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HYDROGEN FROM NATURAL GAS WITHOUT RELEASE OF CO_2 TO THE ATMOSPHERE

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Abstract

The "Hydrogen economy", in which hydrogen will be a main carrier of energy from renewable sources, is a long term prospect. In the near and medium term increasing demand for hydrogen - also as an energy carrier in special niches- will probably be covered by hydrogen from fossil sources, mainly natural gas. This can be acceptable from an environmental as well as an economical point of view, since hydrogen can be produced from natural gas at acceptable costs, without release of CO_2 to the atmosphere. There are two main options for this:

1. Hydrogen from natural gas by conventional technology (e.g. steam reforming) including CO_2 sequestration.
2. High temperature pyrolysis of natural gas, yielding pure hydrogen and carbon black.

Technologies for industrial scale realisation of these options have been developed and evaluated in Norway, which is a large producer and exporter of natural gas. The economy and market opportunities are discussed in the paper. It appears that renewable energy costs must come down considerably from present levels before hydrogen from renewables can compete with hydrogen from natural gas without release of CO_2 to the atmosphere.

1 Introduction

In the light of increasing evidence that the "Greenhouse effect" is a real threat, means to reduce anthropogenic release of CO_2 to the atmosphere must be developed. Hydrogen is a clean fuel in the sense that no CO_2 is emitted by its use. The predominant way of producing hydrogen, however, is from natural gas or other fossil sources, involving coproduction and release of CO_2 to the atmosphere. "Clean" production of hydrogen, e.g. by water electrolysis using renewable energy, is not competitive with present-day renewable energy costs. Therefore, the "Hydrogen economy" must be regarded as a long term prospect. For the near and medium term, methods for hydrogen production that are economic as well as environmentally acceptable - i.e. without release of CO_2 to he atmosphere - are required.

roduction of hydrogen by conventional technology such as steam reforming, combined with CO_2 separation and sequestration, is not a new proposal. A Dutch study of this ption was presented at the 9th WHEC [Reference 1], a Norwegian study at the 10th

WHEC [Reference 2]. The possibilities of decomposing hydrocarbons directly into carbon and hydrogen were also discussed at the 9th WHEC [Reference 3]. Since then, the Norwegian company Kværner Engineering has developed a plasma arc process for cracking of natural gas or other hydrocarbons into hydrogen and carbon black, the "CB & H Process" [Reference 4]. A thorough study of hydrogen production from natural gas and coal, including CO_2 sequestration, will be presented in another paper at this conference [Reference 5]. It can be stated that adequate technology for "CO_2-free" production of hydrogen from natural gas has been developed and demonstrated. Economic and other aspects implied will be discussed in the following.

2 Hydrogen by steam reforming with CO_2 sequestration

2.1 Natural gas processing

The major part of the world's hydrogen production (about 45 M tonnes per annum) is accomplished by steam reforming of natural gas followed by shift reaction:

$$\begin{aligned} &CH_4 + H_2O \rightarrow CO + 3H_2 \\ &\underline{CO + H_2O \rightarrow CO_2 + H_2} \\ &\underline{CH_4 + 2H_2O \rightarrow CO_2 + 4H_2} \end{aligned}$$

It is noteworthy that only half of the hydrogen produced originates from the hydrocarbon, the other half coming from water. Hydrogen can also be produced by partial oxidation ($CH_4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2$), the proportion of hydrogen from hydrocarbon will then be greater. Since steam reforming is strongly endothermic and partial oxidation is exothermic, combined processes (autothermal reforming, etc.) will usually be most efficient. The technology for hydrogen from natural gas is mature and practised on a large scale for production of methanol, ammonia and hydrogen for petrochemical and other uses. State-of-the-art descriptions were given at the 9th WHEC [Reference 6,7].

There are many options for separation of CO_2 from mixtures with other gases. Commonly practised on an industrial scale are chemical scrubbing (e.g. absorption in a monoethanol amine (MEA) solution) and physical scrubbing (e.g. absorption in Selexol or in water at elevated pressure). Such methods are the most likely to be used for CO_2 removal from flue gases from fossil-fuelled power plants, having relatively low CO_2 concentrations. The recovery and disposal of CO_2 from such plants is currently the subject of great international interest, and several studies have been published [Reference 8]. In mixtures with higher CO_2 concentrations, which will occur in IGCC power plants or in steam reforming plants, other options may become available for CO_2 separation. These may be cryogenic separation, selective adsorption on solid sorbents, or separation by selective membranes. The possibility of incorporating selective sorbents or membranes into a steam reforming reactor opens great improvement possibilities, since selective removal of one of the products would shift the equilibrium and create more favourable thermodynamic conditions for the process [Reference 9].

