in Fertilizer and around 0.3 million tonnes in DRI Steel making. Looking to the future, hydrogen has a number of potential applications that could be significantly expanded across a range of end-use sectors from transportation to electric power to industry. For example, hydrogen can be used in fuel cells to power passenger and commercial vehicles, heavy-duty trucks, buses, trains, and waterborne vessels. It is projected that the demand can increase to around 28 Mt by 2050, driven by cost reductions in key technologies, as well as the growing imperative to decarbonize the energy system. Demand will continue to be largely focused in industry sectors, either expanding in existing sectors, such as fertilizers and refineries, or growing into new sectors, such as steel.

Beyond transportation, hydrogen can be used in fuel cells to generate electricity for backup power and/or distributed energy applications, and it can be blended into natural gas for use in a gas turbine to generate electricity. Hydrogen can also be stored for use in power generation to manage load in power systems when intermittent renewables are not available. The fact that hydrogen can be used in so many different applications across multiple sectors makes it a prime candidate for playing a substantial role in transitioning the energy system. The existing and future applications of Hydrogen along with likely cost of production are shown in Figure-4.1 below:



Figure-4.1: Applications and COP of Hydrogen

In the longer term, steel and high-temperature heat production offer vast potential for low emissions hydrogen demand growth. Assuming that the technological challenges that currently inhibit the widespread adoption of hydrogen in these areas can be overcome, the key challenges will be reducing costs and scaling up. MNRE in its presentation to NITI Aayog also identified multiple

usages of Hydrogen mainly as a feed stock in various Industrial applications covering Refining, Fertilizer and Steel and as an energy source in blending with natural gas, heavy duty transport fuel and energy storage. Accordingly, a 60 GW green hydrogen mission to produce around 5 million tonnes of hydrogen is being planned by MNRE. Thus, in future, the usages of the Hydrogen may increase manifold.

Global hydrogen demand was around 90 million tonnes  $H_2$  in 2020, having grown 50% since the turn of the millennium. Almost all this demand comes from refining and industrial uses. Annually, refineries consume close to 40 million tonnes  $H_2$  as feedstock and reagents or as a source of energy.

Today, majority of it is produced by Steam Methane Reforming (SMR) except in China where major demand is met by Coal based Hydrogen using Gasification technology. China 's hydrogen production in 2019-20 was estimated at 22 million tonnes (Mt) making it the world's largest producer. Roughly 14 Mt of hydrogen produced was from coal gasification, with additional hydrogen derived from coking, and under 4 Mt was produced via SMR. An estimated 1 Mt of hydrogen in the chlor-alkali industry was produced via electrolysis. However, in both the technologies i.e SMR and Coal gasification, large amount of CO<sub>2</sub> emissions is generated and that is a matter of concern across the globe. Since, production of green hydrogen produced using SMR or Coal gasification, efforts are being made to capture carbon and either store it underground or used for making different chemicals and feedstock. China is claiming to produce Hydrogen at a cost of USD 0.8-1.5 per kg through coal gasification against estimated cost of USD 3-4 per Kg with renewable energy. Thus, Coal based Hydrogen has large potential if clubbed with CCU or CCUS technology.

Fortunately, hydrogen can be produced in many different ways, which allows many options for meeting demands in relatively low-cost ways. Indeed, the least-cost option for hydrogen production may be different across different regions. To the extent this is the case, the principle of comparative advantage will play an important role in shaping how regions adopt hydrogen as an energy source and what technologies are chosen for production.

Since, India is having large reserves of Coal; this route can meet the major demand of Hydrogen of different sector in the country. The other countries like Australia, who is also having abundant availability of Coal is also exploring options of producing Hydrogen from Coal route as presently it appears to be the cheapest route of producing Hydrogen.

# 5 COAL AVAILABILITY

#### 5.1 Important coal properties in view of gasification

Coal choice is the least flexible factor considering economic, geographical and political reasons. Therefore, it is necessary to adopt a coal gasifier, which best suits to the properties of the coal to be processed. The important coal properties which affect the gasifier performance are proximate and ultimate analysis, cold and hot crushing strength, gasification reactivity & surface area, ash fusion temperature (AFT), slag behaviour, ash composition, caking index, rank and, petrographic characteristics, etc. These properties, as well as their relevance with the type of gasifier, are detailed below:

#### 5.1.1 Cold and hot crushing strength of coal particles

In the case of a moving bed gasifier, the regular and well-dispersed flow of gaseous reactants through the bed, as well as efficient mass and heat transfer between solids and gases, are primary requirements. To achieve this, it requires sufficient bed permeability which is controlled by the coal particle size distribution inside the bed. Further, the particle size distribution of the coal during its travel through drying, pyrolysis, gasification and combustion zones of the gasifier is controlled by cold and hot crushing strength of coal. Thus, cold and hot crushing strength of coal indicates a tendency towards thermal fragmentation as gasification phenomena progress inside the gasifier and thus also controls bed permeability. However, bed permeability is not an important property for the other two types of gasifiers. Therefore, cold as well as hot strength of coal in gasification atmosphere becomes an important parameter to make a decision about the suitability of any coal for moving bed gasifiers. In the present study, we have analysed cold as well as hot strength of coal, char after pyrolysis, and partially converted char/aggregates after gasification phenomena in the laboratory as discussed below. Based upon observed cold and hot strength values, the suitability of coal is decided towards the moving bed gasifier.

#### 5.1.2 Reactivity and Surface Area

Coal reactivity describes how fast the carbon matrix can be consumed under certain conditions. If a gasification process is operated in a temperature range, where the chemical reaction is the rate-controlling step, the reactivity has a significant effect on the process performance. This is especially true for fluid bed gasifiers, where the coal reactivity inevitably determines the carbon conversion which finally reflects in overall gasification efficiency. Further, gasifier sizing, design of gasifier components as well as selection of operating parameters also depend on the reactivity of coal. Reactivity has a significant influence on the degree of char recycle and on the volume of oxidant and gasifying agents required for the gasifier. Therefore, reactivity is considered as one of the important properties for selecting suitable gasifier for a particular coal. Entrained flow gasifiers operate at the highest temperatures among the three basic types of gasifiers. Therefore, entrained flow gasifiers can handle coal of any reactivity. In practice, reactive coals can be gasified at lower temperatures, and hence, at higher cold gas efficiency, whereas less reactive coals may need higher gasification temperatures in order to achieve adequate carbon conversions resulting in lower cold gas efficiency. On the other hand, the operating temperature of a

fluidized bed gasifier varies usually between 800-1050 °C. At the same time, residence time maintained is also low. Therefore, to achieve a high level of carbon conversion, the reactivity of the coal must be sufficiently high in fluidized bed gasifier.

In the case of moving bed gasifier, operating temperature is maintained in between entrained flow and fluidized bed gasifier and residence time is sufficiently high (15-60 minutes or even more). Therefore, it can gasify coal of any reactivity with an acceptable level of conversion. Gasification reactivity is determined by the thermo-gravimetric method as discussed below.

The surface area of coal is an important property towards the exposure of the coaly matter to gasifying agents during gasification phenomena. Higher is the surface area, higher may be the possibility for the contact between gaseous reactants and reactive sites present in the coal, which affects the gasification reactivity of coal. The surface area of coal is determined by gas adsorption technique.

#### 5.1.3 Ash content

Ash content is considered as one of the deciding coal properties for selecting suitable gasifier. Entrained flow gasifiers are usually recommended for coals with low ash content for both economic and technical reasons. If gasifier operating conditions are kept constant, an increase in coal ash content will lead to a decrease in overall efficiency and an increase in slag production and disposal. Whereas, high ash coal can be gasified efficiently in both moving (up to 30%) and fluidized bed gasifiers provided other required conditions are fulfilled. Fluidized bed gasifiers seem to be most suitable for high ash low-rank Indian coals.

#### 5.1.4 Ash composition, ash fusion temperature (AFT), slag viscosity/fluid point

The chemical composition of ash is an important parameter towards the suitability of coal for the particular gasifier. Both, ash fusion temperatures (AFT) and slag viscosity/fluid point depends on the chemical composition of ash. For the entrained flow gasifier, the operating temperature must be above AFT to maintain the slag in the Newtonian flow region. AFT and temperature of slag fluid point increase with SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio, whereas the decrease with alkali concentrations. Entrained flow gasifiers lined with a refractory are susceptible to some of the compounds present in slag (SiO<sub>2</sub>, CaO, iron oxides) which can penetrate deep into the refractory and eventually give rise to cracks that lead to material loss. In the case of the entrained flow gasifier, the AFT and slag fluid point temperature recommended for smooth slag tapping can be lowered by either adding flux (CaO) or blending with coal having low AFT.

In the fluidized bed, it is necessary to process coals with a higher ash fusion temperature than the operating temperature of the gasifier to avoid ash agglomeration. A higher percentage of iron and calcium, in coal ash as well as sodium silicates formed during gasification, is believed to be among the factors that can cause agglomeration and clinker formation in a fluidized bed and moving bed systems. Ash clinkering can also cause channel burning, pressure drop problems and unstable gasifier operation in moving bed gasifiers. Therefore, careful control of the gasifier operating temperature is required when processing coals with high alkali content. In the present study, ash composition is determined by X-Ray Fluorescence spectroscopy as discussed below. Also, we have analyzed initial deformation temperature (IDT), softening temperature (ST), hemispherical temperature (HT) and flow temperature (FT) of ash of the selected coal samples. Further, we have assessed fluid point temperature of slag to understand the performance of analyzed coals towards slag behaviour inside the entrained flow gasifier. This will help us to understand the suitability of coal for the entrained flow gasifier as well as a tendency towards agglomeration and clinker formation in the fluidized bed and moving bed gasifiers.

#### 5.1.5 Free swelling index and caking properties

The caking and swelling characteristics of coal can be described by the free swelling index. Caking of coal particles indicates softening or plastic property of coal when heated, which causes particles to melt together to form larger particles. In the fluidized bed gasifier, the probability of agglomerate formation increases with the increase in the free swelling index (caking property) of the coal. Therefore, caking coals are avoided in fluidized bed gasifiers.

On the other hand, the caking of coal within the moving bed gasifier can also cause pressure drop fluctuations and channel burning, resulting in unstable gasifier operations. Therefore, in moving bed gasifier too, highly caking coal is not desirable. However, to process caking coals, a stirrer connected to the coal plate distributor has been added to the slagging moving bed gasifier. It ensures that strongly caking coals are completely carbonized and converted to free-flowing solids that pass to the lower gasification bed. Entrained flow gasifier can gasify efficiently both caking and non-caking coals. In the present study, the free swelling index has been determined and discussed later.

#### 5.1.6 Coal petrography and rank

Coal petrography relates to the study of different microscopic organic constituents of coal to understand the coal type i.e., maceral composition and coal rank. Macerals are conventionally combined into three main groups: Vitrinite (vit), Liptinite (lipt) and Inertinite (int) based on their chemical, physical and optical properties. Vitrinite is the major maceral contributing to plasticity during caking and acts dominantly as a reactive maceral during combustion. Liptinite produces the highest yield of by-products in coals. Inertinite is generally inert during carbonization, not readily oxidized. The composition of Macerals and the rank of Vitrinite Macerals are found to control the technological behaviour of coal. Preferred coal rank for dry ash and slagging moving bed gasifiers are low and high, respectively. A fluidized bed gasifier can gasify low-rank coal whereas, an entrained flow gasifier can gasify coal of any rank.

#### 5.2 Coal Recourses in India

With 352 BT of coal resources including 177 BT of proved reserves, India has the 4<sup>th</sup> largest reserves of coal in the World. The total World proved reserves of coal are 1074 BT and India accounts for around 10% of the global reserves. US has the largest coal reserves followed by Australia and China.

#### 5.2.1 Category Wise Breakup of Coal Resources

A detailed analysis of the Indian coal reserves by category and depth is given in the tables below:

Coal Type	Proved	Indicated	Inferred	Total	% share
Prime Coking	4667.75	645.31	0.00	5313.06	1.51
Medium Coking	14971.60	71.60 11245.13 1862.86 28079.59		28079.59	7.97
Semi Coking	529.68	991.51	186.33	1707.52	0.49
Sub-Total of Coking	20169.03	12881.95	2049.19	35100.17	9.97
Non-Coking	156416.10	133945.92	25040.13	315402.15	89.57
Tertiary Coal	593.81	121.17	908.67	1623.65	0.46
Grand Total	177178.94	146949.04	27997.99	352125.97	100.00
% share	50.32	41.73	7.95	100.00	

(Resource in million tonne)

Table 5.2.1 - Source: GSI Coal Inventory'2021

It is evident from Table 5.2.1 that 90% of the coal reserves in India constitute noncoking coal or thermal coal which is primarily used for power generation and in industries such as cement and brick-kilns. Whereas approximately 10% of the reserves are coking coal reserves which are majorly used in steel production process. India imports a quarter of its coal requirements.

#### 5.2.2 Depth wise Breakup of Coal Resources

(Resource in million tonne)

Depth Range (m)	Proved	Indicated	Inferred	Total	% share
0-300	125560.46	62910.33	8593.29	197064.08	55.96
300-600	32302.84	65561.98	13101.13	110965.95	31.52
0-600 (for Jharia only)	14056.10	450.44		14506.54	4.12
600-1200	5259.54	18026.29	6303.57	29589.40	8.40
Total	177178.94	146949.04	27997.99	352125.97	100.00

Table 5.2.2 – Source: GSI Coal Inventory'2021

It is evident from Table 5.2.2 that around 56% of the total coal resources or 72% of the total proved reserves lie up to the depth of 300 metres. The shallower the

depth, easier the mining. More than 90% of the coal production in India is done through open-cast mining, which is usually up to a depth of 300 metres, whereas the rest is done through underground mining.

The above tables make it clear that India has huge reserves of coal. Therefore, it would be beneficial for India if it finds a sustainable way of using these reserves as the world including India is gradually transitioning away from coal toward cleaner fuels in the wake of climate change. The use of domestic coal reserves becomes even more important, especially when India does not have other sources of fuel – crude oil and natural gas, 82% and 45% of the requirement of those fuels is met through imports. This exposes India to the vagaries of price volatility and supply insecurity.



#### 5.2.3 R/P Ratio of Different Countries as of 2020

Figure 5.2.3 – Source: BP Statistics 2021

Figure 5.2.3 shows that the proved reserves of coal for India will last 147 years if India produces its coal at the current level as of 2020. This implies that India must figure out a sustainable way to use its coal reserves, otherwise this resource would remain buried under the ground as the transition towards cleaner fuels accelerates.

#### 5.3 Coal Availability for Gasification projects

**5.3.1** A number of efforts have been made to provide support to private sector and public sector for setting up of Coal Gasification plants. This includes concessions in revenue share for commercial auction of coal blocks. If the successful bidder consumes the coal produced either in its own plant(s) or plant of its holding, subsidiary, affiliate, associate for coal gasification or liquefaction or sells the coal for coal gasification or liquefaction process, a rebate of 50% on the revenue share quoted by the successful bidder will be allowed on the total quantity of coal consumed or sold or both for gasification or liquefaction on an yearly basis, subject to the following conditions:

i. At least 10% of scheduled coal production as per approved mining plan for that year shall be consumed or sold for gasification or liquefaction;

ii. Coal Controller's certification would be required for the quantity of coal consumed or sold or both for gasification or liquefaction.

**5.3.2** MoC has also proposed to give freedom to Coal India Limited for utilising coal for its own gasification projects at a rate to be decided by CIL.

**5.3.3** In order to address the problem of shortage of coal for coal gasification, Government has further issued a directive for the introduction of a separate auction window for allocation of coal linkage to coal gasification projects. Ministry of Coal has asked Coal India Ltd (CIL) and Singareni Collieries Company Ltd (SCCL) to add a new subsector for auction of coal linkages to the Non-Regulated Sector (NRS) alongside cement, sponge, iron/steel, aluminium, among others. In view of the Coal Block Allocation Rules, 2017 considering Production of Syn-Gas as one of an end-use sector, Production of Syn-Gas leading to coal gasification will now be considered as a subsector of NRS. The applicable floor price would be decided by CIL, SCCL as per provisions of the NRS linkage auction policy of 2016 and coal quantity and coal grade shall be offered as per availability after taking into consideration, to the extent possible, the demand from industry. The Fuel Supply Agreements (FSA) may be for the complete tenure of 15 years.

