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HYDROGEN PRODUCTION FROM COAL AND COAL BED METHANE, USING BYPRODUCT CO_2 FOR ENHANCED METHANE RECOVERY AND SEQUESTERING THE CO_2 IN THE COAL BED

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ABSTRACT

Rapid advances in proton exchange membrane fuel cell technology are creating renewed interest in hydrogen, the preferred fuel for these fuel cells. A promising strategy for providing hydrogen for fuel cells in countries such as China that are poorly endowed with conventional hydrocarbon resources but coal- and coal-bed-methane-rich would be to produce it from coal and coal bed methane, using the low-cost CO_2 generated as a byproduct of hydrogen production to stimulate the recovery of methane from deep beds of unminable coal. While awaiting the arrival of fuel cells for major commercial applications, hydrogen so produced might be used in the manufacture of ammonia for fertilizer, as a less costly option than making hydrogen from coal only, as is done at present in China. Moreover, the production of hydrogen in this manner would lead to low levels of lifecycle CO_2 emissions, because the CO_2 injected into the coal bed for methane recovery would be sequestered there.

BACKGROUND

Rapid advances being made in proton exchange membrane (PEM) fuel cell (FC) technology for transportation and stationary combined heat and power (CHP) applications are creating renewed interest in hydrogen, the preferred fuel for these fuel cells (Williams, 1998a; 1998b). However, because a H_2 fuel infrastructure is not yet in place, the PEM FC will be introduced into the market for many applications using existing hydrocarbon (HC) fuel infrastructures, with conversion of the HC fuel at the point of use into a H_2-rich gas suitable for fuel cell use—e.g., onsite reforming of natural gas for CHP applications, and liquid HC fuel processing onboard a car. These are likely to be only transitional strategies, however. If the FC car is successfully launched in the market with a liquid HC fuel, the automotive system would generate internal market pressures to shift to H_2 derived from carbonaceous feedstocks via thermochemical processes, as soon as the H_2 infrastructure could be put into place, because of the higher first cost, higher maintenance cost, and lower fuel economy of the gasoline FC car relative to the H_2 FC car (Ogden *et al.*, 1997; Williams, 1998b).

This paper discusses H_2 production from coal and methane recovered from deep coal beds, using the byproduct CO_2 for enhanced coal bed methane (CBM) recovery, with sequestration of the injected CO_2 in these beds. Potential FC and other applications of the H_2 so produced are discussed for China.

CBM RESOURCES AND CURRENT CBM RECOVERY TECHNOLOGY

Coal beds are both source rocks and reservoir rocks for large quantities of methane-rich gas. This gas is typically produced throughout the burial history of the coal in volumes ranging from 150 to 200 normal cubic meters (Nm^3) per tonne of coal, as a result of biogenic and thermogenic processes whereby plant material is progressively converted into coal (Rice *et al.*, 1993). Because coal is a microporous solid with large internal surface areas (tens to hundreds of square meters of per gram of coal), large quantities of methane (CH_4) produced this way will remained trapped in the coal bed, adsorbed on coal surfaces. The large micropore surface areas and the close proximity of CH_4 molecules adsorbed on the internal surfaces (approaching liquid densities) of these micropores make high gas storage densities possible even though the porosity associated with the macropores of the system of natural fractures is low—in the range 1-5% (Rogers, 1994). A good coal bed in the San Juan or Warrior Basin in the United States holds two or three times as much gas as the same volume of a sandstone reservoir of like depth having 25% porosity and 30% water saturation (Kuuskraa and Brandenburg, 1989). In general, gas content increases with increasing coal rank; higher rank coals can contain up to 30 Nm^3/tonne. Some coals have generated more CH_4 than they can store, resulting in expulsion of the excess CH_4 to the atmosphere or into adjacent reservoirs (e.g., trapped under a caprock above the coal bed).