2.2 CO_2 disposal options

There are two main options for large-scale disposal of CO_2 : Ocean disposal and underground disposal. A recent overview was given by the IEA Greenhouse Gas R&D Programme [Reference 10]. The ocean disposal concept was introduced by Marchetti [Reference 11] and is currently subject to renewed interest and evaluation, particularly in Japan. There are several options for ocean disposal, such as pipeline disposal at great depths, ship transport of solid CO_2 for disposal at great depths, and shallow injection followed by gravity-induced transfer to greater depths. The latter concept has been thoroughly studied and evaluated at a Norwegian research institute, the Nansen Environmental and Remote Sensing Center [Reference 12, 13]. Model simulations indicate that the method will be feasible, but some uncertainties remain as to the efficiency and duration of CO_2 retention, and its effects on the marine environment. A pilot scale experiment is being planned in order to obtain reliable data. For underground disposal there are possibilities in aquifers, exhausted gas or oil reservoirs, or in oil reservoirs for enhanced oil recovery (EOR). The CO_2 will be transported as a supercritical liquid from its source to the place of disposal. Suitable formations may be found both on-shore and off-shore. The costs of transportation will of course depend upon distances and volumes. Off-shore locations may be advantageous, since under-water pipelines tend to be more cost-effective.

Possibilities for CO_2 disposal in formations on the Norwegian continental shelf are quite good. The EOR option has been evaluated [Reference 14] and seems to be feasible, but apparently no oil reservoir is yet ripe for tertiary production. Some gas wells are being taken out of production and might serve as CO_2 depositories. Aquifer disposal is, however, the option that is first going to be tested on a large scale. This will happen at Sleipner in the North Sea, a field producing natural gas with a high contents of CO_2. This will be removed by chemical scrubbing on the production platform, compressed and injected into a nearby aquifer. The plant is under construction and scheduled to come into operation this fall. Thus, we shall soon have hands-on experience in the performance and costs of large-scale (in the order of 1 M tonne/year) underground CO_2 disposal.

2.3 Economic aspects

The Norwegian study presented at the 10th WHEC [Reference 2] concerned a state-of-the-art hydrogen production plant (120.000 Nm^3/h H_2) located on the Norwegian coast. CO_2 was to be separated from both process and flue gases by chemical scrubbing and taken to shallow sea disposal by a relatively short pipeline. Assuming a natural gas price of 2,78 USD/GJ, a hydrogen cost of 6,67 USD/GJ was derived (table 1). The study to be presented by Audus and Kårstad in this conference [Reference 5] gives cost estimates for a very large (280.000 Nm^3/h) hydrogen plant at a coastal site in the Netherlands. It is based on state-of-the-art technology [Reference 7] with CO_2 separation by chemical scrubbing from the process gas only, giving 80-85 % recovery. The CO_2 is compressed and transported by pipeline to an off-shore exhausted gas well for disposal. The study is based on realistic and conservative data. Assuming a natural gas price of 3.0 USD/GJ, it ends up with a hydrogen cost of 6,97 USD/GJ.

It is likely that a cost estimate of a large hydrogen plant located on the Norwegian coast would give similar results. Such a plant would probably be located at one of the three gas pipeline terminals on the western coast, for instance the Kårstø terminal which has been in operation for many years. The distance from Kårstø to the Sleipner field is 160 km, but suitable aquifers for CO_2 disposal may be found at half this distance. Considering the possibilities that CO_2 may be separated from some of the gas taken ashore, or from the flue gas from a conceptual gas-fired power station at the site, combined solutions for CO_2 disposal might be conceivable, which would reduce costs. Also, at a pipeline terminal a natural gas price below the assumed level of 3 USD/GJ might be obtainable. Thus, the estimated hydrogen cost of 6,97 USD/GJ is probably conservative.