# **6 GASIFICATION TECHNOLOGIES**

#### 6.1 Introduction

Non-coking coal in the country is mainly consumed by the power sector (~60%). Conventional coal-based power generation technologies have several drawbacks, such as lower efficiency, environmental loading, and damages thereof. Power generation is the foremost source of carbon emission, which accounts for about 40% of total greenhouse gas emissions. Therefore, clean coal technologies are of utmost importance to adopt new coal conversion technologies for improving efficiency, reduction of CO<sub>2</sub> and other pollutant emissions. In this regard, coal gasification may be an attractive alternative to utilize coal efficiently meeting stringent environmental regulations. The clean syngas after removal of various contaminants can be utilized for various applications such as the production of a choice of fuel and chemicals such as hydrogen, methanol, ammonia, fertilizers, substitute natural gas, Direct Reduction of Iron ore (DRI), Fischer-Tropsch liquids, power generation, thermal applications as well as poly-generation from the same platform.

However, the high ash content of Indian coal is a crucial barrier in the development of suitable technology. Gasification had commercial implementations in India mainly for the production of chemicals/fertilizers. Many of the coal-based gasifiers had ceased operations due to problems related to the quality of coal. Recently, Jindal Steel and Power Ltd. (JSPL) have set up a DRI based steel production plant at Angul, Odisha. However, they are also facing problems in their Lurgi Fixed Bed Dry Bottom (FBDB) gasifiers with coals beyond 30% ash. A joint venture of Coal India Ltd. (CIL), Gas Authority of India Ltd. (GAIL), Rashtriya Chemicals and Fertilisers Ltd. (RCF) and Fertiliser Corporation of India Ltd. (FCIL) is in progress to establish coal gasification plants for the production of ammonia and urea at Talcher, Odisha. However, addressing of various operational issues arising due to complex gasification behaviour with high ash Indian coal is the focal challenge for the successful implementation of the project.

Coal analyses such as proximate, ultimate analysis, cold and hot crushing strength, caking index, gasification reactivity & surface area, ash content, Ash Fusion Temperature (AFT), ash composition, slag behaviour, rank, and petrographic characteristics, etc. are essential to understand gasification behaviour/performance of coal and to select, design & develop suitable gasifier.

Over the years, many gasification R&D facilities have been developed. Each facility attempts to exploit the chemistry, kinetics, and thermodynamics of coal gasification. Some of these processes have reached a commercial level. A common feature of all processes is that coal is contacted with gasifying agents mainly oxygen and steam in a reactor at high temperatures mostly under pressurized atmosphere. The solid coal loses moisture, volatile matter and residual char is gasified leaving ash as the residue. Carbon dioxide and heat are produced in-situ by the combustion reaction and further generated heat utilized to drive the endothermic gasification reactions. The art of gasification lies in balancing the exothermic and endothermic reactions while maintaining the required reactor temperature. Although the three main types of gasifiers (i.e. Entrained flow, fluidized bed, and moving bed) can be used to gasify coal, gasifier efficiency and stability are ensured under a range of values of certain characteristics of the coal.

Among the three types of the gasifier, the Entrained flow gasifier is a matured technology but yet to be tested for high ash coal. Fixed bed gasification technology is also a developed one but restricted

up to the coal of 35% ash. Fluidized bed gasifier seems to be suitable for high ash coal but not widely commercialised at this stage. Though, coal ash content is a crucial parameter, other properties such as cold and hot crushing strength, gasification reactivity & surface area, Ash Fusion Temperature (AFT), slag viscosity and behaviour, ash composition, caking nature, rank, and petrographic characteristics, etc. are also imperative towards selection of type of gasifier.

Moreover, the judicious utilization of coal through gasification depends on the blending of high ash coal with low ash feedstocks. As for example, high ash coal can be gasified in an *entrained flow gasifier* after proper blending with low ash coal keeping average ash content of the feed below the desired level. Similarly, the proper blend of high ash coal with low ash can be a feed for *moving bed gasifiers*. On the other hand, high-rank-low- reactive coal can be blended with low-rank high-reactive coal to achieve desired reactivity for its suitability in *circulating fluidized bed gasifier*. Therefore, proper utilization of Indian non-coking coal resources through gasification essentially requires gasification potential mapping of Indian non-coking coal will not only direct to select coal for proper gasification technology, but it will also guide towards utilization patterns and strategies for the gainful management of coal resources having a wide range of physicochemical characteristics.

#### 6.2 State of the art overview of the coal gasification technology

#### 6.2.1 National scenario of coal gasification

Coal is considerable important in India and for the growth of Indian economy. Coal-based thermal plants are generating significant electricity (70%) which is consumed and demanding in energy sector. It is pertinent to mention that high ash content in Indian coal is primarily facing operational challenges in gasifiers and boilers. Heavy slag formation and the corrosion were reported by several operators. Underground coal gasification signifies enormous potential in recovering the high heating value of abundantly available high ash coals by insitu conversion to gas. Ministry of Coal, GOI, has sanctioned a policy for recovering gas from the resources of lignite.

It shows several groups around the globe (China, India, EU, Australia & USA) have contributed extensively on understanding various aspects of underground coal gasification (UCG) either through experiments or using modelling. Mahajani and his group at IIT Bombay collaborates with ONGC and carried out experimental and computational studies on UCG. CSIR-CIMFR join hands with NIET and IIT-Kharagpur for investigating the feasibility study of UCG in the north-eastern region of India, based on the availability of coal in northeast India. Further to note, CSIR-CIMFR, NIET and IIT-KGPs' collaboration developed techniques for measuring gas concentration within the coal steam and equipment to understand the methane concentration in mines underground. CSIR-NCL has contributed to coal combustion using fluidized bed gasifier and they collaborated with GAIL team while working on UCG. It is significant to note that GAIL is of the major part in pursuing UCG in commercial scale.

Jindal Steel Plant is the first company in India to build the coal gasification plant and the first in the world to produce DRI/Steel via coal gasification process. State-run Coal India (CIL) is planning to set up a coal-gasification project with an estimated investment of Rs 5,800 crore in the Dankuni coal complex in West Bengal. The project, which is now in the planning stage, will convert 1.5 million tonne (MT) of the fuel into other products and reduce emissions. Talcher Fertilizers Limited, a joint venture company in Odisha is working on coal gasification to produce urea. Apart from that, IOCL, Reliance are also working in same line.

In India, coal-based energy production is 72% according to 2020 data. Indian coal is basically similar to Gondwana coal with low calorific value (4500 Kcal/kg) containing high amount of ash resulting that Indian power plant using Indian coal consume much more coal, i.e. 0.7 kg/kWh power generation, as compared to USA coal with high calorific values. As a percentage of India's total power production, coal is expected to decline from 70% to 50% over the next decade, according to India's Central Electricity Authority. Syngas produced from coal gasification is piped to industrial usage in Dankuni area near Kolkata.

Many coal-based fertilizer plants can also be economically retrofitted to produce synthetic natural gas. It is estimated that the production cost for syngas could be below US\$6 per million British thermal units (\$20/MWh) rms, coal use will increase as India's overall energy demand grows. Although coal can be of high choice for *hydrogen production*, yet, it has not been encouraged, because hydrogen extraction from coal (with moisture content) will apparently lead to global pollution due to concomitant rise in carbon emission. Coal can be an important source for the production of Hydrogen. However, coal has not been encouraged elsewhere because of concerns that extraction of hydrogen via coal (from the moisture embedded in coal) will lead to carbon emission. It is to be mentioned that 73 MT hydrogen is used for refining, producing ammonia while 42 MT is for methanol, steel and for elsewhere.

The possible industry players who are likely to venture in coal to hydrogen are Air Products, Haldor Topsoe, Thermax, Coal India Ltd, Mahanadi Coalfields Ltd, Larsen & Toubro, Dev Energy, etc. Since it is essential to combine CO<sub>2</sub> capture with gasification, it is likely that the industries which have expertise in gasification as well as CO<sub>2</sub> capture will advance more in the domain of coal to hydrogen. Air Products, Haldor Topsoe, Coal India Ltd, Larsen and Toubro, Thermax and Mahanadi Coalfields Ltd are such companies. BHEL has indigenously prepared high ash coal gasification technology for methanol production. There are also smaller industry players who are likely to venture in this domain like Dev Energy, Praj Industries and other gasifier industries.

In addition, the Committee had the benefit of learnings in the recent gasification projects executed in the country for conversion of coal to methanol which were awarded by DST to Thermax and BHEL R&D. The EIL R&D also has experience on the gasification technology based on a pilot plant built up by them. In India, the RIL had set up huge gasifiers for petcoke gasification in which they are producing hydrogen. IOCL R&D also has a patented technology based on which a pilot plant had been set up for co-gasification of coal, petcoke and bio-mass. Such co-gasification of coal with bio-mass could possibly be also examined to introduce the green factor in production of hydrogen from coal.

#### 6.2.2 International scenario of coal gasification

Coal accounts for over 37% of the world's electricity supply. About 70% of world steel production depends on coal feedstock. Coal is the world's most abundant and widely distributed fossil fuel source. However, each year burning coal produces over 14 billion tonnes of carbon dioxide (CO<sub>2</sub>), which is released to the atmosphere, most of this being from power generation. Development of new 'clean coal' technologies is attempting to address this problem so that the world's enormous resources of coal can be utilized for future fuel generations without contributing to global warming.

The coal gasification technology has attracted increasing importance internationally due to the low production cost of hydrogen and sustainability. In the United States, 95% of hydrogen is produced by a reaction between a methane source, such as natural gas, and high-temperature steam (700°C–1,100°C), referred to as steam methane reforming (SMR). About 4% is produced through coal gasification, and 1% is produced from electrolysis.

China, the biggest producer of coal on the planet also shares the highest production of hydrogen (20 million tonnes per year), mostly from coal gasification. Currently, 70% of China's hydrogen comes from nearly 100 coal gasification plants installed in the country. The overall production cost of hydrogen from coal in China is also lower (0.95-1.90/kg H<sub>2</sub>) compare with the natural gas derived hydrogen (1.27-2.37/kg H<sub>2</sub>) and green hydrogen (3.94-5.54/kg H<sub>2</sub>). Despite the low production cost, the emission of CO<sub>2</sub> from the coal-gasifier is huge. It is estimated that 8 kg coal produces about 1 kg of hydrogen in China and emit 20 kg of CO<sub>2</sub> in the environment.

Since ratification of the Paris Accord in 2016, governments around the world have made increasingly strong commitments to profoundly reduce their greenhouse gas emissions. Many nations have selected economy-wide net-zero greenhouse gas emissions targets in their planning (by 2040–2060) and in many cases have made commitments to rapidly reduce greenhouse gas emissions much sooner. They have matched these commitments with ambitious investments in clean energy production and use as part of a decarbonization strategy. In concert, many large and significant companies have committed themselves to net-zero goals between 2030 and 2050, including leading energy, chemical, shipping, and aviation companies. Against this backdrop, low-carbon hydrogen has emerged as an important option to provide net-zero emissions energy services.

The greenhouse gas free hydrogen (blue hydrogen) can be produced by combining carbon capture and storage technology with the existing gasifiers. In terms of gasification with carbon capture, there are currently three facilities producing hydrogen from coal, coke and petroleum coke at scale, with a combined capacity of around 0.6 MtH<sub>2</sub>/y, namely Great Plains and Coffeyville in the USA and Sinopec Qila in China. These plants demonstrate that large-scale production of low emissions hydrogen using carbon capture can already be technically and commercially feasible.

Canada became host to the world's largest green hydrogen plant, with its 20-megawatt (MW) nameplate capacity. Along the way, the European Union established a green hydrogen installation target of 40 gigawatt (GW) by 2030, while Chile announced a 25 GW target by

2030 in its effort to become the "cheapest producer of green hydrogen on Earth." Other countries in this rapidly growing list include South Korea, Australia, Saudi Arabia and Portugal.

Japan and Australia have announced the launch of a new brown coal-to-hydrogen project. This project will use brown coal in Australia to produce liquefied hydrogen, which will then be shipped to Japan. The project is located in the Brown Coal Reserves in the state of Victoria and is run by Kawasaki Heavy industries. This project is significant because it will help Japan to meet its "net zero emission" target by 2050. Japan, which is the fifth largest energy consumer of the world, has aimed to boost the annual hydrogen demand by ten times amounting to 20 million tonnes by 2050. It is equal to 40 percent of the current power generation in the country. Japan, the world's fifth-largest energy consumer, aims to boost its annual hydrogen demand tenfold to 20 million tonnes by 2050, equivalent to about 40 percent of its current power generation. Australia, already dominant in the global liquefied natural gas (LNG) trade, is hoping liquefied hydrogen will give it a greener market for its coal and gas.

Envirotherm GmbH have built 14 gasifiers for syngas production for ammonia and urea synthesis using coal, mostly in China and India. Their technologies are based on slagging fixed bed gasifier (BGL type) and circulating fluidised bed for gasification as well as combustion.

#### 6.3 Comparison between various types of gasifiers for coal to hydrogen production

Gasification is recognized as the process converting any carbon-based raw material into synthetic gas using air, water vapor or oxygen. Using gasification techniques, many raw materials and wastes, such as coal, car tires, sewage sludge, sawdust, wood and plastic waste can easily and effectively be converted into useful outputs. At the end of any gasification process, a product gas may include some or all of the outputs that may generally contain CO,  $H_2$ ,  $CH_4$ , ash, tar,  $H_2S$ ,  $NH_3$ , HCl and HCN. The product gas then needs to be purified from the contaminants, particles and some other substances which really decrease its calorific value by applying various gas clean-up processes, and the useful gases, such as CO,  $H_2$  and  $CH_4$  are separated accordingly. In the gasification process, it is clear that four different types of coal are generally utilized in a suitable manner, which are (i) lignite (low rank), (ii) sub-bituminous coal (low rank), (iii) bituminous coals (medium rank), and (iv) anthracites (high rank). However, it is important to note that, according to the open literature, these materials are generally gasification, (ii) moving bed gasification, (iii) fluidized bed gasification, (iv) entrained flow gasification and (v) plasma gasification. A schematically illustration can be seen from Fig.6.3 and are compared by considering their various parameters as tabulated in Table 6.3.

Fixed bed gasifiers are divided into updraft gasifier and downdraft gasifier. In the updraft gasifier, the fuel is loaded from the upper part of the reactor, and the gasifying agent is supplied to the system from the lower part of the reactor. Synthesis gas produced is taken out from the upper part of the reactor. In downdraft gasifiers, fuel is loaded from the upper part of the reactor, and the gasifying agent is sent to the system through the channels opened in the middle of the reactor, and synthesis gas is taken out from the lower part of the reactor.

Moving-bed gasifiers commonly operate at moderate pressures (25-30 atmospheres). Feedstocks in the form of large coal particles and fluxes are loaded into the top of the refractory-lined gasifier vessel and move slowly downward through the bed, while reacting with high oxygen content gas introduced at the bottom of the gasifier that is flowing counter currently upward in the gasifier. The basic configuration is the same as per the fixed bed gasifier.

Fluidized bed gasifiers have many different designs such as bubbling bed, circulating bed, internally circulating bed, spouted bed and dual bed. In these reactors, fuel is loaded into the system from the side of the reactor. It is quickly mixed with the bed material and heated to bed temperature in a very short time, and gasifying agent is introduced from the lower part of the reactor while the synthesis gas exit takes place from the upper part of the reactor, and the slag exits from the lower part of the reactor. Among all of the fluidized bed gasifiers, circulating fluidized bed gasification (CFBG) has a high mass and heat transfer rate as well as highly efficient gas-solid contacting. At low operating temperature of CFBG, a longer residence time of solid can be achieved leading to a higher gasification yield. CFBG process is more energy efficient as it is an endothermic process. Only the required heat will be generated to maintain the process at the optimum temperature. Practically, all the heat produced will be utilized throughout all the processes, as it is an adiabatic and isothermal process.

In entrained-flow gasifiers, fine coal feed and the oxidant (air or oxygen) and/or steam are fed cocurrently to the gasifier. This results in the oxidant and steam surrounding or entraining the coal particles as they flow through the gasifier in a dense cloud. Entrained flow gasifiers operate at high temperature and pressure and extremely turbulent flow which causes rapid feed conversion and allows high throughput. The gasification reactions occur at a very high rate (typical residence time is on the order of few seconds), with high carbon conversion efficiencies (98-99.5%). The tar, oil, phenols, and other liquids produced from devolatization of coal inside the gasifier are decomposed into hydrogen (H<sub>2</sub>), carbon monoxide (CO) and small amounts of light hydrocarbon gases. Entrainedflow gasifiers have the ability to handle practically any coal feedstock and produce a clean, tar-free syngas. Given the high operating temperatures, gasifiers of this type melt the coal ash into vitreous inert slag.