Worldwide CBM resources are estimated to be 85-262 trillion Nm^3 (Rice *et al.*, 1993); the corresponding energy value is 3,400-10,400 EJ, equivalent to 0.3-0.9 times the mean estimate of remaining recoverable conventional natural gas resources worldwide (Masters *et al.*, 1994). In China, CBM resources are estimated to be 30-35 trillion Nm^3 (1,200-1,400 EJ) at depths less than 2000 m (Rice *et al.*, 1993; Sun and Huang, 1995); another estimate (Murray, 1996) is that the upper limit of the total CBM in place in China is 75 trillion Nm^3 (3,000 EJ). For comparison, total fossil fuel consumption

in China was 30 EJ in 1990, and the mean estimate of the remaining recoverable conventional natural gas resources in the United States is 700 EJ (Masters *et al.*, 1994). The fraction of the CBM resource that can be recovered economically depends on both the quality and accessibility of the resource and the recovery technology employed.

CBM is recovered commercially in the United States, mostly in the San Juan Basin of New Mexico and Colorado and the Black Warrior Basin of Alabama and Mississippi (McCabe *et al.*, 1993). U.S. CBM production grew rapidly from 1.1 billion Nm^3 in 1988 to 25.5 billion Nm^3 in 1996 (97% of which was not associated with coal mining operations), when CBM accounted for 6% of total U.S. natural gas production.

Current practice is to depressurize the coal bed (usually by pumping water out of the reservoir), which leads to desorption of the gas from the micropores of the coal matrix, its diffusion through the coal matrix to macrofractures in the coal called "cleats," and its flow through the cleats to the wellbore for recovery. The process is simple and effective but slow and inefficient (Gunter *et al.*, 1997). Typically there is a significant time lag (days to months) between the beginning of the dewatering process and the time when substantial gas recovery rates are realized.

CO_2 INJECTION FOR CBM RECOVERY AND CO_2 SEQUESTRATION IN THE COAL BED

An alternative approach to CBM recovery that may prove to be more efficient than present technology involves injecting a gas into the CBM reservoir; CO_2 is especially promising as a candidate injection gas because it is twice as adsorbing on coal as is CH_4; it can therefore efficiently displace the CH_4 adsorbed on the coal (Gunter *et al.*, 1997). CO_2 injection makes it possible to maintain reservoir pressure and produce CH_4 quickly. As CO_2 moves through the reservoir it displaces CH_4. The limited experience to date indicates that very little of the injected CO_2 shows up in the production well until most of the CH_4 has been produced (Gunter *et al.*, 1997), so that the prospects for permanent sequestration of the injected CO_2 appear to be good.

Of course, CO_2 sequestration in the coal bed would prevent subsequent mining of the coal. However, deep or otherwise unminable coal beds for which coal mining is uneconomic might prove to be attractive for CBM recovery and CO_2 sequestration. For example, 90% of the nearly six trillion tonnes of U.S. coal resources deposited at depths less than 1800 m is unminable with current technology, either because the coal is too deep, the seams are too thin, or mining would be unsafe (Byrer and Guthrie, 1998). Deep coal beds can contain especially large amounts of CH_4. In the United States, more than 2/3 of the 2.3 trillion Nm^3 of CBM in the Piceance Basin (McFall *et al.*, 1986) and approximately ½ of the 0.6-0.9 trillion Nm^3 of CBM in Menefee coal in the San Juan Basin are below 1500 m (Crist *et al.*, 1990). The CH_4 content of coals tends to increase with depth, both because the higher formation pressures are beneficial as a driving force for gas production, and because more gas can be adsorbed in the micopores of coals as the pressure increases (Rogers, 1994). Moreover, deep coals are common in many parts of the world (Kuuskraa and Wyman, 1993). But data on resources deeper than about 1500 m are sparse because such resources are of little interest for coal mining (Rogers, 1994), and, in the CBM community, conventional wisdom is that such deep coals will have extremely low permeabilities and thus would be unproductive. However, Kuuskraa and Wyman (1993) have argued that early estimates of low permeability for deep coals might be overly pessimistic and that under favorable geological conditions (such as low stress and dry coals) CH_4 recovery from deep coals might turn out to be a productive option.