From a site at a natural gas terminal, export of hydrogen to the European continent by pipeline may be conceiveable. A gas pipeline with a capacity of 16 billion Nm³/year could handle a mixture of 10 % hydrogen in natural gas (Hythane). This would correspond to 80 % of the output of a large (280.000 Nm³/h) hydrogen plant. The cost of the mixture would be about 3,3 USD/GJ, i.e. 10 % above the natural gas cost. It may well be that the advantages of Hythane, as a clean fuel, will be worth this price increase. According to a study of pipeline transport of Hythane [Reference 15] the cost of transportation of a 10 % mixture should not deviate much from the cost for pure methane.

3 Hydrogen by the CB&H process

3.1 Process description

In 1990 Kværner started development of the innovatory Kværner CB&H process. By pyrolysis of hydrocarbon feedstocks the process produces two valuable products, i.e. hydrogen and carbon black. Since 1992 a full-scale pilot plant has been operated, and the development has resulted in a technology which now is ready for commercialisation.

In a reactor a plasma torch supplies the necessary energy to pyrolyse the feedstock. The plasma gas is hydrogen, which is recirculated from the process. Thus, apart from the feedstock and the electricity demand of the plasma torch, the process is self-sufficient.

A heat-exchange system conducts the heat-transfer from the high-temperature products to the feedstock and plasmagas, which are pre-heated to set values. Excess heat is used to produce steam for external use. In large plants, it might be feasible to use excess heat in order to generate electricity. The recuperated process heat results in a decreased energy demand of the process.

Considering the CB&H process primarily as a hydrogen-supplier, the process has the following advantages compared to other hydrogen processes:

- lower hydrogen production cost (se section 3.2).

- higher feedstock efficiency. The CB&H process yields almost 100% efficiency. The impurity level of the process is restricted to the impurities introduced by the feedstock. Hence, no CO_2 is liberated from the process and other non-hydrocarbon impurities are in the ppm level.
- high flexibility concerning the feedstock. All hydrocarbons ranging from light gases to heavy oil residues can be utilized as feedstock, and a change in feedstock does only affect the product-ratio between hydrogen and carbon black. This implies that the Kværner CB&H process can meet demands in process plants or refineries where the feedstock mix and availability of hydrogen clean-up gases varies over the year.
- high process modularity. Kværner has engineered and cost-estimated plants with capacities ranging from 1 - 360 million $Nm^3$of hydrogen per year. Higher hydrogen production rates are easily attainable by adding more modules.

3.2 Cost of hydrogen

Figure 1 shows the estimated costs for several hydrogen production processes based on renewable sources or natural gas. The figure reveals that traditional steam reforming and Kværner's CB&H process are the most economical alternatives regarding the hydrogen production cost.

A more detailed comparison between the steam reforming process and the CB&H process is given in table 1. According to the table the production of hydrogen from natural gas is slightly more expensive by use of the steam reforming process than the CB&H process. If CO_2 sequestration is implemented in the steam reforming process, however, the CB&H process is undoubtedly the most favourable alternative based on costs. A lower investment cost as well as the valuable production of carbon black makes Kværner CB&H process the most economical alternative.

Figure 2 shows the price relationship between hydrogen and carbon black for two plants with hydrogen production capacities of 100 and 500 million Nm^3/year. The price relationship will vary within a certain interval depending on the costs of the input parameters, i.e. natural gas and electrical power. It appears that in the large-scale plant, with upper-level input costs and a carbon black sales price of 150 USD/tonne (which is considered conservative, also in the metallurgical market) the hydrogen cost will be 0,07 USD/Nm^3, corresponding to 6,48 USD/GJ.

One may conclude that hydrogen may be produced, either by the CB&H process or by conventional natural gas processing with CO_2 sequestration, in a price range of 6,5 - 7 USD/GJ. Provided that the carbon black produced can be sold at reasonable prices, the CB&H process seems to have a competitive edge.

For comparison, the study of [Reference 5] estimates the cost of electrolytically produced hydrogen. Based on an electricity price of 0,045 USD/kWh, a hydrogen cost of 24 USD/GJ is derived. From the relationship between electricity price and electrolytical hydrogen cost given in [Reference 2], it appears that even at an electricity price of 0,01 USD/kWh, electrolysis in a 200 MW plant yields a hydrogen cost of 11

USD/GJ. Thus, electricity will have to be practically free of charge for electrolytic hydrogen to be competitive with hydrogen from natural gas.