*Fig.6.3: Schematic illustrations of gasifier types (a) Updraft Gasifier, (b) Downdraft Gasifier, (c) Fluidized Bed Gasifier, (d) Entrained Gasifier, (e) Plasma Gasifier.* 

Table 6.3: Compariso	n between the	different type	es of gasifie	ers available
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	СТ	SGET	SS	RZT	Scale	CGE
Fixed Bed Gasifier	Lignite Subbituminous	460-650 °C	Normal (5 cm)	1000-1100 °C	Large	80%
Fluidized Bed Gasifier	Lignite Subbituminous	800-1000 °C	Small (0.6 cm)	800-1000 °C	Large	78-81%
Entrained Bed Gasifier	All types of coal	900-1600 °C	Very small (0.015 cm)	1990 °C (max)	Large	80%
Plasma	All types of coal	1250 °C	Normal (5 cm)	1500-5500 °C	Small-	80-90%

Gasifier	(min)		Modular	

CT: Coal Type, SGET: Synthesis Gas Exit Temperature, SS: Sample Size, RZT: Reactor Zone Temperature , CGE: Cold Gas Efficiency.

Among these gasification processes, the entrained and plasma gasification of coal may generally be carried out at higher temperatures between 1200 and 1700 °C, respectively. However, the others may require lower operating temperatures than 1200 °C. Plasma gasification is recognized as a relatively new technology using plasma torch in order to produce clean and renewable fuels but the main disadvantage of this technology is the requirement of high energy input as it operate at a higher temperature then other types of gasifiers. During the process, the raw materials in the system are decomposed. Due to very high temperature, higher conversion efficiency can potentially be achieved. The products released during the process are essentially syngas and slag.

**Considering the important advantages and disadvantages of different type of gasifiers, circulating fluidized bed gasification process can suitably be applied to produce the syngas from coal.** During such a circulating fluidized bed coal gasification process, CO and H<sub>2</sub> can be obtained after gas cleaning. Coal loaded into the circulating fluidized bed gasification unit is gasified at moderate temperature. The synthesis gas produced in it is passed through the heat exchanger and sent to the gas cleaning unit after being cooled accordingly. Also, the purified gas is pressurized by a compressor, and sent to a hydrogen separation unit, using techniques, such as pressure swing adsorption, cryogenic distillation or membrane separation. At the end of the separation process H<sub>2</sub> fuel is obtained. According to the literature results, it can be said that, at the end of the gasification of coal, approximately 0.1-0.17 kg of hydrogen gas is produced from 1 kg of coal.

# 7 SYNGAS CLEANUP FOR DOWNSTREAM PROCESSING

#### 7.1 Coal Gasification for Hydrogen

The syn-gas produced from coal gasification can be converted to liquid fuels and chemicals as well as H<sub>2</sub>. Hydrogen production from coal gasification route is becoming important considering the present thrust for switchover to a hydrogen-based energy economy and the vast reserve of coal present in India. The hydrogen produced from coal is designated as either black or brown hydrogen as per the current convention of colour coding for the hydrogen, depending upon the feedstock used and the processes involved. The black hydrogen is defined as derived from bituminous while the brown is from lignite coal. After combining with carbon capture, storage and utilization the black/brown hydrogen can be converted to blue hydrogen if the overall process does not emit CO<sub>2</sub>. Gasification is heart of the process for converting coal to syngas which after required cleaning from the contaminants, as required to protect downstream process catalysts from poisoning and also to meet emission regulations, can be converted by water gas shift reactions to a hydrogen rich gas which can be further purified to very high purity hydrogen (>99.9%). Gasification is the partial combustion of carbonaceous feeds in the presence of a controlled amount of oxygen, where oxygen/carbon ratio is adjusted to ensure that most of the feed carbon is converted to CO and most of the hydrogen to molecular hydrogen. Gasification processes are operated either at a near atmospheric pressure or at an elevated pressure in presence of steam, air/oxygen. The equilibrium considerations suggest slower decomposition of steam and CO<sub>2</sub> with increasing pressure. However, the effect of pressure up to 30 kg/cm<sup>2</sup> on product composition is not much. Most commercial gasifiers operate at elevated pressures. In gasification the carbonaceous feed particles pass through three major reactions i.e., combustion (reaction with  $O_2$ ), Boudouard reaction (reaction with  $CO_2$ ) and steam gasification (reaction with steam). After entering into the gasifier the feed is volatilized at 1000–1500 °C and the resulting hydrocarbons react to give carbon monoxide and hydrogen (syngas) as per the following overall equation.

 $CH_xO_y$  + (1- y)  $H_2O \rightarrow (x/2 + 1-y) H_2$  + CO

Typical composition of the raw syngas is shown in Table 7.1.

Composition	Range
H <sub>2</sub>	25 - 30 % (v/v)
СО	30 - 60 % (v/v)
CO <sub>2</sub>	05 - 15 % (v/v)
H <sub>2</sub> O	2-30 % (v/v)
CH <sub>4</sub>	0-5 % (v/v)
H <sub>2</sub> S	0.01 - 1 % (v/v)
N <sub>2</sub>	0.5 – 4 % (v/v)
NH <sub>3</sub> + HCN	0-0.3 % (v/v)
Ar	0.2 – 1.0 % (v/v)
COS	0-0.1 % (v/v)
Ni & Fe Carbonyls	1 - 4 ppmv

#### Table 7.1. Typical composition of the syngas produced by gasification

#### 7.2 Syngas Conditioning and Clean up

Syn gas clean-up for removing particulates, trace metal contaminants and acid gases are required before downstream processing for either chemical synthesis such as methanol or  $H_2$  production.

#### 7.3 Particulate Removal

Raw syngas leaving gasifier contains fine ash, char and slag that need to be removed prior to downstream processing. The char and ash can be removed from the gas in two-stage water wash. This consists of a quench pipe and carbon separator followed by a packed tower, the carbon scrubber. In the quench pipe around 95 % of the carbon is removed by direct water spray. In the scrubber the gas is washed in counter-current flow in two packed beds. A circulation system is employed over the lower bed using a circulating pump. The upper bed is washed with return water from the carbon recovery section.

Refractory lined cyclones and high temperature candle filters are commercially established technology for dry ash removal from syngas. Ceramic cross flow candle filters are mostly used for the dust removal from hot gas. Moving bed filters are also being developed for more economic removal of these char particles by dry process. The organizations like Westinghouse and Southern Research Institute (SRI), Coal Research Institute (CRI), UK, University of North Dakota Energy and Environmental Research center (UNDEERC) etc. are engaged in the research on the development of new hot gas filters for particulate removal.

High-temperature, high-pressure (HTHP) filters are key components in the successful development of advanced coal-based integrated gasification combined cycles. They operate best in the temperature regime 300-500 °C. The first step in syngas clean-up is to remove the particulate in order to protect the downstream processes from fouling. The HTHP filters also protect the downstream gas turbine from particle-related damage and clean the process gas to satisfy dust emission standards.

Dusty syngas enters the filter unit through a tangential inlet nozzle and then flows in an annulus between the vessel wall and a metal shroud. The vessel is refractory lined and the surface covered with a metal liner to protect the refractory from erosion damage and also to assist the free flow of captured particulate out of the vessel. The centrifugal action induced by the tangential entry separates out the coarser, denser particles in the annular region and the gas with fine particulate, flows into the central filtration zone of the vessel. Particulate collects on the outside of the elements, the clean gas passes into the plenum, up the support pipe and on to the exit of the filter unit.



Figure 7.3. Candle Filter Unit for Ash removal

Syngas leaving candle filters are also quenched and scrubbed with water for very fine particulate removal. Water scrubbing can remove effectively all ash particles along with chlorides, ammonia, and part of  $H_2S$  and other contaminants from syngas. The water scrubbed syngas is reheated to ~200 °C and sent for downstream COS removal by catalytic hydrolysis and/or sour water gas shift followed by cooling in the low temperature gas cooling system prior to mercury and acid gas removal respectively.

Alkaline Earth Silicates (AES) fibre based filter from TENMAT, with proprietary high temperature bonding agents and is able to resist temperatures up to 1000°C, which removes the need to cool down the gases before they reach the filter elements. Being a spark-proof mineral composite material, the filter removes the need for costly anti-spark arresters. Its homogenously porous nature also avoids potential issues of preferred paths through the walls of the filter elements, and due to its strong mechanical properties, the filter elements can undergo numerous regenerative cycles via counter-flow pulses of cleaning gases, without the wear & tear usually witnessed on fabric filters when regenerated in the same way. Claimed filtration efficiency of the filter is >99.99% with filtration capability of < 1 micron particle size. Porvair Filtration Group has a portfolio of filtration systems that operate with IGCC (Integrated Gasification Combined Cycle) gasification of coal and petroleum coke. They design and manufacture filtration and separation equipment for processes such as PFBC (Pressurized Fluidized Bed Combustion), biomass and pelletised MSW (Municipal Solid Waste). Similarly Siemens Westinghouse has an offering for particulate control device (PCD) for filtration of gasification ash from synthesis gas. Commercial hot gas clean-up filters are also available from the 3M company (SiC candles, and silicon carbon coated SiO2 ceramic fiber candles), Pall Advanced Separation Systems Corporation (sintered iron aluminide and silicon carbon ceramic candles), McDermott Technologies (composite candle filters), Honeywell (composite candle filters) and U.S. Filter/Schumacher (granular SiC candles).

#### 7.4 Disposal of Coal Gasification Ash and Slag

As opposed to conventional coal combustion, fly ash volumes are relatively low in coal gasification, though it varies with the type of gasifier and nature of coal used. This is since gasifiers operate at temperatures higher than the fusion point of ash (typical of IGCC systems such as those offered by GE Energy, CB&I E-Gas<sup>™</sup> and British Gas / Lurgi). At such high temperatures, most of the inorganics in coal convertsto **slag or bottom ash**. Slag production is a function of ash content, so

even low-ash coal produces more slag than petroleum coke under similar gasification conditions, and Indian high ash coal is expected to produce even more. Regardless of the feed, as long as the operating temperature is above the fusion temperature of the ash, slag will be produced.

Because of its particular properties and non-hazardous, non-toxic nature, slag is relatively easily marketed as a by-product for multiple advantageous uses, which may negate the need for its long-term disposal. This can improve the economics of disposition of solid by-product from gasification processes. However, such applications must be established in a timely manner to prevent high volumes of solid waste generation and disposal. For example, a 100-MW power plant based on IGCC technology using 1000 tons of 10% ash coal per day may generate over 110 tons/day of solid waste or slag, consisting of vitrified mineral matter and unburned carbon.

Slag captures toxics/heavy metals into glassy matrices and does not allow them to leach out of the material, unlike thermal coal plant combustion ash from which toxic species can possibly leach and find their way into groundwater and surrounding soils.

**Ash** (sometimes referred to as Char) is the finer component of the gasifier solid residuals, composed of unreacted carbon (sometimes as much as 10 percent) with various amounts of siliceous ash. Storage of this material can be hazardous due to its potentially flammability or dust explosion risk. It can be recycled back into the gasifier to increase carbon usage and has been used as a supplemental fuel source for pulverized coal combustion. The irregularly shaped particles have a well-defined pore structure and have excellent potential as an adsorbent and precursor to activated carbon.

In a project between the Department of Energy (DOE) and CAER, potential uses of char were investigated, specifically as adsorbers for emissions control. Carbon char has the potential to control mercury (Hg) and nitrogen oxides (NOx) emissions. Not having to treat the char is significant, as treatment would be an additional cost.Additionally, gasification char adsorbed significantly more NOx than all other test materials in a US DOE study except for a specially designed activated carbon NOx adsorber. Naturally, the gasifier char, a "waste," is significantly less expensive than a specialty adsorber and being able to put it to good use makes plant operations more economical.

Some examples of coal gasification slag utilization are summarized below:

- Preparation of high modulus sodium silicate by mechano-chemical synergistic activation of coal gasification slag [3 -Jiangshan Qu et al, Science of The Total Environment, Volume 801, 20 December 2021, 149761]
- Asphalt paving aggregate hot mix and seal coat aggregate[4 US patent US5166109A, S.B Alpert et al, 1991, since expired)

Additional ideas listed on the NETL website [2] include:

- Construction structural backfill
- Portland cement aggregate/additive; cement kiln feed
- Asphalt shingle roofing granules
- Pipe bedding material
- Blasting grit or other abrasive application
- Polishing media
- Snow and ice control

- Mineral filler
- Landscaping
- Road drainage media
- Water filtering medium
- Water-jet cutting a new application for boiler slag
- Building materials: Slag lightweight aggregate (SLA) and ultra-lightweight aggregate (ULWA)

The Tampa Clean Coal Technology Project in Polk County, Florida, USA, was able to has successfully processed slag for use in cement production. In order to meet the required slag specification, the size of the fines handling system was doubled, and additional slag handling equipment was installed to deal with unconverted carbon in the fines. The plant was modified after startup to better separate unconverted carbon for recycling or sale and produce slag that is more consistently suitable for the cement industry [5 - McDaniel, J. and M. Hornick, "Polk Power Station - 5th Commercial Year of Operation," Paper presented at 2001 Gasification Technologies Conference, San Francisco, California, USA October 2001].

Another study of SASOL coal gasification ash from South Africa similarly suggested use in cement [6 - Coal gasification ash and Weathered fly ash, as partialreplacement of Portland cement – effect on selected durabilityproperties of concrete, Maboea and Otieno, MATEC Web of Conferences 199, 02021 (2018)]

As coal gasification technologies are explored for syngas and hydrogen production, it becomes imperative – especially for Indian high-ash coal – that slag and ash utilization methods be developed, tested, and commercialized in order to address the costly problems associated with its disposal as solid waste. There appears to be inadequate research in this area within India to date, and a serious, outcome-oriented thrust is required to ensure that solid waste disposal costs and environmental impact do not negate the potential national benefits of a coal gasification programme.

# 7.5 Water gas shift reactors

In the commercial gasifiers the produced syngas contains high amount of CO (Table 7.5), which can be converted to  $H_2$  by water gas shift reaction (WGSR).

Gasification	Composition of syngas (%)								
Technology	N <sub>2</sub>	H <sub>2</sub>	СО	CO <sub>2</sub>	H <sub>2</sub> O	CH <sub>4</sub>	H <sub>2</sub> S	NH₃	Total
Техасо	5.8	27	35.6	12.6	18.6	0.1	-	0.1	99.8
Koppers Totzek	1.4	32.8	58.7	7.1	-	-	-	-	100
Shell Lurgi	5.1	29.7	60	2.3	2.1	-	0.8	-	100
Winkler	3.0	49.5	25	18	-	3.0	1.5	-	100

Table 7.5 Typical composition of Syngas produced by various gasifiers

This is an important reaction for extra  $H_2$  production from syngas coming out from coal gasifier. In this catalyzed reaction, steam and CO react to produce  $H_2$  and  $CO_2$  and the reaction is represented as:

 $\rm CO + H_2O \rightarrow H_2 + CO_2$ 

If the shift reaction is carried out after the removal of sulfur from the syngas then it is called as sweet-gas shift reaction and if it is done before sulfur removal then it is called as sour-gas shift reaction.

Sweet water gas shift is not preferable in coal gasification applications because of the sensitivity of the catalyst towards sulphur and chloride poisoning. The need for cooling the syngas before sulphur removal through conventional solvent based processes also leads to moisture condensation that then requires additional reheating and reinjection of steam into the treated gas after H<sub>2</sub>S removal in order to provide required moisture for shift. The process thus overall becomes energetically inefficient.

Sour shift is preferred in coal gasification since the moisture entrained following water scrubbing of hot syngas can be fully utilized to drive the shift reaction towards more  $H_2$ . The sour shift is based on cobalt-molybdenum catalyst and operates in the temperature range 260-450 °C.

A typical set up of sweet–gas shift reaction is comprised of two high temperature shift (HTS) and one low temperature shift (LTS) conversion with cooling between the reactors whereas for sour-gas shift it consists of two to three conversion stages with heat exchangers and sometime steam addition as required. Sweet-gas shift can operate with less steam, which is intended on the economic point of view. Within two HTS steps sweet–gas shift can reduce CO concentration from 44.6 % to 2.1 % at the end of run (EOR) conditions. Without steam, the residual CO can be further converted to 0.5 % (EOR) in LTS step. Using the sour-gas shift CO can be converted from 44.6 % to 1.8 % (EOR) within two steps using a slightly higher amount of steam as compared to sweet-gas shift concept. To reach a CO level below 1 %, it is necessary to add a remarkable quantity of steam before running the shift reaction in a third reactor.

Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> is the catalyst for the LT reaction (200–250 °C) whereas Fe<sub>2</sub>O<sub>3</sub>–Cr<sub>2</sub>O<sub>3</sub> catalyst is used in the HT (320-450 °C) reaction. Although the Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> catalysts typically have relatively higher selectivity than Fe<sub>2</sub>O<sub>3</sub>–Cr<sub>2</sub>O<sub>3</sub>, their main disadvantage is lower resistance to S- and Cl-impurities. Apart from the LT/HT catalysts, some catalysts can also be used over wider ranges of temperatures, e.g. Co–Mo/Al<sub>2</sub>O<sub>3</sub>, which is also stable towards S-impurities. The WGSR may be carried out in conventional catalytic packed-bed reactors by the addition of such oxides with feed. The size of the catalyst bed is smaller for the sweet-gas shift reactor. However, the sweet-gas shift catalysts are usually more expensive compared on a unit price basis. Sweet-gas shift catalysts have also more complex start-up procedure and are more sensitive with respect to poison and maloperation. Typical characteristics of water shift catalysts are shown in Table 7.5.1 below.

Table 7.5.1. Characteristics of water shift catalysts

Catalyst	Composition	Temperature	Active	Poisons
		Range (° C)	State	
High Temperature	89 % Fe <sub>2</sub> O <sub>3</sub> , 9 % Cr <sub>2</sub> O <sub>3</sub> , and	350-400	Reduced	S, Cl
Shift	2 % CuO			
Low Temperature	33-42 % CuO, 39-65 % ZnO and	200-250	Reduced	S, Cl
Shift	20-33% Al <sub>2</sub> O <sub>3</sub>			
Sour Shift	$3\%CoO_3$ and $13\text{-}15\%MoO_3$	260-450	260-450 Sulfided	
			(20 ppmv)	

Following the thermodynamics of the shift reaction, the conversion of CO to  $H_2$  is favored at higher temperatures, which allow recovery of the heat of reaction at sufficient levels to generate highpressure (HP) steam. Therefore, HP steam can be used within the plant; this consideration may also influence the decision for using sweet-gas or sour-gas shift reaction. Depending upon the downstream use of the syngas the position of shift reactors and AGR loop can be modified to optimize the process economy. For power application sour gas shift reaction is favorable whereas for chemical use sweet gas shift is required. If high purity hydrogen is required in maximum quantity, the recycling of tail gas from the PSA unit to the shift reactor is required. Such recycling of tail gas increases hydrogen recovery by 98 % and can help in reducing the gasifier size by 10 %.

The major components of the syngas produced in the gasifier are CO and  $H_2$ . Other gases like  $CO_2$ ,  $H_2S$  and COS are also present in considerable amount in it. A typical composition of the syngas produced before and after shift reaction is summarized below (Table 7.5.2):

Parameters	H <sub>2</sub>	N <sub>2</sub>	СО	Ar	CH <sub>4</sub>	CO <sub>2</sub>	H <sub>2</sub> S	COS	H <sub>2</sub> O	Total
Mole % (before	43.01	0.35	49.98	0.05	0.39	4.36	1.61	0.08	0.16	100
shift reaction)										
Mole % (after shift	61.53	0.24	1.12	0.03	0.26	35.50	1.14	50	0.18	100
reaction)								ppmv		

Table 7.5.2. Typical composition of syngas before and after shift reaction

Amongst these components of the syngas  $H_2S$  and COS (carbonyl sulfide) are highly corrosive. Hence, these are required to be removed from the syngas gas before its downstream use. Carbon dioxide may also be captured /sequestered to reduce the environmental emission from turbine placed in the downstream. These gases ( $H_2S$ , CO<sub>2</sub> and COS) are also termed as acid gas. From the above table it is also evident that maximum amount of  $H_2$  (around 60 %) is produced during the gasification followed by shift reactions. This  $H_2$  can be used in various purposes if it is separated from the syngas.

# 7.6 COS Removal

Depending upon the configuration of the coal gasifier used and its operating conditions typically 3-10% of the sulphur present in coal is converted to carbonyl sulphide (COS). In application such as coal to methanol (CTM), <1 ppm sulphur is recommended in the syn gas feed to the reformer.

The COS present in syngas is converted to  $H_2S$  by catalytic hydrolysis process and then the  $H_2S$  is removed by downstream AGR unit. The syngas after water scrubbing is passed through the catalytic hydrolysis unit that uses an activated alumina based catalyst which normally operates at 175-200 °C.

COS + H<sub>2</sub>O ----->CO<sub>2</sub> + H<sub>2</sub>S

CKA-3 of Haldor Topsoe is a promoted alumina-based catalyst used for hydrolysis of carbonyl sulfide (COS). As per the company brochure "The CKA-3 catalyst has a high activity even at low temperatures, which is essential for maximum conversion of COS, as the chemical equilibrium is favourable at low temperature. Whether the end product is chemicals or power, hydrolysis of carbonyl sulfide can be used in some gasification based plants to provide a more optimal process layout. In the COS hydrolysis reaction, the carbonyl sulfide is converted to hydrogen sulfide and carbon dioxide, which are easier to remove in the Acid Gas Removal (AGR) unit. As a result, the cost and size of the AGR can be reduced, which leads to significant savings".

Filsorb S 201 from DORF KETAL is a mixed metal oxide catalyst with CuO and ZnO in proprietary composition for COS removal. The catalyst is claimed to be suitable for removing COS from the gas as well as liquid phase. The product purity normally achieves below the detectible limits.

# 7.7 Mercury Removal from Syngas

Current commercial practice is to pass cooled syngas from low temperature gas cooling system (LTGC) through sorbent beds upstream of the Acid Gas Removal unit to remove Mercury and other trace elements (As, Se, Cd, Sb, P).

UOP has commercial mercury sorbents such as Copper impregnated Alumina as well as zeolite based sorbent called HgSIV which is modified with silver for enhanced mercury removal. These sorbents which can remove ppm level elemental mercury from syngas. The process is based on dual bed Temperature Swing Adsorption (TSA) where when one bed is in operation for mercury removal the other bed will under regeneration by thermal desorption.

Eastman Chemical company has applied activated carbon based mercury control technology in one of their chemicals from coal facility. The sorbent is based on Calgon's HGR-P sulphur impregnated activated carbon pellets. The operating conditions are as follows:

- Temperature of sorption: ~30 C
- Bed operating pressure: ~60 bar
- Gas contact time: ~20 s (based on total packed volume)
- Mercury removal efficiency from syngas: 90-95%
- Carbon lifetime: 12-18 months

Putting two beds in series up to 99% mercury removal is possible. The positioning of the bed is upstream of the sulphur recovery unit

The above processes for mercury removal from syngas is optimal for lower temperature operation (up to 200 °C) which implies pre cooling of syngas that leads to overall process inefficiency due to requirement of elaborate water circulation and treatment systems downstream of the gasifier.

The zeolite and activated carbon based sorbents are ineffective at temperature beyond 200 °C. Current trend is to develop warm or high temperature removal system for heavy metals including mercury from syngas. In this context NETL in collaboration with Johnson Matthey has developed Pd based sorbents (US 7033419) which have shown promise for high degree of mercury removal (99%) at temperature beyond 280 °C.

#### 7.8 Acid Gas Removal

The acid gas present in coal gasifier off-gas consists primarily of  $H_2S$ , COS and CO<sub>2</sub>.  $H_2S$  and COS are present in trace level typically 100 ppm and 10 ppm respectively while CO<sub>2</sub> is present in percentage range of 5-15%. To meet environmental regulations in terms of meeting stack gas content of SO<sub>2</sub> in the IGCC scheme of power production, the sulphur content in syngas is typically brought down <10 ppm level. Whereas when the target is production of chemicals from syngas, in order to minimize poisoning of the downstream catalysts the sulphur level needs to be lowered down to < 0.1 ppm.

Established processes for acid gas removal are based on solvents which could be either physical solvents or chemical solvents. The classification of solvents as physical or chemical is based upon the nature of interaction of the acid gases with the solvent molecules. In case of physical solvents interaction while in the case of chemical solvents which are typically basic in nature there occurs chemical bond formation through acid base interaction of the acid gases from the solvent. Typically the solvent based processes involve counter-current sorption of acid gases from the syngas in an absorber with a regenerable lean solvent. The acid gas lean syngas coming out from the top of the absorber is sent for downstream systems processing while the acid gas rich solvent leaving the bottom of the absorber is sent to the regenerator where the solvent is stripped with steam under low pressure to remove absorbed sulphur. The concentrated acid gas rich in H<sub>2</sub>S that exits top of the regenerator is sent for the sulphur recovery unit consisting the Claus plant where H<sub>2</sub>S is converted to elemental sulphur or sulphuric acid. The regenerated lean solvent from the bottom of the regenerator is cooled by a heat exchanger against the rich solvent followed by water cooling before being sent to the top of the absorber for the next absorption cycle.



Figure 7.8 Typical flow sheet of an absorber based AGR unit

Physical solvents are organic solvents having affinity for acid gases. The physical solvents generally require high partial pressure of the acid gases in the syngas as well as low operating temperature for good separation. On the contrary the chemical solvents can operate at near ambient temperature and are more effective for low acid gas partial pressure compared to the physical solvents. The physical solvent based processes tend to also co-adsorb more CO<sub>2</sub> than chemical solvent based AGR. In order to reject CO<sub>2</sub> to concentrate sulphur acid gas from the regenerator overhead to 15-25% level, as typically required for feeding the Claus SRU, multiple step depressurization of the rich solvent with supplemental nitrogen purging is required. Because of the requirement of refrigeration along with more complex solvent regeneration the physical solvent based AGR becomes 2-3 times costlier than Chemical solvent based AGR.

For chemical synthesis applications, that require syngas with <1 ppmv sulphur, physical solvent based process such as Rectisol (methanol) and Selexol (dimethyl ethers of polyethylene glycol) are often the preferred choice. This ensures essentially total removal of sulphur ( $H_2S+COS < 0.1$  ppmv). The process also remove traces of HCN,  $NH_3$  also trace metal contaminants to provide additional downstream catalyst protection.

A wide range of absorption processes is used for the removal of acid gases from syngas as stated below:

#### **Chemical absorption:**

In this process acid gas components react with the solvent molecules and dissolve in the solvent. The following chemicals are used for the chemical absorption of the acid gases from the syngas.

- (i) Monoethanol amine (MEA)
- (ii) Di-ethanol amine (DEA)
- (iii) Methyl-diethanol amine (MDEA)
- (iv) FLEXSORB (hindered amines)

#### Physical absorption:

In this process syngas components are physically absorbed into the solvent molecule. The important physical absorbents are di-methyl-ethers of polyethylene glycol (Selexol) and refrigerated methanol (Rectisol).

#### Physical & Chemical absorption:

Some compounds like Sulfinol and amisol absorb acid gas components by physical attachment as well as chemical reactions.

Amongst these processes, MDEA, Selexol and Rectisol are widely used in many commercial gasifiers. The characteristics of these mostly used absorption process for acid gas cleaning is summarized in Table 7.8
Solvent &	Removal	Process	Quality of	Developed	Remarks
Process	(%)	parameters	treated gas	bv .	
MDEA	H <sub>2</sub> S:98	T: 30-35 ° C	H <sub>2</sub> S:10-20	Union	Lowest capital cost. Moderate
(Chemical)	_99		ppmy	Carbide	operating temp. Only limited
	0	(Amhient)	PPIII		nhysical COS absorption takes
	$20^{2}$				physical cos absorption takes
	50	5			place
		P:< 30		Chemical,	
		kg/cm²		Shell	
Selexol	H₂S :	T:-7 to-4°	H₂S: < 30	Allied	Higher cost than MDEA but
(Physical)	99	С	ppmv	Chemical	overall system cost including
	CO <sub>2</sub> :	P: 70		Corp.,	sulfur recovery (SR) process and
	Variable	kg/cm <sup>2</sup>		Union	tail gas treating (TGT) could be
				Carbide,	more cost effective
				UOP	
Rectisol	H <sub>2</sub> S:	T:-35 to-60	H <sub>2</sub> S :< 0.1	LINDE AG	Highest cost,
(Physical)	99.5 -	°C	ppmv		High selectivity for H <sub>2</sub> S over CO <sub>2</sub> ,
	99.9	P: 82	CO <sub>2</sub> :		Ability to remove COS
	CO <sub>2</sub> :	kg/cm <sup>2</sup>	several		
	98.5		mole % to		
			few ppm		

Table 7.8 Comparison amongst the mostly used absorption methods for acid gas removal

The energy demand of physical absorption processes is predominantly caused by compression and pumping of solvent. It can be as low as 0.03 kWh per kg of carbon dioxide removed from a gas under elevated pressure. Chemical absorption processes need heat for regeneration, which strongly depends on solvent concentration in the aqueous solution. At high pressure (pressurized gasification), chemical absorption requires around 0.11 kWh/kg CO<sub>2</sub>.

The absorbed  $H_2S$  gas can be desorbed at elevated temperature and the elemental sulfur can be recovered by the Claus process, which consists of multistage catalytic oxidation of hydrogen sulfide according to the following overall reaction:

 $2H_2S + O_2 \rightarrow 2S + 2H_2O$  (i)

Each catalytic stage consists of a gas re-heater, a catalyst chamber, and a condenser. The Claus process involves burning one-third of the  $H_2S$  with air in a reactor furnace to form sulfur dioxide (SO<sub>2</sub>) according to the following reaction:

 $2H_2S + 3O_2 \rightarrow 2SO_2 + 2H_2O$  + heat (ii)

The furnace normally operates at combustion chamber temperatures ranging from 980 to 1540 °C with pressures rarely higher than 0.7 kg/cm<sup>2</sup>. Before entering a sulfur condenser, hot gas from the combustion chamber is quenched in a waste heat boiler that generates high to medium pressure steam. About 80 percent of the heat released could be recovered as useful energy. Liquid sulfur from the condenser runs through a seal leg into a covered pit from which it is pumped to trucks or

railcars for shipment to end-users. Approximately 65 to 70 % of the sulfur is recovered. The cooled exit gas from the condenser is then sent to the catalyst beds. The remaining uncombusted two-third of the hydrogen sulfide undergoes Claus reaction (reacts with  $SO_2$ ) to form elemental sulfur as follows:

 $2H_2S + SO_2 \rightarrow 3S + 2H_2O + heat$  (iii)

The catalytic reactors operate at lower temperatures, ranging from 200 to 315°C. Alumina or bauxite is sometimes used as a catalyst. Because this reaction represents an equilibrium chemical reaction, it is not possible for a Claus plant to convert all the incoming sulfur compounds to elemental sulfur.

In addition to the oxidation of  $H_2S$  to  $SO_2$  and the reaction of  $SO_2$  with  $H_2S$  in the reaction furnace, many other side reactions can and do occur in the furnace. Several of these possible side reactions are:

 $CO_2 + H_2S \rightarrow COS + H_2O$  (iv)  $COS + H_2S \rightarrow CS_2 + H_2O$  (v)  $2COS \rightarrow CO_2 + CS_2$  (vi)

From the condenser of the final catalytic stage, the process stream passes to some form of tail gas treatment process. The tail gas, containing  $H_2S$ ,  $SO_2$ , sulfur vapor, and traces of other sulfur compounds formed in the combustion section, escapes with the inert gases from the tail end of the plant. Thus, it is frequently necessary to follow the Claus unit with a tail gas clean-up unit to achieve higher recovery.

Conventional solvent based AGR units, both of amine and physical solvent types and conventional Claus sulfur recovery (SR) units with their associated tail gas treating (TGT) units, are easily capable of meeting the fairly stringent emissions regulations.

# 7.9 Adsorptive Separation of Acid Gases

Some adsorbents like ZnO/CuO,  $Cr_2O_3$ ,  $Al_2O_3$  etc. can adsorb the acid gas components. In case of adsorption processes H<sub>2</sub>S is converted to metal sulfide (315-530 ° C), which produces SO<sub>2</sub> during regeneration at 590-680 °C. The equations for the conversion of sulphur in the above processes are described below:

Desulfurization

 $ZnO + H_2S \rightarrow ZnS + H_2O (315-530 \degree C)$ 

Regeneration

 $ZnS + 3/2 O_2 \rightarrow ZnO + SO_2 (590-680 ° C)$ 

These metal oxides also adsorb  $CO_2$ . Cited energy requirements for adsorptive removal of  $CO_2$  is in the range of 0.16 to 0.18 kWh/kg  $CO_2$  at  $CO_2$  concentrations of 28 to 34 mole % in the feed gas and 0.55 to 0.7 kWh/kg  $CO_2$  at a  $CO_2$  concentration of 10 to 11.5 mole %.

Research Triangle Institute (RTI) with DOE/FETC sponsorship has been developing zinc titanate sorbent technology since 1986. In addition, RTI has been developing the direct sulfur recovery process (DSRP) with DOE/FETC sponsorship since 1988. Fluidized-bed zinc titanate desulfurization coupled to the DSRP is currently an advanced, attractive technology for sulfur removal/recovery for IGCC systems.