An hypothesis advanced by Gunter, which remains to be verified by experience, is that with CO_2 injection it will often be feasible to recover about 90% of the CBM in place in a homogeneous coal seam, irrespective of the reservoir permeability, down to permeabilities of about 1 millidarcy, a relatively low permeability (private communication, Bill Gunter, March 1998). In contrast, with conventional CBM recovery technology the recovered fraction over a typical project lifetime increases with reservoir permeability (Kuuskraa and Boyer, 1993). Thus the gain relative to conventional recovery technology would tend to be greater for reservoirs with low permeability.

There has been a modest amount of field experience with this technology. Amoco conducted a pilot test of CO_2 injection for CBM recovery in the San Juan Basin in December 1993; this was followed by another San Juan Basin CO_2 pilot project conducted by Meridian (now Burlington Resources) in 1995. In the spring of 1998 a project was launched to test this technique in Canada's Alberta Basin, under the auspices of the Alberta Research Council. Although the recovery of CBM via CO_2 injection into deep coal beds is not yet commercial, the technology could be commercialized in 5-10 years if there is sufficient market interest.

Key to the viability of this approach to CBM recovery is having a cheap source of CO_2 at the prospective CBM recovery

site. One potential source of low-cost CO_2 is at plants that produce H_2 from a carbonaceous feedstock, where CO_2 is produced in a relatively pure stream as a byproduct of H_2 manufacture. Thus locating plants that produce H_2 from coal or another carbonaceous feedstock near CBM reservoirs could provide low-cost CO_2 for CBM recovery.

LAUNCHING A CBM INDUSTRY IN CHINA USING BYPRODUCT CO_2 AT NH_3 PLANTS

China is well-positioned to launch an industry that produces CBM via CO_2 injection. It has large CBM resources and produces large quantities of low-cost CO_2 as a byproduct of making H_2 from coal as an intermediate product in ammonia (NH_3) manufacture. Because of the scarcity of its resources of conventional natural gas (the feedstock from which most of the world's NH_3 supplies are derived), most of the NH_3 China produces is derived from coal. Moreover, China is building up a capacity to make fertilizer from coal using modern gasification technology. China has in operation, under construction, or on order, 25-30 modern, oxygen-blown gasifiers; many are for gasifying coal and nearly all are for chemical process applications—mostly for NH_3 production. Chinese interest in such technology arises because nitrogen fertilizer demand is growing and much of the existing coal-based NH_3 production involves small, inefficient, and polluting plants, many of which are likely to be replaced with larger, cleaner, and more cost-competitive plants. The modern coal gasification technology now being introduced could be used to make H_2 for fuel cell applications when fuel cells are established in China's markets.

When NH_3 is produced from coal, the byproduct CO_2 generation rate is about one kmol of CO_2 per kmol of NH_3. The CO_2 potentially available for CBM recovery depends on the fertilizer produced. If the desired product is ammonium nitrate (NH_4NO_3), all the CO_2 is available. If instead the product is urea (NH_2CONH_2), about half the CO_2 is needed for urea manufacture. In either case excess CO_2 could be used for stimulating CH_4 recovery from deep beds of unminable coal, if such beds were located nearby. China should consider locating near prospective CBM recovery sites new plants for making NH_3 from coal and using the low-cost byproduct CO_2 for stimulating the production of CBM.