4 Carbon black, a huge market potential

Carbon black holds a large market potential, both within the traditional rubber industry as well as within new markets, as represented by the metallurgical industry. Thus, production of valuable carbon black will contribute to the CB&H process by reducing the hydrogen production cost, as shown in figure 2.

The world wide production of carbon black is approximately 6 million tonnes, where the capacity in Western Europe amounts to some 1 million tonnes. The demand within metallurgical industry for carbon black material in Europe is approximately 2 million tonnes, this is equivalent to 8 billion Nm^3H_2

Carbon black is dispersed as a filler in rubber, plastics, paints and inks in order to modify the mechanical, electrical and optical properties of the product. Particularly important is carbon black's unique ability to reinforce rubber. The consumption of carbon black within different sectors is shown in figure 3. The figure shows that 90% of the applications are utilized in the rubber industry.

4.1 Traditional carbon black for the rubber industry

The main consumption of carbon black (70%) is utilized for the production of tyres, as shown in figure 3. In addition 10% is used for manufacture of other products used within the auto industry (fan belts, hoses), while another 10% of the carbon black produced is used as industrial rubber (conveyor belts, cable insulation, footwear).

The carbon black industry produces a whole range of carbon black grades, which are classified according to surface area and structure of the carbon black aggregate. The higher the surface area, the finer is the carbon black grade and the higher is the price obtained per unit of carbon black. The consumption of fine grade carbon black is also higher than the corresponding for medium/coarse grades. The European market volume for fine grade and medium/coarse grades of carbon black is shown in figure 4. The figure also shows the corresponding amount of H2 which is produced along with carbon black when utilizing the natural gas based Kværner CB&H process.

Traditionally carbon black for rubber application is produced by use of processes characterised by low feedstock efficiency and high CO_2 emissions. In addition the processes produce small quantities of NOx, SOx and heavy metals. In comparison the Kværner CB&H process, which produces clean, valuable hydrogen and carbon black, exhibits a feedstock yield of nearly 100% and gives emissions in the ppm level, makes a better choice.

Following the years of low investment in the carbon black industry, there is not expected significant new capacity on stream the next two or three years. Hence, shortness of supply of carbon black remains a possibility[Reference 16].

4.2 New markets for Carbon Black

The metallurgical industry is identified as an interesting and new carbon black market. Petrol coke has generally been utilized as carburizer and reduction material in this industry. However, pyrolytically produced carbon black is expected to exhibit several advantages compared to petrol coke. The pyrolytically produced carbon black is fully clean, highly reactive and pulverized. The introduction of such a carbon material in the metallurgical industry will bring along several positive effects, such as reduced sulphur emission and a reduced carbon consumption, which in turn will lead to reduced CO_2 emission. Utilizing a clean carbon material may also result in products containing a low level of impurities as well as a reduced energy demand. Considering the fact that the availability of relatively clean petrol coke is decreasing, makes the potential of pyrolytically produced carbon black within the metallurgical industry even higher.

Following applications of carbon black in the metallurgical industry have been evaluated as interesting:

- reduction material for production of SiC (Si, FeSi)
- carbon additive/carburiser to the steel- and foundry industry.

4.2.1 Reduction material for production of SiC (Si, FeSi)

Reduction material for the production of SiC needs to have a high reactivity towards gaseous SiO and to give a high yield of quartz. Carbon Black produced by use of the Kværner CB&H process has shown excellent properties concerning these requirements.

The total production of SiC in Europe today is approximately 130 000 ton/year, with a consumption of reduction material of about 150 000 ton/year. By replacing all or part of the total carbon (petrol coke) used in production of SiC in the Achison process with carbon black, a potential of 10-20% higher production yield is achieved, giving a price level of > 250 USD/ton (cif).

4.2.2 Carbon additive/carburiser to the steel and foundry industry

Carbon material for this application must have properties as

- fast solution rate into the metal bath
- high fix carbon, low level of sulphur, nitrogen, moisture, ash and volatiles

The total consumption of carbon additive in Europe is approximately 300 000 ton/year, where high and regular qualities account for respectively 20% and 40% of the total. High qualities includes graphite type carbon, synthetic graphite as well as calsined petroleum coke with a low sulphur content. The price level for high quality material is > 615 USD/ton (cif)

Regular type qualities include petroleum type coke and metallurgical coke. The price level > 310 - 460 USD/ton (cif). The level will depend upon the sulphur content, which normally is between 0.2 and 1,2%.