The DSRP unit consists of essentially two fixed-bed catalytic reactors. A sulfur condenser follows each reactor. Without cooling, the regenerator off-gas is mixed with the coal gas slipstream and fed to the first DSRP reactor, which operates at 649 °C and 21 kg/cm<sup>2</sup>. Approximately 95 % of the sulfur gas in the inlet stream of the first reactor, is converted to elemental sulfur. Controlling the stoichiometric ratio at the inlet of the reactor can optimize this conversion. The outlet gas of the first DSRP reactor outlet composition can be controlled, in order to obtain the H<sub>2</sub>S/SO<sub>2</sub> = 2:1 ratio. The cooled gas stream is passed to the second DSRP reactor where 80–90 % of the remaining sulfur compounds are converted to elemental sulfur at 399 ° C by the high pressure Claus reaction. The total efficiency of the two reactors for the conversion of sulfur compounds to elemental sulfur is about 99.5 %.

The chemical reactions involved are:

#### Reactor 1.

 $H_2 + 1/2SO_2 \Leftrightarrow H_2O + 1/4S_2$   $H_2 + 1/3SO_2 \Leftrightarrow 2/3H_2O + 1/3H_2S, 500-650 °C, 20 \text{ atm.}$   $CO + 1/2SO_2 \Leftrightarrow CO_2 + 1/4S_2$   $S_2 \Leftrightarrow 1/4S_8$   $CO + 1/2S_2 \Leftrightarrow COS$ **Reactor 2.** 

 $2H_2S + SO_2 \Rightarrow 3/nS_n + 2H_2O$ , 250–400 °C and 20 atm.

CLAUS with *n* = 1, 2, . . . , 8

The main advantage of this process is the practically complete energy coupling with the sorbent regeneration system. This is because  $SO_2$  is catalytically reduced to elemental sulfur using a small slipstream of the coal gas at the pressure and temperature conditions of the regenerator off gas. Also, elemental sulfur recovery is an attractive option because sulfur can be easily transported, sold, stored, or disposed of. Nonetheless, the process is difficult and hazardous to operate due to the presence of this elemental sulfur. Furthermore, the expensive catalytic reducing agents used penalize the operating costs.

High sulfur coal gasification plants that use COS hydrolysis, together with conventional AGR and SR units, have been able to achieve nearly 98 % sulfur recovery, equivalent to sulfur emissions of about

0.10 lb/MMBtu of coal input. By also using TGT units, even higher sulfur recoveries, up to 99.8 %, can be achieved.

The use of warm gas cleaning techniques for desulfurization of syngas is rare in the world. Its future prospect is also not bright because of mercury, which is very difficult to remove at high temperature. However, research is on progress to develop direct sulfur recovery process (DSRP), where desulfurization of syngas and recovery of sulfur is performed simultaneously. RTI has developed improved catalyst for the desulfurization of syngas in warm state in the transport gasifier.

# 7.10 Selection of AGR process

During the selection of a suitable AGR process the following points need to be considered.

- (i) Raw syngas composition, its temperature and pressure
- (ii) Target syngas purity
- (iii) Process complexity, utility requirements and cost
- (iv) Corrosion and degradation/solvent loss





Figure 7.10 Suitability of AGR process at various conditions Sulfur in product gas, (ml/m<sup>3</sup>)

The rectisol process is used in maximum plants for AGR. Rectisol acid gas cleaning unit consists of an absorption section for the removal of sour gas compounds and desorption section for the regeneration of lean solvent as shown in Figure. The column absorber is operated at high pressure and low temperature, in which sour gas compounds ( $H_2S/COS$  and  $CO_2$ ) are removed by methanol. The treated gas is routed to downstream process units. It is absolutely water free and free of

impurities. To recover the valuable product compounds like  $H_2$  and CO, the loaded methanol is first depressurized, to flash small amount of the co-absorbed components.



Figure 7.10.1 Rectisol process flow sheet

This flash gas is routed back to the feed gas line to recover the products. Due to a further pressure reduction,  $CO_2$  is flashed from the loaded methanol in the  $H_2S$  enrichment column.  $N_2$  is used as stripping gas to improve reduction of  $CO_2$  in the methanol. The flashed  $CO_2$  is sent for sequestration, while the stripped  $CO_2$  (including stripping  $N_2$ ) is vented to atmosphere as tail gas. Methanol from the sump of  $H_2S$  enrichment column is routed to the hot regeneration column. The  $H_2S$  and remaining  $CO_2$  are stripped by means of methanol steam, generated in the re-boiler. The  $H_2S$  fraction is routed to a Clause plant for sulfur removal. Lean methanol is collected from the bottom of the column, cooled down and routed back to the absorber column as regenerated washing agents. Carbonyl sulfide (COS), which is usually present at a several hundred ppmv level in syngas from coal is difficult to remove quantitatively in AGR units. Further removal of sulfur may therefore be accomplished by the addition of a COS hydrolysis unit (before the AGR), which catalytically converts COS to  $H_2S$ , which can be easily scrubbed out in the AGR unit.

# 7.11 H<sub>2</sub> production

After the removal of acid gas components from the shifted syngas it contains mainly  $H_2$ . However, some impurities like CO, CO<sub>2</sub>, and  $H_2S/SO_2$  may be present in minute amount. The available technologies for the purification of hydrogen are:

- (i) Pressure swing adsorption (PSA)
- (ii) Membrane technology

The PSA units are developed exploiting the properties of adsorbents to adsorb more impurities at high pressure than at low pressure. A pressure swing cycle of short duration helps the adsorption of impurities on to adsorbent from the feed gas to produce pure hydrogen and their desorption in the purge gas. The basic flow diagram of the PSA is shown in Figure 7.11

Hydrogen product (high purity)



# Fig. 7.11 Basic flow diagram of the PSA for H<sub>2</sub> purification



Typical pressure profile changes at various internal steps of PSA is shown in Figure 7.11.1

#### inte

Fig. 7.11.1 Pressure profile trend of typical PSA cycle

Various types of adsorbents are used in the adsorption column for the purification of hydrogen in a PSA unit. Some adsorbents commonly used in a PSA column and the impurities adsorbed by these are mentioned in Table 7.11

Adsorbent	Impurity adsorbed
Silica gel	Ethane, propane, butane and heavier hydrocarbons
Activated carbon	Methane, carbon dioxide
Molecular sieve 5A	Methane, carbon monoxide, nitrogen
Activated alumina	Water

Table 7.11	Adsorbents	for PSA	and removal	l of imr	ourities

Molecular sieve is used at the top layer of the adsorption column in PSA unit along with consecutive layers of various adsorbents below this layer.

Capacity of the PSA unit can be improved by controlling the following factors:

(i) Using high performance adsorbents

High performance adsorbents would have high selectivity towards CH₄ or CO, higher density and higher loading capacity.

(ii) Using faster PSA sequence

A faster PSA sequence indicates the shorter adsorption time and longer purges. It also includes maximum number of equalization.

- (iii) Using large adsorber vessels
- (iv) Using more adsorber vessels

Where high purity hydrogen is required, the hydrogen product may be separated via palladium diffusion membrane. In addition to the use of palladium diffusion membranes, which have a high selectivity for hydrogen separation, micro porous ceramics have also been investigated. A measure of success has been achieved, for example, using micro porous silica, which has a much lower cost than Pd although a lower hydrogen selectivity.

Polymeric membranes, which presently attain  $CO_2/H_2$  selectivity up to 10, can also be used to separate  $H_2$  from syngas. Ceramic membranes achieve  $CO_2/H_2$  selectivity up to 15 on laboratory scale, expensive palladium metal membranes even achieve 100 at temperatures between 300 and 400°C. Application of membrane reactor (MR) technology brings to the WGSR the ability to operate at significantly lower  $H_2O/CO$  ratios (1–2 compared to 9.8), integrated gas separation, the generation of high purity  $H_2$ , and conversions above the equilibrium limit.

# 8 CARBON CAPTURE, UTILIZATION AND SEQUESTRATION IN COAL-BASED HYDROGEN SYSTEMS: BRIEF OVERVIEW

A large amount of CO<sub>2</sub> emission to the tune of 10-15Kg/kg of Hydrogen is generated in producing Hydrogen from Coal through Coal gasification route. Thus, to make Coal to Hydrogen a sustainable alternate to meet Hydrogen requirement, the effective utilization or sequestration of CO<sub>2</sub>by adopting suitable technology (CCUS) technology is going to play a major role.Globally, the largest usages of CO<sub>2</sub> are for enhanced oil recovery (EOR) and recycling CO<sub>2</sub> infertilizer production for urea but the same is difficult to adopt in India as most of the oil fields are located on the western side and coal field are located on the opposite i.e eastern side and hence such applications shall require laying of large cross-country pipelines. Further, utilization in fertilizer shall require large capex investment upstream i.e for production of ammonia, which is must for converting CO<sub>2</sub> into urea.

TERI reported that a comprehensive study undertaken for identifying  $CO_2$  sequestration potential in off-shore and onshore in India in 2009 revealed that potential storage of 345 MtCO<sub>2</sub> in majorcoalfields and 2–7 GtCO<sub>2</sub> in oil and gas reservoirs exist in India. These storage sitestend to be relatively small and disparate. Even if some sites are considered for sequestration, many of the coal seams would still be in use for many years, preventingtheir use as  $CO_2$  storage sites. Additionally, there are some concerns that some of the coal seams in India aretoo shallow to provide a long-term  $CO_2$  storage option. Thus, detailed studies may be required before considering carbon sequestration as a possible alternate for carbon mitigation strategy. Thus, it is essential that focus ned to be brought on carbon capture and utilization potential.

Technology development for CCU is in nascent stages and different options are being explored by various countries. Some of the technologies being considered are:

- **CO<sub>2</sub> to Methanol**: This requires large Hydrogen. Further, demand of Methanol in the country is very small in comparison to the volume of CO<sub>2</sub> likely to be generated from Coal gasification. A number of R&D programs are being undertaken by companies like CIMFR, BHEL, IIT-Thermax, EIL and IOC but proven technologies are yet to be established.
- CO<sub>2</sub> to Bio-Ethanol: Gas fermentation technology has been developed for converting CO rich gases or CO<sub>2</sub> with hydrogen support into Bio-Ethanol through Gas Fermentation Technology. Some plants in Germany and China have been established and are in operation. One plant is under execution in India also. As Bio-Ethanol production is the Government of India priority also to reduce import of crude oil, this can be explored as a potential option for CO<sub>2</sub> utilization.
- **CO<sub>2</sub> to Carbon Black and Graphite Electrode:** Technology up to TRL-6 level have been developed by a company Solid Carbon in USA for converting CO<sub>2</sub> into Carbon Black and Synthetic Graphite for Electrodes. However, this process requires large electricity and thus can be considered if renewable power is available. No commercial pilot plant has been set up yet.
- CO<sub>2</sub> to carbon flakes: RMIT Australia is working on a technology for splitting CO<sub>2</sub> into Carbon flakes and Oxygen using special alloy of gallium and Indium. Although, technology has been tested at laboratory scale and pilot testing are in progress at industrial scale, commercialization of the technology may take another few years. Once developed, this can and convert the same into

- Dry Reforming of CO<sub>2</sub>: Several steel technology suppliers are working on Dry reforming of CO<sub>2</sub> using Methane, either recovered from Coke Oven Gas or taken from natural gas intoSyn Gas i.e CO and Hydrogen for use in Steel making. Presently, one syngas based steel amking plant is already in operation in India and if such technologies are fully developed, this can provide opportunity not only for bulk recycling of carbon but also minimize the cost of production of various chemicals by using CO2 as the main feedstock. This technology is also in pilot scale and yet to be tried on commercial scale.
- CO<sub>2</sub> Electrolysis: Solid oxide electrolysis cell (SOEC) has been developed and are used commercially for producing carbon Mono Oxide from Carbon Oxide. The technology is known as eCO technology but presently used on a smaller scale for producing CO. The carbon mono oxide so produced is of more than 99.95% purity and can either be used in Iron and Steel making for reducing Iron Ore as a substitute of coal and thereby minimizing the import of Coking Coal or alternatively can be used for producing Bio-ethanol using gas fermentation technology. A simplified flow sheet of eCO is shown below in Figure-8.0:



Figure-8.0: CO<sub>2</sub> conversion into CO

By combining electrolysis with low-carbon energy sources, water and CO<sub>2</sub> can be converted into chemical feedstock with a minimal or even negative carbon footprint

Looking into present status of commercial application of various CCU technologies mentioned above, it is observed that Electrolysis of CO combined with Bio-Ethanol production using CO and  $CO_2$  may be provide a long-term sustainable solution not only for bulk utilization of captured  $CO_2$  but also providing Hydrogen for different application at the targeted price of USD 1-1.5 per kg using domestic coal resources in comparison to green or SMR based Hydrogen. In future, other technologies , as and when developed , can be introduced depending upon techno-economic analysis.

The carbon that is released from the burning of the natural gas is then recycled through a series of steps that heat, cool, and compress it to result in low-cost and emission-free electricity. It may be possible to extend this to the  $CO_2$  produced out of Coal-to-hydrogen Water-Gas-Shift systems since the  $CO_2$  produced would be as clean as or cleaner than what one might expect from natural gas combustion. This could be a potentially exciting option for India though detailed due diligence would be needed.

## 3. Quest Carbon Capture and Storage by Shell (Canada)

Quest is Shell's carbon capture plant at their Scotford Upgrader power plant in Alberta, Canada, where they make crude oil from the bitumen found in sand. The oil-making process requires

hydrogen to make the oil lighter but – as with the proposed coal-to-hydrogen initiative for India – it is accompanied by carbon dioxide emissions.

Quest's capture technology is conventional, using a commercially available amine to absorb  $CO_2$ . The  $CO_2$  is then desorbed from the amine and compressed into a liquid, which is then piped 65km away from the plant through various well sites and then injected over 2km underground, into suitable porous rock formations.

Besides the above, the presentation made by M/s Dastur & Co. showed various technologies commercially available from their technology partners for CCUS as can be seen in the attached Annexures. Besides the Geological sequestration and EOR, they have shown technologies which can convert  $CO_2$  to Chemicals and Aggregates.

# 9 HYDROGEN USAGE

## 9.1 Refinery & Fertilizer

#### Introduction:

As of today, primary consumption of pure Hydrogen in India is in refineries and fertilizer segment which is more than 99%. The details of  $H_2$  consumptions,  $H_2$  sources and consumers within refineries and  $H_2$  generation for balance requirements are described in following sections

## 9.1.1 Utilization of hydrogen in Refining Sector

The Indian refining sector has seen tremendous growth, from a single refinery with a capacity of 0.25 MMTPA in 1901 to 23 refineries with a cumulative capacity of 249.37 MMTPA till date. Given the evolution in demand for middle distillates, which are expected to peak by 2030, a focus in the industry has also been to improve petrochemical yields, making the operations within the refinery more complex.

The production and recovery of hydrogen, in context of refining operations, is essential to processes that convert crude oils to light, high quality products. The product slate of a refinery, therefore, is closely correlated with the availability and consumption of hydrogen. Recent years have been seeing a higher demand for hydrogen from the oil refineries. This is a resultant of several factors such as decreasing quality of crude which are now heavier (have a high C:H ratio) and sourer (have a high sulfuric content) on one end, and, stricter emission norms which require that oil products used by end user have lower levels of impurities. Further, the use of heavier crude oils and more bottom of-the-barrel processing has increased the hydrogen demand in hydrocracking and heavy oil hydrotreating units, while, new specifications for low sulphur fuels signal increased hydrogen consumption in hydrotreaters. At the same time, limits on the aromatics content of gasoline and requirements for oxygenates have led to lower severity in the catalytic reformer and as a result hydrogen production in this unit has decreased over time.

Hydrogen, therefore, constitutes a significant portion of refinery processing and operational cost and is usually regarded as a utility within the industry. The supply and use of hydrogen, as a result, is to the extent possible maintained internally, to avoid constraints on the refinery operations. If the demand for hydrogen, however, exceeds the available supply from within the internal refining processes, then the incremental demand is met by increasing hydrogen plant production.



The Figure below shows the hydrogen using units in a typical refinery.

Figure 9.1.1: Typical refinery configuration indicated with H2 consuming unit

Following are the list of units where H2 are generated and consumed.

## Typically hydrogen is produced in the following Units in Refinery:

- 1. Hydrogen Generation Unit (HGU)
- 2. Catalytic Reforming Unit (As a By-Product) CRU PSA

Hydrogen is consumed in the following Units in Refinery:

- 1. Diesel Hydrotreating Unit (DHDT)
- 2. Diesel Hydro Desulphurization Unit (DHDS)
- 3. Once Through Hydrocracking Unit (OHCU)
- 4. Kero Hydro Desulphurization Unit (KHDS)
- 5. Naphtha Hydrotreating Unit (NHT)
- 6. Isomerisation Unit (ISOM)
- 7. FCC Gasoline Desulphurization Unit
- 8. Vacuum Gas Oil Hydrotreating (VGO-HDT)
- 9. Catalytic Dewaxing Unit

Hydrogen generation plants are based on either steam-reforming of naphtha / natural gas. In most of the refineries, hydrogen production is supplemented by hydrogen recovered from the off gases from different hydrogen consuming processes. These off gases usually contain significant amount of hydrogen along with light hydrocarbons such as methane, ethane and propane etc.