Here the results of modeling CBM recovery and use in conjunction with NH_3 manufacture from coal are described. It is assumed that NH_3 plants are located near sites with deep unminable coal deposits containing CBM, so that byproduct CO_2 can be used for stimulating CBM recovery, with sequestering of the injected CO_2 in the coal bed. Two alternative CBM uses are considered: (*i*) *Case I*: CBM is used to produce additional H_2 and thus more NH_3 (for NH_4NO_3 and for NH_2CONH_2 production in Cases Ia and Ib, respectively); (*ii*) *Case II*: CBM is used to produce electricity in a gas turbine/steam turbine combined cycle power plant (in conjunction with NH_4NO_3 and NH_2CONH_2 production from coal in Cases IIa and IIb, respectively). Material and energy balances for Cases Ia and IIa are shown in Figures 1 and 2, respectively. In all instances production rates, costs, and CO_2 emission rates are compared to *Base Cases*, in which H_2 and electricity are produced from coal only, with venting of the excess CO_2 generated in H_2 manufacture. For all cases, CBM recovery and use are considered in conjunction with the manufacture of 26 PJ per year of H_2 from coal for fertilizer applications—an amount of H_2 adequate for producing 920 thousand tonnes of NH_3 per year.

It is assumed that CH_4 is recovered from a 10 m-thick, homogenous coal bed at an average depth of 856 m (the average depth of all CBM wells drilled in the United States in 1990) and that the CH_4 in the bed has a concentration of 15 Nm^3 per tonne of coal. Adopting the "Gunter hypothesis," it is assumed that 90% of the CBM in place is recovered over the facility life. The assumed lifetime CBM recovery rate is 1.78 million Nm^3 per hectare (compared to 2 to 4 million Nm^3 per hectare for the CBM-rich regions of the San Juan Basin) and the assumed land area per well is 65 hectares (a typical value for the San Juan Basin), so that the lifetime CBM recovery per well is 115 million Nm^3. For a particular well the CBM production profile will vary markedly over time. However, detailed modeling of the CBM recovery profile at each well is not attempted. Rather, each case considered involves a large number of wells, and it is assumed that production can be arranged so that the aggregate output of all wells can be maintained at a relatively constant level over the assumed 25-year life of the CBM recovery facility.

It is assumed that the CO_2 recovered at the H_2 plants at 1.3 bar is compressed to 100 bar and transported by pipeline to the injection sites, that the number of injection wells equals the number of CBM recovery wells, and that the CO_2 injection rate per well is 2.3 tonnes/hour, which is less than the injectivity (maximum injection rate) if the reservoir permeability is greater than about 1 millidarcy. It is assumed that the CO_2 injection wells are evenly distributed in a square array, at the center of which the conversion facility is located. The number of injection wells, assumed to be proportional to the CO_2 injection rate, is 200, 79, 154, and 82 for Cases Ia, Ib, IIa, and IIb, respectively.

Estimates of the lifecycle costs (in 1991$) of the recovered CBM and the H_2 and electricity produced from coal and CBM

are developed in a report on which this paper is based (Williams, 1998d), assuming a coal price of $1 per GJ, a 10% discount rate, and neglecting taxes/subsidies. Energy quantities are represented on a higher heating value basis.

The estimated CBM costs are made up of two components: (*i*) *CBM recovery cost estimates* per well developed by Kuuskraa and Boyer (1993) for CBM reservoirs in the San Juan Basin of Colorado, using conventional CBM recovery technology, except that the cost for well drilling and completion is assumed to be $249,000 per well—the average for all CBM wells drilled in the United States in 1990 (Pelzet, 1991), plus (*ii*) *cost estimates for stimulating CBM recovery with CO_2 injection* [costs for CO_2 compression, transport, and injection, based on analyses relating to CO_2 sequestration in depleted natural gas fields (Blok *et al.*, 1997) and aquifers (Hendriks, 1994), modified as appropriate to reflect assumed coal bed reservoir characteristics]. The estimated CBM production cost is $2.20-$2.25 per GJ (in 1991$). For comparison, the average U.S. wellhead natural gas price (in 1991$) in 1996 was $1.80 per GJ (EIA, 1997).