The volume and price estimates for the utilization of carbon black within the metallurgical industry is incorporated in figure 4.

5 Hydrogen and carbon black interplay

The interplay between carbon black and hydrogen is in earlier sections described by reduced hydrogen production costs when the carbon black revenue is included in the calculations. Such an interplay may also result in reduced CO_2 emissions when combining the CB&H process with other industrial processes. Hence, figure 5 shows the effect when a gas fired power plant is combined with Kværner CB&H process and a metallurgical process, e.g. SiC production. The CB&H plant needs electric power to run the reaction from natural gas to carbon black and hydrogen. This will increase the CO_2 emission, but this increase will be more than compensated for by the decrease in CO_2 emission which is obtained by the utilization of carbon black and hydrogen as feedstock in SiC production and electrical power generation, respectively. An example of achievable reduction in CO_2 when producing 20 000 tonnes of carbon black from natural gas is given below:

Generation of 85 GWh electrical power in a gas fired power plant:	+ 29 000 tonnes CO_2
Reduction in CO_2 emission due to substitution of natural gas with hydrogen in the power plant:	- 45 000 tonnes CO_2
Reduction in CO_2 emission due to lower specific energy consumption in SiC production and a higher energy efficiency because of a better reduction material:	- 22 000 tonnes CO_2
Reduction in CO_2 emissions due to higher production yield and better feedstock utilization:	- 20 000 tonnes CO_2
Total reduction:	58 000 tonnes CO_2

In addition to reduced CO_2 emissions, utilization of carbon black as reduction material in the metallurgical industry will also result in better product quality and reduced SO_2 and dust emissions.

In the scenario given in figure 5, hydrogen could be utilized for other purposes than as a power plant fuel. Hence, a hydrogen peroxide plant or a methanol plant might be incorporated in the process system. This would give a more valuable utilization of the produced hydrogen. The reduction in CO_2 emissions would however be lower than the corresponding obtained when hydrogen was utilized as fuel.

6 Conclusions

With present-day prices of natural gas and electricity, hydrogen from natural gas is by far the cheapest option for hydrogen supply. Such production can be accomplished without release of CO_2 to the atmosphere. One possibility is to use high temperature pyrolysis, as in the Kværner CB&H Process, producing pure hydrogen and carbon black. Another possibility is to use conventional processing (steam reforming) followed by CO_2 sequestration, which will add some 25 % to the cost of the hydrogen produced.

Carbon black is not an environmental problem, but a useful product with a great market potential in the metallurgical industry. The cost of the co-produced hydrogen will depend on the prices of the input commodities (natural gas and electricity) and on the revenues from the carbon black. With current gas and electricity prices and conservatively estimated carbon black prices, a hydrogen cost of about 6,5 USD/GJ is calculated. The hydrogen cost by steam reforming including CO_2 sequestration is estimated at about 7 USD/GJ. Thus, costs in the range from 6,5-7 USD/GJ can be foreseen for hydrogen from natural gas without release of CO_2 to the atmosphere. Hydrogen produced by water electrolysis will not be able to compete with this unless electricity becomes very much cheaper - or natural gas much more expensive - than they are to day.

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Table 1 HYDROGEN FROM NATURAL GAS WITHOUT RELEASE OF CO_2

	Steam reforming	CO_2 seque-stration	Sum	CB & H process
Investment, USD/Hm^{3}H$_2$h^{-1}	890	722	1612	1471
Capital cost x)	17.18	13.94	31.1	29.4
Gas cost x)	39.34	-	39.3	48
Electricity cost x)	0.91	0.77	1.7	33
Op. & maint. x)	4.28	0.71	5	14.3
Miscellaneous x)	5.4	0.46	5.9	-
Steam x)	-	2.05	2.1	- 5
Carbon black x)	-	-	-	- 53.6
Sum x)	67.11	17.93	85.1	66.1