The off-gases with high hydrogen purity can be used directly as hydrogen source, if the contaminants are within acceptable limits. The off-gases with low purity hydrogen generally are routed to fuel gas header and the streams with intermediate hydrogen purity can be purified provided it is economical. Hence in addition to hydrogen generation and consuming units, hydrogen purification units form an important part of the hydrogen distribution system and establish the overall material balance of hydrogen in the refinery.

# 9.1.2 Refining capacity & hydrogen consumption

The level of hydrogen consumption in case of oil refineries is dependent on several factors including specific density of crude oil; sulphur ratio of the crude oil; the complexity of the refining operations and the quantum of bottom-of-the-barrel refining; the product slate of the refinery; and, the emission norms applicable at the end-user level.

Currently, Indian Refining sector is using approximately 2.1 MMTPA of Hydrogen. Out of the total hydrogen consuming in Refinery, part of the hydrogen gets produced as by-product of Catalytic Reforming Process (~15%) depending on the Refinery Configuration. Rest of the Hydrogen are primarily produced from Steam-Methane Reforming (SMR) +WG Shift of Fossil Fuel. Normally Hydrogen is produced on site for captive consumption. Typical Hydrogen consumption of Refinery is ~ 1% of the Refining Capacity.

To meet the increasing demands of petroleum feeds stock, there are expansions plan most with all refineries as shown in Table 9.1.2 along with estimated H2 demands.

Table 9.1.2: Proposed capacity expansion and H2 demands						
Refineries	2020-21	2024-25	2029-30	H2 demand		
IOCL-Total	80.3	98.3	120.6	0.340		
IOCL-Guwahati	1	1.2	1.2	0.002		
IOCL-Dibrugarh	0.7	0.7	0.7	0.000		
IOCL-Bongaigaon	2.4	2.7	5	0.022		
IOCL-Barauni	6	9	9	0.025		
IOCL-Gujrat	13.7	18	18	0.036		
IOCL-Haldia	8	8	8	0.000		
IOCL-Mathura	8	9.2	9.2	0.010 (*)		
IOCL-Panipat	15	15	25	0.084		
IOCL-Paradip	15	15	25	0.084 (*)		
CPCL Manali	10.5	10.5	10.5	0.000		
CPCL CBR	0	9	9	0.076		
BPCL-Total	27.5	27.5	32	0.038		
BPCL-Mumbai	12	12	12	0.000		
BPCL-Kochi	15.5	15.5	20	0.038 (*)		
Numaligarh Refinery Ltd (OIL)	3	9	9	0.090		
HPCL-Total	15.8	24.5	24.5	0.237		

HPC-Mumbai	7.5	9.5	9.5	0.017
HPC-Vizag	8.3	15	15	0.220
MRPL	15	15	18	0.025
PSU-Total	141.6	174.3	204.1	0.730
BORL - Bina Refinery	7.8	7.8	15	0.061
HMEL	11.3	11.3	11.3	0.000
HRRL	0	9	9	0.076
RRPCL	0	-	60	0.506 (*)
JV-Total	19.1	28.1	95.3	0.642
Nayara	20	20	46	0.219
RIL	68.2	75.7	98.2	0.253
PVT-Total	88.2	95.7	144.2	0.472
Total	248.9	298.1	443.6	1.844

(\*) likely to be explored for sourcing H2 from coal gasification

# 9.1.3 Utilization of hydrogen in Fertilizer sector:

Hydrogen is a key input for production of Ammonia  $(NH_3)$ , which is used as a base material for urea and other complex fertilizers. Hydrogen is produced through SMR+WG Shift with NG as feedstocks in most of the fertiliser plants. Hydrogen is used to produce Ammonia through Haber process.

Ammonia is used for production of Urea and non-urea fertilizer like Diammonium Phosphate (DAP).

Hydrogen consumption in urea production is approximately 0.1 kg/kg of urea. Hydrogen consumption in DAP production is approximately 0.035 kg/kg of DAP.

Currently India consumes 3.1MMTPA of  ${\rm H_2}$  for production of approximately 15 MMTPA of Ammonia.

# 9.2 Hydrogen based Iron & Steel Making

## 9.2.1 Introduction

India is blessed with abundant high-quality Iron Ore, both hematite ( $Fe_2O_3$ ) and magnetite ( $Fe_3O_4$ ) but presently hematite is mainly used in the manufacturing of the Iron and Steel as magnetite resources are yet to be fully explored. The basic process of Iron and steel making involves removal of oxygen by using reducing agents like carbon and hydrogen along with other gangue materials viz. alumina, silica, present as impurities, with the help of various fluxes. In addition to reducing agent, some heat source is also required for carrying out the reaction as well as melting to produce liquid metal. There are two main methods used globally to produce steel from iron ore:

- Primary Route: blast furnace-basic oxygen furnace (BF-BOF); and
- Secondary Route: scrap/direct reduced iron-electric arc furnace/induction furnace (Scrap/DRI-EAF/IF)

More than 70% of the global steel production is through BF-BOF route and only 30% of the production is made by other routes like using scrap or direct reduced iron (DRI). As the carbon is used as a source of heat as well as reducing agent, large amount of Carbon Di-Oxide is emitted in the process and thus the iron & steel sector has been recognized as single largest industrial sector contributing 7-9% of global greenhouse gas emissions. Thus, world over efforts are being made to de-carbonize the sector by using alternate reducing agent like Hydrogen. Presently, direct use of Hydrogen has not been adopted commercially in the Iron and steel sector but a small percentage of steel is produced by using Natural gas or syn gas in which 50-60% Hydrogen (natural gas is reformed using steam to produce H<sub>2</sub> +CO whereas Syn Gas is produced through Coal gasification) is used along with carbon mono oxide (CO) to produce reduced iron in solid state which is converted into molten form using electrical energy.

India is the second largest steel producing country in the world and it is expected that the annual production will increase from present level of around 110 million tons to 255 million tons by 2030 i.e. more than doubled. The coal required for BF-BOF route is called "Metallurgical Coal or Coking Coal" and very limited reserves of such coal is available in India. Thus, more than 80 % requirement of such coal is met by import and in 2020 alone India has imported in excess of 50million tones of coking coal. The availability of Natural Gas is also limited and major demand is met by import only. Natural Gas based steel production through DRI route has been adopted by some of the countries having cheap availability of gas but its contribution is less than 10% globally. Since, India is having large reserves of thermal coal, adopting Syn gas-based steel making can avoid large import of coking coal besides developing a transition route for adoption of Hydrogen in future as and when commercially proven technologies are available.

Hydrogen generation is an energy intensive process and the desired objective of decarbonizing the sector will not be achieved if it is produced using fossil fuel-based grid power. It is estimated that coal-based power plants emit on an average of 915 grams of carbon dioxide (CO<sub>2</sub>) per kilowatt hour of electricity produced. Hence, around 50 Kg of  $CO_2/$ kg of  $H_2$  will be emitted and thus the indirect emission shall be much more than direct emission from BF-BOF as around 70-80 Kg of hydrogen shall be required (theoretically 54 Kg) for each tons of steel. Hence, emphasis is being made to generate Hydrogen using renewable power or using coal by adopting Coal gasification technologies. As, the technologies for Coal to Hydrogen also are in nascent stages, except one pilot project recently been commissioned in Victoria, Australia as a joint collaborative project of Australia-Japan and India has successfully adopted Syn Gas based Iron and Steel making on commercial scale, it may be advantageous if for future expansions Syn Gas based route is adopted with an option to convert into hydrogen at a later date. However, such process also shall require carbon mitigation measures through carbon capture utilization (CCU) as large CO2 is generated in the process of gasification as well as downstream processing for producing Hydrogen through coal.

Although, India is targeting 500 GW of renewable power by 2030 to meet its more than 50% requirement, as committed in COP26 in Glasgow, Hydrogen generation using renewable power provides an opportunity to decarbonize "hard to abate" steel sector. The hydrogen

generated using renewable power is called as "green hydrogen" and steel produced by such sources as "green steel". Although, the use of Hydrogen, particularly green hydrogen, holds the promise of carbon-neutral steelmaking in long term but storage and transportation of Hydrogen poses lot of technical constraints as it is required to be cooled down below minus (-)253 <sup>o</sup>C for transportation. Thus, coal-based Hydrogen along with CCU provides an opportunity for the steel sector to decarbonize as Hydrogen can be generated at site using Gasification and downstream processing technologies and no storage and transportation may be required. Hydrogen based shaft furnace direct reduction is gaining attention as this technology still in the nascent stage and some advanced steelmakers where large renewable power like wind, solar or hydro power is available are pursuing this process with very heavy investment. Numbers of countries like Japan, Australia, and Korea etc. are working on low Carbon Di-Oxide route, but cost of Hydrogen is becoming prohibitive for use of Hydrogen in Iron and Steel making. In case, Hydrogen can be produced economically using large reserves of thermal coal of India, this can not only help in reducing the import of large quantity of coking coal but will also make steel production sustainable and improving competitiveness of Indian steel sector globally.

#### 9.2.2 Reduction Reaction of Iron Oxide

Iron ore reduction in steel production as of now is mainly with carbon monoxide gas, generated from carbon-based reductant. Carbon monoxide reduction reaction (Fe<sub>2</sub>O<sub>3</sub> + 3CO  $\rightarrow$  2Fe + 3CO<sub>2</sub>) separates oxygen from iron ore (Fe<sub>2</sub>O<sub>3</sub>), and CO<sub>2</sub> is generated at this stage.

Hydrogen has the potential to replace fossil fuels in primary production facilities in the iron making - both in existing installations (partial substitution of the fossil fuel input) and in the longer term, in 100% or near-100% hydrogen-based facilities. The reduction of hematite into Iron takes place through a set of reactions as follows:

 $3 \text{ Fe}_2\text{O}_3 + \text{CO} = 2\text{Fe}_3\text{O}_4 + \text{CO}_2$  (Exothermic & takes place @ temp below  $800^{\circ}\text{C}$ )

 $Fe_3O_4 + CO = 3FeO + CO_2$  (Endothermic & takes place @ temp below  $800^{\circ}C$ )

The heat generated during exothermic reaction is sufficient to compensate for heat loss due to subsequent endothermic reaction. In case Hydrogen is used, both the reactions are endothermic and instead of GHG emissions ( $CO_2$ ) is replaced by water vapours as follows:

 $3 \text{ Fe}_2\text{O}_3 + \text{H}_2 = 2\text{Fe}_3\text{O}_4 + \text{H}_2\text{O}$  (Endothermic & takes place @ temp below  $800^{\circ}\text{C}$ )

 $Fe_3O_4 + H_2 = 3FeO + H_2O$  (Endothermic & takes place @ temp below  $800^{\circ}C$ )

At a temperature of between 800-  $1000^{\circ}$ C, the FeO is again converted into Iron (solid) as follows:

FeO +CO = Fe (S) +  $CO_2$  (Exothermic)

If Hydrogen

FeO  $+H_2 = Fe(S) + H_2O$  (Endothermic)

Iron produced by using Hydrogen is called as Direct Reduced Iron (DRI) or Briquetted Iron as further melting is not possible into liquid stage. However, when carbon is used as a reducing agent as well as fuel source, further melting takes place and Iron is produced as a liquid metal along with other element like carbon, silicon, manganese etc.

# 9.2.3 Use of Hydrogen in Iron Making

From the above, it is clear that Hydrogen can also be used as a reducing agent but the main issue is of endothermic reaction which shall require some external source of heat for maintaining reaction temperatures. The idea of using hydrogen as a reductant is primarily related to reduction in  $CO_2$  emission in steelmaking.  $CO_2$  emissions results from the use of carbon for both the energy and the chemical reduction needed, the major contributor being the blast furnace, in which the solid iron ore, in the form of lump, sinter or pellet, is transformed into liquid pig iron. The basic concept of hydrogen ironmaking is to substitute C (or CO) reductant with H<sub>2</sub>,

Thus, hydrogen as a reducing agent separates oxygen from iron ore  $(Fe_2O_3)$ , producing iron (Fe) together with water (H<sub>2</sub>O) as shown in Fig. 9.2.3 Replacing the coal/coke with hydrogen will not generate any CO<sub>2</sub>, a greenhouse gas. Harmless H<sub>2</sub>O is formed, instead of the greenhouse gas CO<sub>2</sub>.



Fig. 9.2.3: Reduction Reaction of iron ore with hydrogen and carbon monoxide

The majority (90%) of iron from ore is produced using the blast furnace, which produces liquid iron saturated in C. The other alternative is the direct reduction (DR) processes, whose product is solid iron (DRI-direct reduced iron, also named sponge iron, or HBI–hot briquetted iron). DRI production is either coal based or gas based. Gas based processes are more common globally, though coal based DRI plants are predominant in India due to non-availability of gas. In the gas-based process, the reduction occurs as a series of gas-solid reactions as shown above with the reactant gases being CO and H<sub>2</sub>. The reactor is generally a vertical shaft furnace, whose reducing gas (CO-H<sub>2</sub> mixture) is obtained by natural gas reforming. Common gas-based processes for DRI production are MIDREX and HYL-ENERGIRON whereas Rotary kiln furnaces are employed for coal-based process.

Thus, there are two ways in which hydrogen can be used in steel production:

- As an auxiliary reducing agent in the BF-BOF route (H<sub>2</sub>-BF)
- As the sole reducing agent {direct reduction of iron ore by hydrogen

DRI (H2-DRI)}

Possibility of substituting 100% of the carbon (monoxide) with H<sub>2</sub> could be envisaged in case of DRI production through gas-based shaft furnace. In case Syngas's is used instead of only Hydrogen (as presently being used in Gas based DRI or Gasification based DRI), reduction of iron oxide takes place involving two key reactions of CO and hydrogen. The reduction with CO is exothermic, and that released energy helps drive a parallel endothermic reduction by hydrogen. Switching to a hydrogen-only system significantly changes this thermodynamic balance, so the hydrogen must be preheated before it enters the furnace. Looking into large reserves of domestic thermal coal, it is advantageous to bring focus on Syn gas based production instead of conventional BF-BOF process.

## 9.2.4 Carbon Emission in Steel Sector

The carbon footprint of world average steel is about 1.85 t  $CO_2$ -e/t of steel against 2.5-2.8 t  $CO_2$ -e/t of steel in India. Various countries and regions are pursuing research to develop new technology initiatives to reduce  $CO_2$  emission in the iron and steel making. In BF/BOF route, Coal is one of the major contributors in the cost of production of steel and constitutes nearly 40% of the total cost besides it is the source of GHG emission. Typical GHG emissions in various steps of BF-BOF route is shown in Figure-9.2.4 below:



Figure-9.2.4: GHG reduction in BF-BOF process

To minimize GHG emission, efforts are being made world across to find alternate reducing agent and Hydrogen is being considered as the most preferable choice. Total global steel production is forecasted to grow by 30% from present level of 1860 million tons to about 2.2 billion tonnes by 2050. In 2019 the contribution of steel sector was 2.6 Gt of  $CO_2$  out of world total emission of 33.1 Gt i.e. around 7% and it is expected that if the same trend continues (business as usual), the total  $CO_2$  emission from the industry may cross more than 4.3 Gt by 2050 as shown in Figure 9.2.4 (1), forecasted by the International Energy Agency.



Figure 9.2.4 (1) : Projected CO<sub>2</sub> emission by Global Steel Sector (Source: IEA)

Although, trials at 1 T/hr steel production are under way using green hydrogen and efforts are also being made to inject green hydrogen in Blast Furnaces as well as increase hydrogen content in gas based DRI, it is expected that commercial hydrogen-based technology may be available by mid of the century and thereafter only true green steel may be produced. In order to limit global warming to well below 2°C, net anthropogenic greenhouse gas (GHG) emissions must reach zero between 2050 and 2070 and become negative thereafter.