Results for Case I H_2 production analyses are summarized in Figure 3. The amount of H_2 produced by the coal/CBM system is 1.82 times and 1.17 times that produced in the Base Case (which involves the same amount of coal feedstock) for Cases Ia and Ib, respectively. In Case Ia the estimated cost of H_2 produced from CBM is $4.8 per GJ_{H2} (65% of the cost of H_2 produced from coal in the Base Case), and the average H_2 cost is 16% less than in the Base Case; in Case Ib the cost of CBM-derived H_2 is $5.8 per GJ_{H2} (higher than in Case IIa because of the much smaller scale of the H_2 plant); in this case the average cost of H_2 is only slightly less than in the Base Case. There is no unique way to assign to subsystems credits for CO_2 sequestered at the system level. Here these credits are assigned to activities associated with CBM production (and thereby to the manufacture of H_2 from CBM), because all costs associated with CO_2 injection are assigned to CBM production. As a result, the net lifecycle CO_2 emissions for CBM-derived H_2 are negative. CO_2 emissions per GJ associated with H_2 manufacture from coal in Case I are also 1/3 less than in the Base Cases, because it is assumed that CBM rather than coal is used to provide the external electricity and heat needed to make H_2 from coal. The average net emission rate for the entire coal/CBM system of H_2 production is 6.0 kgC/GJ_{H2} (15% of the Base Case emission rate) for Case Ia and 19.0 kgC/GJ_{H2} (50% of the Base Case emission rate) for Case Ib.

In Cases IIa and IIb, CBM production supports 50%-efficient combined cycle plants at 545 MW_e and 279 MW_e, respectively, of which 378 MW_e in Case IIa and 135 MW_e in Case IIb is in excess of onsite electricity needs (for both H_2 production from coal and CBM recovery). For Cases IIa and IIb the estimated cost of electricity produced in CBM-fired combined cycle plants is about 13% less per kWh than the cost of electricity from coal in steam-electric power plants in the Base Cases. Moreover, local air pollutant emissions would be much less, in light of the fact that natural gas-fired combined cycle power plants have the lowest local air pollutant emissions of all fossil fuel thermal-electric power generating technologies. As for CBM-derived H_2, net lifecycle CO_2 emissions for CBM-derived electricity are negative; CO_2 emissions per GJ of H_2 produced from coal in Case II are also 1/3 less than in the Base Cases. The average net emission rates for the entire system of H_2 production from coal plus electricity production from CBM are ¼ as much for Case IIa and ½ as much for Case IIb as in the Base Cases (which are assumed to produce from coal as much electricity as in Cases IIa and IIb).

Initially, electricity generation might be preferred to H_2 production for use of the CBM, because with H_2 production a large amount of capital equipment downstream of CBM production (e.g., for H_2, NH_3, and nitrogen fertilizer production) would be idled at high cost if there were substantial unexpected reductions in the CBM recovery rate. In contrast, if electricity is produced from CBM and most is exported to the electricity grid, the capital at risk would be much less both because of the low capital intensity of combined cycle power plants and because unexpected shortfalls in CBM recovery could probably be readily compensated for by other underutilized electric generating capacity on the grid. However, once CO_2 injection technology is well-established in the market, CBM use for producing additional H_2 should be considered wherever there is a sufficiently large market for H_2 (e.g., for extra NH_3 production in the near term or for FC applications in the longer term), in light of the much lower cost of making H_2 from CBM than from coal.

POTENTIAL FOR COAL- AND CBM-DERIVED H_2 FOR TRANSPORTATION APPLICATIONS IN CHINA

The demand for transportation services is growing rapidly in China. However, China has only modest oil resources to support its transportation needs; ultimately recoverable conventional oil resources in China are estimated to be about 400 EJ, 4% of the global total (Masters *et al.*, 1994). But China has abundant coal and CBM resources, which might be effectively utilized in providing transportation services if PEM FC vehicles are successfully launched in the market. China is beginning to explore FC options for transportation. In February 1998, China's State Science and Technology Commission (SSTC) issued a request for proposals from international FC companies to work with China in developing a

FC transit bus demonstration project in Beijing. The intent is that if this demonstration is successful, it would be followed by commercial production of FC buses in China.