x) All costs in U.S. mills/Sm^{3}H$_2$

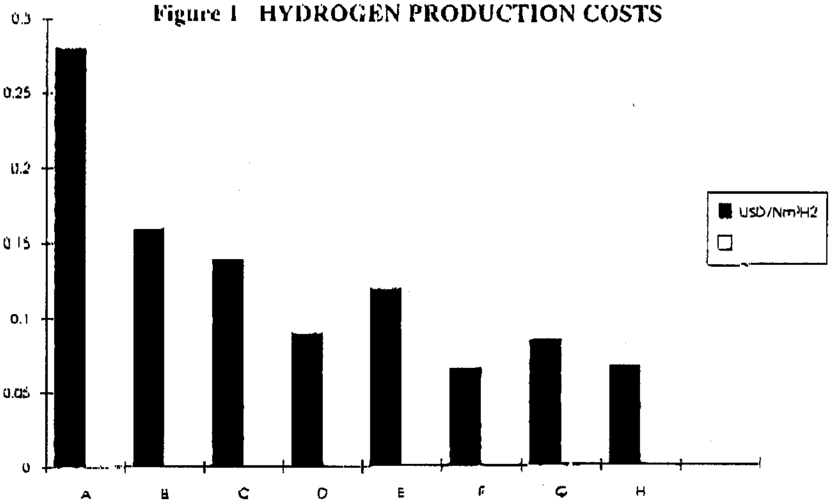


Figure 1 HYDROGEN PRODUCTION COSTS

A: Wind power, 0,06 USD/kWh
B: Hydro power, 0,03 USD/kWh
C: Geothermal power (Hot Elly electrolysis)
D: Hydrogen from biomass
E: CB & H process, without carbon black revenue
F: CB & H process, with carbon black revenue
G: Steam reforming of natural gas, with CO_2 sequestration
H: Steam reforming of natural gas, without CO_2 sequestration

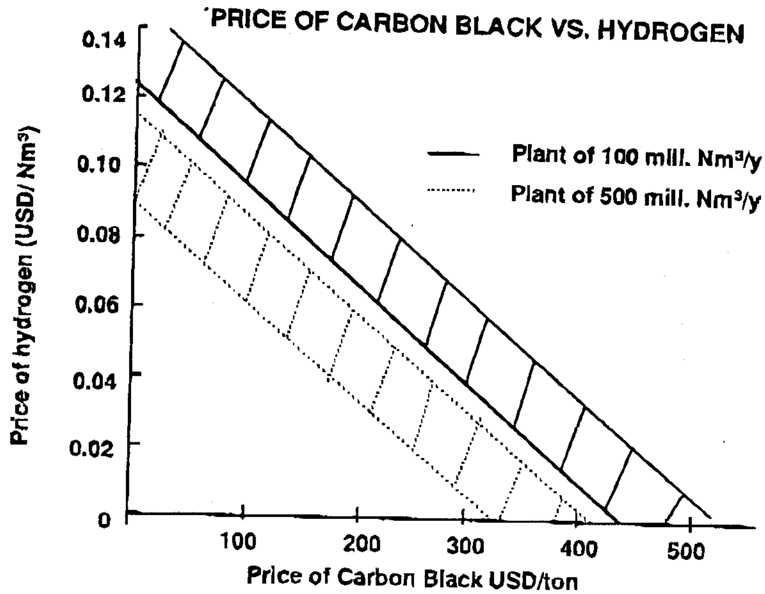


Fig. 2 Price of carbon black vs hydrogen for different hydrogen production capacities