# 9.2.5 Heat Requirement in Iron reduction

The energy consumption in BF/ BOF, DRI (assuming MIDREX)/EAF and likely in Hydrogen route is indicated below:

S.No	Route	Energy Needed
1.	Standard BF-BOF Route	18.8 GJ/tHRC (mostly Coal)
2.	DRI +EAF	15.6 GJ/tHRC (Gas +Electricity)
3.	Hydrogen based route +EAF	14.7 GJ/tHRC (mostly electricity)

From the above energy comparison, it can be observed that energy consumption for the two direct reduction routes is similar, slightly lower than that of the standard BF-BOF route, and the hydrogen-based route is at the same level as the natural-gas-based route. But it is CO<sub>2</sub> emission which may be reduced considerably if Hydrogen becomes techno-economically viable option and Hydrogen as well as Electricity both are generated by renewable means instead of fossil fuel. If fossil fuel is used as a source of Electricity generation, reduction in GHG emission in comparison to DRI (gas based) +EAF may not be significant. Presently, hydrogen production via steam methane reforming (SMR) is the most popular method but in this process both hydrogen and carbon dioxide are produced. Thus, the hydrogen produced is called as "grey hydrogen".

## 9.2.6 Hydrogen Utilization in the Blast Furnace

World over efforts are being made to inject Hydrogen enriched gases or Hydrogen directly inside the Blast Furnace to reduce GHG emissions but such technologies are yet to be employed on a commercial scale. One of the major Blast Furnace supplier SMS-Paul Wurth claimed to have successfully completed trial runs of injecting Hydrogen enriched gases in the Blast Furnace and the same can be implemented in the existing as well as new furnaces. While substitution of expensive Coke with lower-cost fuels such as Pulverized Coal Injection (PCI), Coke Oven Gas (COG), Natural Gas (NG), and Tar (etc.) contributes to reducing OPEX, COG and NG are particularly interesting for CO<sub>2</sub> emission reduction due to their higher hydrogen content. As India is having large reserves of thermal coal, injection of Hydrogen enriched syn gas into Blast Furnace provides large opportunities for minimizing dependency on imported coking coal as well as reducing CO<sub>2</sub> emission.

## 9.2.7 Syngas as an Enabler for Integration of Green/ Grey Hydrogen

With a focus on green steel manufacturing, hydrogen is the most promising reducing gas when considering fossil fuel replacement. Many steelmakers are currently investigating strategies for hydrogen integration in the steel production chain. With proposed reduction of iron ore using hydrogen in DRI facilities, hydrogen combined with syngas injection and top gas recycling can serve as a viable transitional step in current conventional blast furnace operation. The combination of syngas shaft injection with H<sub>2</sub>, NG or hot syngas injection to the tuyere, depending on hydrogen availability has the potential to reduce CO<sub>2</sub> emissions by 34% without hydrogen and 48% with hydrogen. These CO<sub>2</sub> reductions can further be increased by adding carbon capture technology and substituting PCI with charcoal. As, steel production capacity in India need to be doubled from 145 million tonnes to 300 million tonnes by 2030 to meet continuously increasing demand of steel and the very fact that no other proven technology is available as on date, it is worth considering to stall new blast furnace with provision of Hydrogen / Syn gas through Coal gasification, initially Syn Gas only injection can be considered as a transition to shift to Hydrogen. Figure



The suggested methodology is shown in Figure-9.2.7 below:

Figure 9.2.7: Different injection possibilities for the enhanced blast furnace

#### 9.2.8 Challenges in Hydrogen utilization in BF

It is not feasible to use only hydrogen in a BF for reduction, as operational requirements limit higher  $H_2$  substitution and full  $H_2$  operation. Increasing  $H_2$  concentration will gradually transform the exothermic reaction to endothermic, lowering down the temperature and disturbing the basic chemistry inside the BF, and influence temperature, gas utilization rate, reaction rate etc.

**Pre-heating of hydrogen**: Reduction with carbon is exothermic but that with hydrogen is endothermic, causing a temperature decrease. Pre-heating of hydrogen will be necessary for the large amount hydrogen injection.

**Effect on Gas permeability:** BF is a reactor that continuously performs heating, reduction, and melting. Less carbon (coke) and more hydrogen input to BF will lead to less coke support, which causes less gas permeability, and less contact with high-temperature gas, making it difficult to melt. Ensuring maximum gas permeability for stable reaction and melting with less coke in the BF will be required.

Use of hydrogen can be a good way to achieve partial substitution of fossil fuels, while still using existing equipment.  $H_2$  in BF has the potential to reduce emissions in coke plant also as it reduces the amount of coke needed. Hydrogen usage in BF can be considered as a transition towards  $H_2$ -DRI to provide emission reductions in the near-term.

## 9.2.9 Hydrogen Utilization in Direct Reduced Iron (DRI) Iron making

The H<sub>2</sub>-DR route is derived from direct reduction, a well-established process, which is usually operated with natural gas or coal. Natural gas-based direct reduction (NG-DR) is a proven technology. Nearly around 100 million Tonnes of Iron is produced by this route which is finally converted into desired grades of steel using Electric Arc Furnaces. In this process, natural gas is reformed into CO + H<sub>2</sub> and presently 50-60% Hydrogen based gas is used in this process. Alternately, Syn Gas produced through Coal Gasification can be used as the composition of the gas is same as of reformed natural gas but this technology requires additional facilities like , Gasifiers, Gas Cleaning , Air Separation unit etc and makes the project capital intensive. First such plant in the world has been set up in India and is operating successfully. In addition, the main issue is of  $CO_2$  emission which need to be addressed by suitable carbon capture and utilization scheme.

Efforts are being made to increase Hydrogen percentage in the Syn gas / reformed Natural Gas so that in future such facilities can be converted into Hydrogen based iron making as and when such technologies become techno-economically viable. However, Gas/Syn Gas based DRI process can be utilised as an entry point for  $H_2$ -DR. The operating gas mixture could be gradually enriched with hydrogen, but its share is limited by hydrogen availability, emissions, costs, and process requirements. This enables a very high degree of flexibility, which can pose a strong strategical advantage.

The utilisation of hydrogen accelerates the reduction process (in comparison to the usage of coke as a reducing agent). Due to the endothermic reaction, heat must be added in the process. The additional heat can be provided by burning excess hydrogen or using electricity.

The off-gas of this process is mainly water vapor, which could be used for hydrogen production to improve the energy efficiency of water electrolysis. The product of this process is a carbon-free direct reduced iron (DRI) or sponge iron with an iron content of approximately 95% and no carbon content.

100% Hydrogen based steelmaking was tried earlier also and hence this is not a new concept. Cleveland-Cliffs, Lurgi and LTV Steel built a 400,000 ton/year "Circored" (Metso-Outotec, Circored process) direct reduction plant in Trinidad that used hydrogen from a steam reformer as its reductant and energy source. The plant was started up in 1999, but the fluidized bed reactor had numerous problems and it produced only about 150,000 tons by the time it was shut down in 2001. The process was based on use of fine ore instead of pellets to the traditional blast-furnace/basic oxygen furnace (BF/BOF) steelmaking route and to the DR route (CO/H<sub>2</sub> based reduction in shaft furnaces using DR-pellets) and completely eliminates the need for expensive and energy intensive pelletizing. The typical flow sheet of Circored I shown in figure-9.2.9





#### 9.2.10 Midrex Hydrogen Based DRI Technology

There has been increasing focus to mitigate  $CO_2$  emissions in the iron and steel industry. The natural gas-based MIDREX Direct Reduction Process paired with an electric arc furnace (EAF) has the lowest  $CO_2$  emissions of any commercially proven steel-making route based on Iron Ore; yet, there is even more room for lower emissions through use of hydrogen as a fuel and chemical reactant. The best possibility for drastically reducing the steel industry's  $CO_2$  footprint is the use of pure hydrogen as the energy source and reductant for direct reduced iron (DRI) production in a MIDREX Shaft Furnace. This concept, known as MIDREX H2<sup>TM</sup>, holds

great promise to be developed and realized in either new or existing DRI Plants. A major obstacle to implementing hydrogen direct reduction ironmaking in the difficulty of producing pure hydrogen without a large  $CO_2$  footprint as nearly 55KWh of electricity is required for generating per kg of Hydrogen. In case, fossil fuel-based electricity is used,  $CO_2$  emission will be much more than conventional DRI making process. The typical flow sheet of MIDREX modified process is shown in Figure-9.2.10 and 100% H<sub>2</sub> based in Figure-9.2.10 (1).



FIGURE 9.2.10 MIDREX Process with Hydrogen Addition



FIGURE-9.2.10 (1): MIDREX H2<sup>™</sup> Process

There are a number of considerations for the MIDREX  $H_2^{TM}$  Process, first of which is temperature. With so much hydrogen, the DRI cools off as the reducing gas enters the shaft furnace because of endothermic reaction. Thus, it is necessary to add natural gas to maintain the desired reduction temperature. According to Midrex modelling, the addition of natural gas at a rate of 50 Nm<sup>3</sup>/t DRI should accomplish this.

The second issue is DRI carbon content. The vast majority of DRI is used in EAFs. EAF steelmaking practice today generally employs carbon added either in metallic charge materials such as DRI, HBI and pig iron or as pure carbon. Burning this carbon with injected oxygen creates significant heat which reduces electricity consumption and enables faster melting. Since pig iron is made from BF hot metal that is saturated with carbon, it contains 4-4.5 percent carbon. DRI can have 1-4.5 percent carbon depending on the process, reducing gas used and the way the DR plant is operated. Most EAF steelmakers prefer to use DRI with 1.5-3 percent carbon, but the optimum carbon level varies based on metallic charge mix and the steel grade produced.

With high amounts of hydrogen in the reducing gas, it will be necessary to add hydrocarbons at some place in the process to achieve the desired carbon level. DRI carburizing options include addition of hydrocarbon to the cooling zone or in the furnace lower cone. Addition of 50 Nm<sup>3</sup>/t of natural gas for temperature control results in DRI carbon of about 1.4 percent. The next evolution in steelmaking will be to melt iron without using carbon, but this will be very energy intensive since the melting point of steel increases as carbon content decreases.

Hence, it can be seen that although 100% Hydrogen based Iron making is feasible but it has several associated issues which are yet to be addressed. Under the present circumstances, Syn Gas enriched with additional hydrogen as shown in above Figure appears to be more appropriate than 100% H<sub>2</sub>.

#### 9.2.11 Suggestive Hydrogen Reduction Route for Iron & Steel making

DRI is the most appropriate process where natural gas can be replaced by Hydrogen. Thus, green hydrogen-based DRI and scrap in combination with EAFs can be the only feasible route if Hydrogen is to be used in the Iron and Steel making. All major European steel players are currently building or already testing hydrogen-based steel production processes, either using hydrogen as a PCI replacement in BF/BOF route or using hydrogen-based direct reduction. Since, Hydrogen reduction is endothermic in nature, it becomes difficult to maintain the temperature in BF hearth zone with Hydrogen and possibility of freezing of furnace cannot be ruled out. Similarly, EAF-based steel production may be difficult to operate with renewable power and thus presently achieving carbon neutrality at this point has remote possibility. The most commonly talked Green Hydrogen based Iron and Steel making process is depicted below in Fig-9.2.11 below:



Figure 9.2.11: Hydrogen Based Steel Making

From the above, it can be seen that external energy shall be required for the following:

- As Electricity for producing Hydrogen (Likely Renewable)
- To Heat Hydrogen to a temperature above, likely 900 <sup>o</sup>C
- To pre heat charge, i.e. Pellets, as Hydrogen reduction is endothermic and there is no other fuel charged in the feed
- Supply additional heat during reduction to compensate for endothermic reaction and to maintain reaction temperature
- As Electricity for melting of Briquettes / Scrap in EAF

It is estimated that for a 1 million Ton Steel plant, around 500 MW of renewable energy source is required for generating just Hydrogen only. This shall require not only in setting up of large renewable power sources but also large capacity Hydrogen Electrolyser's or alternate technology need to be developed to meet large Hydrogen requirement of Iron and Steel plants. In addition, there are many issues to overcome for hydrogen to become a major economic factor. For instance, hydrogen has a high energy density by weight, but a low energy density by volume when not compressed or liquefied, thus the high cost of a hydrogen fuel cell has been a major obstacle in its development. Other related issues, such as storage, distribution infrastructure and hydrogen purity and concerns for safety will have to be overcome for the Hydrogen Economy to take off.

# 9.2.12 Global Initiatives in Green Hydrogen Usage in Steel Industry

Many steel companies across the globe have programs under development to transform themselves into green steelmaking operations based on the use of 'green hydrogen' and achieve net zero carbon in steel making. This is mainly because numbers of European countries have started imposing carbon border tax and thus present method of making steel put them in disadvantageous position. However, such policies are yet to be implemented in India. The major Pilot/Demonstration Plants for Hydrogen Steelmaking Trials started across the globe are:

- Thyssenkrupp began trials at one of its blast furnaces in Duisburg, Germany, injecting about 1,000 m<sup>3</sup> of hydrogen per hour into the furnace. It is claimed that this approach could reduce blast-furnace CO<sub>2</sub> emissions by about 20%. Thyssenkrupp intends to reduce its CO<sub>2</sub> emissions by 30% by 2030. It is reported that on 11 November 2019, Thyssenkrupp Steel was the first company globally to inject hydrogen into a blast furnace during operation. A particular focus of the test phase was to gather extensive information on the positioning of the hydrogen lance in the furnace, on flow and pressure conditions. In the second phase, the tests will be extended to all 28 tuyères of the blast furnace. The focus of research will then be on the impact of hydrogen technology on the metallurgical processes in the blast furnace.
- ArcelorMittal, Hamburg (Germany) is developing hydrogen use in an existing commercial natural gas DRI plant (100,000 tons/year).
- SSAB (Sweden) has initiated HYBRIT Project (Hydrogen Breakthrough Ironmaking Technology) for development of hydrogen steelmaking using a newly built direct reduction plant (7,000 tons/year). HYBRIT process aims to replace the coke and other fossil fuels used in traditional, blast furnace—based steelmaking and instead relies on hydrogen created with renewable electricity.
- Baowu Steel Group (China) launched the "China low carbon metallurgical technology innovation alliance" and established a "low-carbon metallurgical innovation research center" for research on the industrialization of hydrogen steelmaking using the existing 400 m<sup>3</sup> test BF in Xinjiang.
- SuSteel project aims to use hydrogen plasma to reduce iron ore. The advantage of this concept is that it produces crude steel from iron ore in one step. SuSteel's hydrogen plasma smelting reduction technology uses electricity to shred hydrogen gas as it passes through a hollow graphite electrode into a conical reactor. This process creates a stream of hydrogen atoms, ions, and molecules at temperatures over 20,000 °C. The plasma melts and reduces finely ground iron ore to create a pool of liquid steel. Pelletizing is not required, and the graphite electrode adds just enough carbon to the metal to form crude steel, so the metal can avoid a trip through an electric arc furnace and proceed directly to secondary steel refining
- HyREX (POSCO Hydrogen Reduction Process): FINEX (Fine Iron ore Reduction), a coal and oxygen-based iron ore reduction process using fluidized bed reactors, has been developed by POSCO. Based on this capability, HyREX process has been designed to use green hydrogen and renewable electricity. The HyREX process is aimed to be developed by 2030 after running pilot plant tests, and demo plant is to be operated by 2040 and commercialization is slated for 2050. FINEX process, where iron ore fine and coal go through the fluidized reduction furnace and the melter gasifier to be processed into molten iron, uses 25% hydrogen and 75% carbon monoxide generated during the process as reducing agents, whereas HyREX will use 100% hydrogen.

## 9.2.13 Challenges in promoting Hydrogen Based Steel Making

Today, hydrogen-based steel production using a combination of DRI and EAF appears to be technically feasible and being considered to be part of a potential long-term solution for decarbonizing the steel Industry. The question is not whether but when and to what extent this transformation will happen. Following external factors may shape the future development and time to adoption of green hydrogen-based steel:

## 1. Power supply

Green hydrogen-based steel creates a need for a significant capacity increase in electricity derived from renewables (for preheating the charge material as well as for EAF besides for Hydrogen generators). To put this into perspective, availability, steady supply, and competitive renewable energy costs are key decisive factors for the technology shift.

# 2. Hydrogen-supply security

The future shift to hydrogen-based steel relies heavily on the broad availability of green hydrogen on an industrial scale. Producing one million tons of hydrogen-based steel requires a green hydrogen amount of 70-80,000 tons i.e. around 70-75Kg /ton of steel. Around 50-55 Kwh of energy is required to produce 1 Kg of Hydrogen. Thus, a capacity of 500-600 MW, is needed to produce this amount of green hydrogen. For a country like India, where steel production capacity is required to be increased from present level of 140 million tons to around 300 million tons in the next 10 years to meet consumption demand, considering such Hydrogen based steel plants at present may put India into disadvantageous position and India may become an import based country from presently of export based country. This is mainly because of the fact that providing the required production capacity and infrastructure for hydrogen-based steel production on a large scale has a significant impact on the timeline for the commercial availability of hydrogen-based steel.