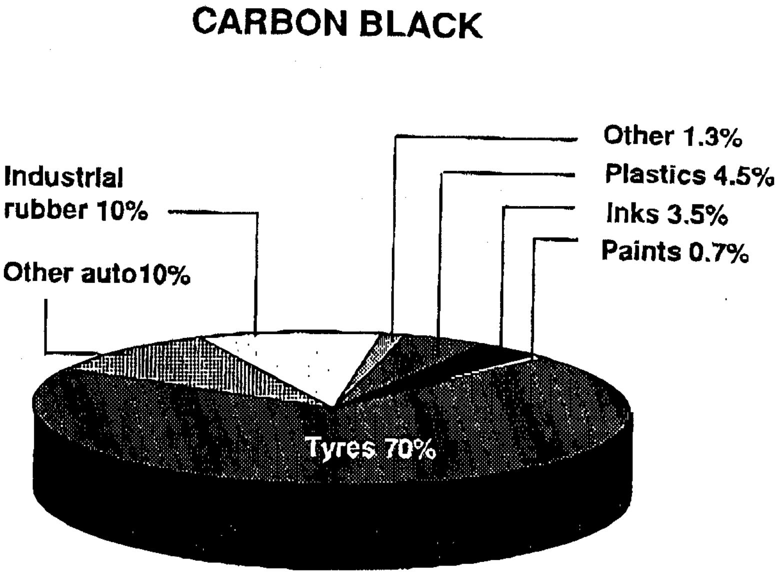


Fig. 3 Application of Carbon Black

THE CARBON BLACK MARKET IN WESTERN EUROPE

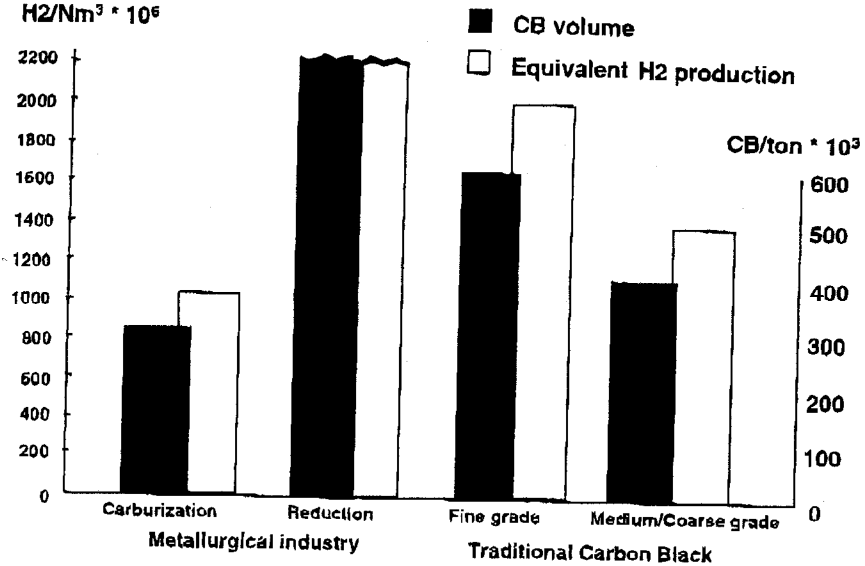
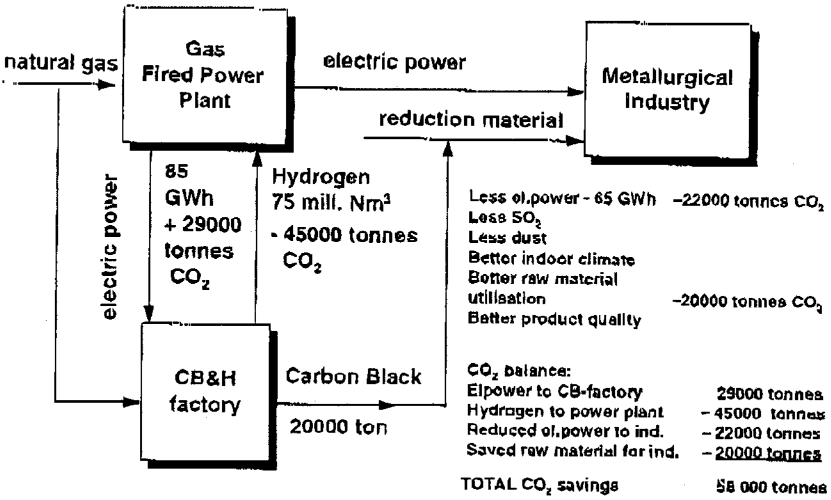


Fig. 4. Volume market for carbon black use in metallurgical industry and rubber industry. The equivalent production of hydrogen by use of the Kværner CB&H process is also shown.

HYDROGEN AND CARBON BLACK INTERPLAY



Less el.power - 65 GWh -22000 tonnes CO_2
Less SO_2
Less dust
Better indoor climate
Better raw material utilisation -20000 tonnes CO_2
Better product quality

CO_2 balance:

Elpower to CB-factory	29000 tonnes
Hydrogen to power plant	- 45000 tonnes
Reduced el.power to ind.	- 22000 tonnes
Saved raw material for ind.	- <u>20000 tonnes</u>
TOTAL CO_2 savings	58 000 tonnes

Figure 5 CO_2 savings when combining a gas fired power plant, Kværner CB&H process and processes within metallurgical industry