#### 3. Raw material

To switch production from BF/BOF to DRI/EAF using hydrogen, raw material changes are necessary and will especially increase demand for DR pellets. The security of DR supply in the case of a massive switch to hydrogen-based steel production is uncertain and could result in rising price premiums, negatively affecting the economics of the new production method. This shall require large investment in Beneficiation and Pelletization industry.

#### 4. Production technology

The basic production method for DRI/EAF powered with natural gas is already established and working on a large scale in certain markets that benefit from an abundant supply of cheap natural gas. Moving forward, switching the process to an entirely hydrogen-powered process is technically feasible, although the overall cost is still high, and the technology has yet to be proven on a large scale. Based on the present research going on across the globe, any techno-economically viable technology for Hydrogen before 2040-50 appears to be remote possibility. On the upside, however, it is considered relatively easy to switch a DRI/EAF production method powered by natural gas over to hydrogen.

## 5. Willingness to pay

Considering steel's vital role in the global economy, customer support, acceptance, and eventually demand are required for the success of green hydrogenbased steel. Only if customers value carbon-reduced/neutral products, and are willing to pay for decarbonisation, can this shift in production technologies happen.

## 6. Regulation

The economics of increasing the share of hydrogen-based steel are dependent on continuing political momentum for decarbonisation via measures such as carbon dioxide pricing and carbon border tax to avoid carbon leakage. Equally important is the provision of start-up capital and subsidies for initial investments to compensate for the capex requirements of the technological shift. Depending on scale, a plant based around DRI and EAF using hydrogen would have significant higher capex requirements in comparison to BF/BOF or present Coal based DRI/EAF.

## 7. Investment

The use of Hydrogen based technology shall make major portion of existing BF-BOF operations redundant as facilities like Coke Oven, Sintering, Blast Furnaces, Basic Oxygen Furnaces etc shall not be required. This shall require huge investment besides retiring existing installation in advance. This is being debated globally and becoming one of the major bottlenecks in adopting hydrogen-based technologies. As climate /green funds are being promoted by numbers of countries, it is expected that such funds may help in expediting the transition from carbon-based steelmaking to hydrogen-based steel making.

#### 9.2.14 Way ahead for steel plants

India is having limited availability of coking coal as well as natural gas and majority of the requirement is met by import. Looking into large import bill of Coking Coal to meet present steel making requirement, alternate route of steel making need to be explored. As the cost of renewable power in India is decreasing, Hydrogen based steel making appears to be an alternative. However, in view of techno-economic constraints mentioned above and additional safety measures required for handling Hydrogen, use of Hydrogen in Iron and steel making in India may take longer time than expected. Thus, the only option available to take advantages of Hydrogen reduction is either use of Natural Gas in reformed form to replace part of the Pulverized Coal Injection in Blast Furnace or in DRI or Syn Gas through Coal gasification in DRI as both the gases are having in excess of 50-60% Hydrogen. Through this process, use of Hydrogen will be economically utilized without resorting to the need of producing just Hydrogen and increasing cost of production of steel and making industry globally non-competitive. In all the Integrated Steel Plants, Coke Oven gas is also available which has more than 50-60% of Hydrogen besides 25-30% CH<sub>4</sub> and a small percentage of CO, to start with use of such gas can be explored to replace part of PCI in Blast Furnace. Some research works are in progress for dry reforming of methane (present in Coke Oven Gas) by using  $CO_{2 \text{ captured}}$  from Blast Furnace to produce Syn Gas or Hydrogen and re-injection of the same in the Blast Furnace. Success of such projects will ensure availability of low-cost Hydrogen from Coal source and may result in large utilization of it in the iron and steel making. The experience gained with limited Hydrogen can be utilized in future for developing 100% Hydrogen based Iron and Steel making.

## 9.3 Hydrogen in transport sector

Hydrogen can play a significant role in decarbonising the transport sector. Hydrogen Fuel cell electric vehicles (FCEVs) are powered by hydrogen. They are more efficient than conventional internal combustion engine vehicles and produce no tailpipe emissions—they only emit water vapor and warm air.

Hydrogen has a higher density as compared to other fuels; thus, it produces more energy in lesser weight due to which it is a viable decarbonised fuel option for heavy commercial vehicles/trucks plying on longer routes. Battery electric trucks are not viable since, the heavy load of batteries significantly reduces their capacity to carry freight.

According to IEA statistics, more than 40 000 FCEVs were on the road globally by the end of June 2021. Global FCEV deployment has been concentrated largely on passenger light-duty vehicles, which accounted for three-quarters of FCEV stock at the end of 2020, with buses making up ~15% and commercial vehicles meeting the remaining 10%.

Korea, the United States and Japan have focused their efforts on deploying passenger cars, and together hold 90% of the market share in this segment, however they have a very small number of buses and commercial vehicles. Meanwhile, China adopted policies for fuel cell bus and commercial vehicle uptake, and now dominates global stocks in these segments (93% of buses and 99% of commercial vehicles in 2020).

In Europe, numerous announcements in 2020-21 showcase stronger efforts to deploy fuel cell buses and trucks. Several manufacturers and projects aim to deploy thousands of buses in the next decade. Hyundai has already delivered 46 heavy-duty trucks to Switzerland as of July 2021 and plans to deploy 1 600 vehicles in the country by 2025, while the Port of Rotterdam and Air Liquide have created an initiative to deploy 1 000 fuel cell trucks by 2025 and a joint call signed by over 60 industrial partners aims for up to 100,000 trucks by 2030. Based on current and announced capacity, the IEA estimates that fuel cell manufacturing could enable a stock of 6 million FCEVs by 2030, satisfying around 40% of Net Zero Emissions by 2050 Scenario needs.

# 10 COST ECONOMICS: COAL TO HYDROGEN

As per the information received from SINOPEC China against with Coal landed price of Rs 3500 per ton, Hydrogen Cost can be around USD 1 per Kg. However, this is without CCUS and Hydrogen Compression Unit required for transportation and Ash Disposal unit. It seems that their ash content is less than 20% and hence they have used Entrained Gasifiers. This hydrogen is used in a Refinery and Hydrogen is supplied at 5 MPa. More than 80% requirement of Hydrogen in Fertilizer, Methanol and Petrochemical sector in China is met by Coal Gasification route as it is one of the lowest costs varying USD 0.9 per Kg to around USD1.5 per kg. Some of the oil refineries also in China are using Hydrogen from coal instead of Steam Methane Reforming. This is mainly to reduce import of oil and natural gas by China and India also is in the same state and can minimize dependency on imported Oil and Natural gas if Coal to Hydrogen is promoted. Since Indian coal may have higher ash content but cost of such coal would be lower, the higher capital cost in case of Indian coal may be compensated by the lower input cost of coal. The estimated cost of hydrogen from coal, based on information from various sources, could be in the range of \$ 1.3 - 1.5 per Kg.

An estimate of Hydrogen Cost based on the Natural Gas & Coal as feed stocks, with different feed stock cost has been made based on published information from several sources as given below:

Basis: Plant capacity: 150 Tons/day hydrogen generation plant			
Feedstock: Natural gas			
Case 1: Natural gas price - \$ 7/MMBTU			
Cost of hydrogen production:	~ Rs 115/kg		
Contribution of natural gas feedstock:	~73%		
Capex:	~9%		
Others (Fixed and variable costs):	~18%		
Case 2: Natural gas price - \$ 10/MMBTU			
Cost of hydrogen production:	~Rs 151/Kg		
Contribution of natural gas feedstock:	~79%		
Capex:	~7%		
Others (Fixed and variable costs):	~14%		

Basis: Plant capacity: 150 Tons/day hydrogen generation plant			
Feedstock: Coal			
Case 1: Coal price – Rs 1500/ ton			
Cost of hydrogen production:	~ Rs 102/kg		
Contribution of Coal feedstock:	~16%		
Capex:	~44%		
Others (Fixed and variable costs):	~40%		
Case 2: Coal price – Rs 3000/ ton			
Cost of hydrogen production:	~Rs 118/Kg		
Contribution of natural gas feedstock:	~28%		
Capex:	~38%		
Others (Fixed and variable costs):	~34%		

Engineers India Limited has also made an estimate of Hydrogen production cost with Natural Gas and typical Indian Coal as feed stocks. The findings are as below:

# Natural Gas as Feed Stock

Hydrogen can be produced through SMR technology with Natural Gas as feedstock. Major components of a typical NG based plant are Steam Reformer, Shift Reactor, PSA & U&O facilities. Based on in-house data available in EIL, hydrogen production cost for a typical hydrogen unit is as below:

Case-1	
Parameters	Value
NG price	US\$ 7/ MMBtu
Cost of H2 production	Rs 1,01,000 / MT
Case-2	
Parameters	Value
NG price	US\$ 10/ MMBtu
Cost of H2 production	Rs 1,33,000 / MT

Case-3

Parameters	Value
NG price	US\$ 12/ MMBtu
Cost of H2 production	Rs 1,54,000 / MT

# **Coal as Feed Stock:**

Hydrogen can be produced with coal through Gasification technology. Major components of the plant are Gasification section, Air Separation Unit, Shift & Gas cleaning section, PSA & U&O facilities. Typical Indian coal characteristics have been used here. Based on in-house data available in EIL, hydrogen production cost for a typical coal-based hydrogen unit is as below:

Case-1	
Parameters	Value
Coal Price	1500 Rs/ MT
Cost of H2 production	Rs 1,21,000 / MT
Case-2	!
Parameters	Value
Coal price	3000 Rs/ MT
Cost of H2 production	Rs 1,57,000 / MT
Case-3	
Parameters	Value
Coal price	1000 Rs/ MT
Cost of H2 production	Rs 1,10,000 / MT

The estimates both in case of coal and for natural gas are without CCS/CCUS, which is likely to cost around 0.5 \$/ Kg. and has been separately dealt in this report.

# Green Hydrogen Production from Renewable Power

The emphasis in recent time has been on Green Hydrogen which is produced from renewable power using water electrolysis. The current cost of green hydrogen as reported by various agencies is around 4.5 \$ per Kg. The recent policy initiatives regarding waiver of transmission charges and green power banking etc may cut down the cost to around 3 \$ per kg. which is higher than hydrogen cost from fossil sources. The production cost of Green Hydrogen is dependent on green power tariff, electrolyser capital cost, its efficiency and its utilization.

# International Published data on Hydrogen Production Costs

As per an international publication in Science Direct of 2019, the costs of hydrogen production costs derived from each method is estimated as below:

1.	Steam Methane Reforming without CCS	:	2.08 \$/Kg
2.	Steam Methane Reforming with CCS	:	2.27 \$/Kg
3.	Coal Gasification without CCS	:	1.34 \$/Kg
4.	Coal Gasification with CCS	:	1.63 \$/Kg
5.	Solar/wind power based Electrolysis	:	5.78-6.03 \$/Kg

Since Green Hydrogen at present is expensive and Natural Gas prices are more volatile besides, India being dependent on imports for Natural Gas, it makes a case for coal to hydrogen route.

# **11 RECOMMENDATIONS & WAY FORWARD**

## 11.1 Gasification Technologies

The technology for gasification should be the one suitable for high ash Indian coal, with an eventual goal to produce hydrogen. Based on the advantages and disadvantages of different techniques available for coal to hydrogen production, circulating fluidized bed gasification is found out to be the best suitable technique as per the requirement of final product and environmental concerns. The committee therefore propose that the technology to be selected for coal gasification be based on a circulating fluidized bed gasification system followed by a solid separation and, subsequent elaborate gas purification. Indian coal being of low grade with high ash content (upto 45%), makes both stable operation of circulating fluidized gasification as well as synthesis gas cleanup challenging. The target is to select an optimized process for this purpose, involving process intensification concepts and judicious integration of the entire plant. Although numbers of institutions are already working on development of indigenous gasification technology for high ash coal, commercialization of the same may take a longer time. To meet objective of Coal gasification Mission i.e achieving around 100 million tonnes coal gasification by 2030 and to promote coal to hydrogen to make cheap hydrogen available in the country, external technological support may be necessary. The committee is of the opinion that some policy intervention may be required for interim period to reserve some of the low ash coal for Coal to Hydrogen or Coal Gasification so that the execution of coal to hydrogen project can be expedited.

## **11.2** Water Gas Shift Reactor and H<sub>2</sub> Purification

After syngas clean up, we have to for water gas shift to increase hydrogen generation followed by hydrogen PSA purification process. The gasification technologies are fairly mature but need to be demonstrated on a semi commercial stage. The water gas shift reactor is still to be proven, while both the concepts are quite scientifically known. There are other elements which need to be tried out to make to demonstrate the process in a comprehensive manner. These are things like membrane reactor for water gas shift, ammonia/ H<sub>2</sub>S removal systems and thermal management systems to get better overall cold gas (hydrogen conversion) and hot gas efficiency.

## 11.3 CCUS for Blue Hydrogen

The carbon intensity of coal/lignite/petcoke based hydrogen (black/brown hydrogen) can and must be reduced by combining carbon capture sequestration & utilization (CCUS) technology with a conventional gasifier approach. This is one area of major focus in view of the climate commitments made by India and if we have to build up plants for converting Coal to Hydrogen, we will have to select suitable technological options in the overall scheme of things.

The key drivers for CCS or, alternatively, for effectively utilisation (CCU) of the CO<sub>2</sub> generated from coal-to-hydrogen plant, are (a) the fossil carbon inherent in the source feed which would be emitted if not captured or utilized and (b) the need to maximize the hydrogen by converting the CO present in the generated syngas to additional hydrogen via an energy-intensive water-gas shift (WGS) reaction.

A number of CCS projects are being considered, but majority of the processes are in developmental stage except some technologies like methanol or bio-ethanol or electrolysis of carbon di-oxide which have been used on commercial scale. It is expected that more options may be available in time to come . The CCS efforts fall into two broad categories, dedicated geological storage (such as salt caverns) and enhanced oil recovery (pumping the carbon dioxide into aged wells and displacing an equivalent amount of crude oil that is then brought up to the surface). Both possibilities exist in India, but the farther the carbon dioxide produced by gasification has to travel from point of generation to point of use, the less the net benefit derived by CCS.As potential for enhanced oil recovery appears to be limited, there is a need to accord focus on utilization i.e CCU. Several companies across the globe have declared various CCU projects which are at different stages of execution around the world today. The advantage of CCU technologies would be the possibilities of integrating this within the complex of coal gasification, gas cleaning, water gas shift reactor,  $CO_2$  removal/capture unit and conversion to chemicals or aggregates. One of the option which can be considered is electrolysis of  $CO_2$  to convert the same into CO or dry reforming of  $CO_2$  to convert the same into syn gas and its utilization downstream.

# 11.4 Utilization of Hydrogen

Currently, the refineries and fertilizer plants are the largest consumers of hydrogen which is primarily being produced from natural gas as grey hydrogen, though the refineries have plans to use the  $CO_2$  for conversion to chemicals and becoming net zero refineries. In case we could produce hydrogen from coal along with CCUS, we will be able to produce blue hydrogen from indigenous sources rather than imported natural gas. Looking into future demand of hydrogen of around 11.7 million tonnes by 2030 and 28 million tonns by 2050, the Committee is of the opinion that ample opportunities are there for utilisation of hydrogen produced from coal in the refineries, fertilizer units, steel plants and transport sector etc and this can be the cheapest source of hydrogen.

#### 11.5 Way Forward

The Committee is of the view that India has an opportunity to produce hydrogen from domestic coal and we may aggressively pursue this option in our overall hydrogen ecosystem. We could set-up a couple of semi-commercial/demonstration gasification units for conversion of coal to hydrogen. We may also go for integrating the CCUS units along with gasification so that the blue hydrogen thus produced is more acceptable. The gasification technologies may be selected based on assessment of the potential for eventual commercial upscaling and keeping the option of biomass co-gasification along with coal subject to availability of bio-mass in the close vicinity of such gasification units.

It is suggested that the coal gasification units be established near the hydrogen demand centres and/or near the coal mines. The cost of transportation of hydrogen would be high at present and hence we need to examine the economics before deciding upon the locations of gasification plants. Alternatively, we could establish the plants closer to the natural gas grid so that hydrogen thus produced could be injected to some extent i.e. up to 18-20% into the natural gas pipelines, as to that extent of hydrogen injection in the natural gas pipelines may not need the modification of the gas pipelines. This will facilitate the utilization of hydrogen produced from coal in the industries currently using imported natural gas. The Government may consider deputing a team of experts led by senior officials of the Ministry of Coal & Coal India to visit a few of the globally best of the units for Gasification, Gas Cleaning & Conditioning, Water Gas Shift Reactors, CCUS technologies for carbon capture and preferably conversion to chemicals and aggregates.
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## **12 ANNEXURES**

File attached separately