Hydrogen derived from coal plus CBM in the manner described above for NH_3 manufacture (with energy/materials balances similar to those for Case Ia) could support FC vehicle fleets at large scales in China. This point is illustrated by a *gedanken* experiment developed in a companion paper (Williams, 1998c). In this experiment H_2 is so produced in quantities adequate to support a hypothetical future automotive fleet of 350 million H_2 FC cars that are driven 15,000 km per year and have a fuel use rate of 2.35 liters of gasoline-equivalent per 100 km [a fuel use rate expected for H_2 FC cars that have the acceleration, hill climbing ability, rolling resistance, aerodynamic drag, and weight characteristics established for cars under the Partnership for a New Generation of Vehicles in the United States (Ogden *et al.*, 1997)]. For such a fleet (Williams, 1998c): (*i*) the fuel cost per km for typical FC car owners in China would probably not be greater than for owners of gasoline internal combustion engine cars of comparable size and performance; (*ii*) the coal requirements for supporting such a fleet would be only 13% of total coal use in China in 1990; (*iii*) a fleet of this size could be supported for 100 years with just ¼ of China's estimated CBM resources of 35 trillion Nm^3, and (*iv*) the lifecycle CO_2 emissions for H_2 production and use in this system (including the assumed use of coal-derived electricity to operate H_2 compressors at car refueling stations) would amount to only 6% of China's CO_2 emissions in 1990. And local air pollutant emissions of such a vehicle fleet would be zero.

Because of its high population density, China might view buses and two- and three-wheeled vehicles as preferable to automobiles for passenger transportation. This *gedanken* experiment suggests, however, that such alternatives should be preferred because they would cause less congestion and noise or because adoption of the automobile culture would require sacrificing other development goals—not because of concerns about energy resource constraints, local air pollution, or greenhouse gas emissions. Hydrogen derived from coal plus CBM used in FC vehicles could support high levels of transportation services in China, making it possible for China to avoid heavy dependence on petroleum for transportation, potentially at competitive costs, with very little local or global air emissions. Moreover, because it presently has very little HC fuel infrastructures in place for transportation, it has the opportunity to "leapfrog" directly to H_2 FC technology, obviating the need for the costly HC-fueled FC transition technology that is being considered for launching FCs in transportation markets in some industrialized countries.

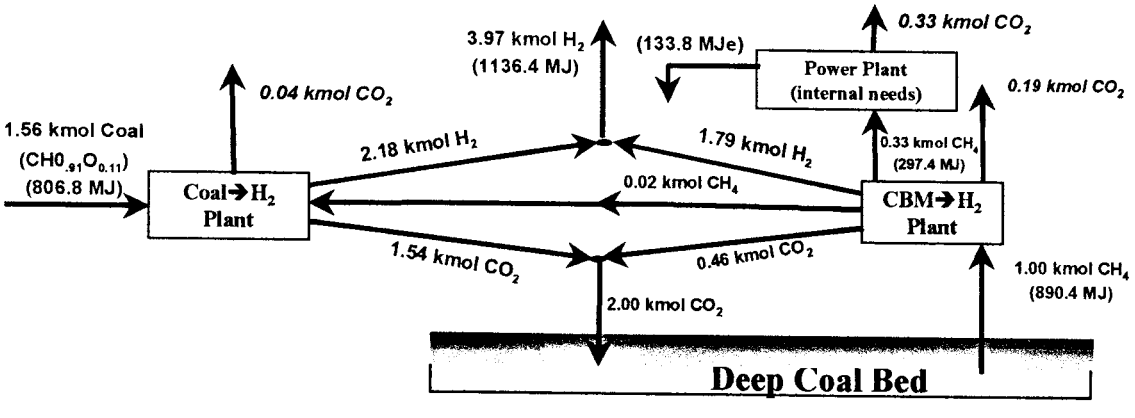

Figure 1: Material and Energy Balances for H_2 Production from Coal and CBM, Using the CO_2 Separated at the H_2 Production Plant to Recover CBM, with Coal Bed Sequestration of the Injected CO_2 (Case Ia) [Detailed material balances are indicated only for C (e.g., inputs of O_2 and H_2O into the system are not shown).]

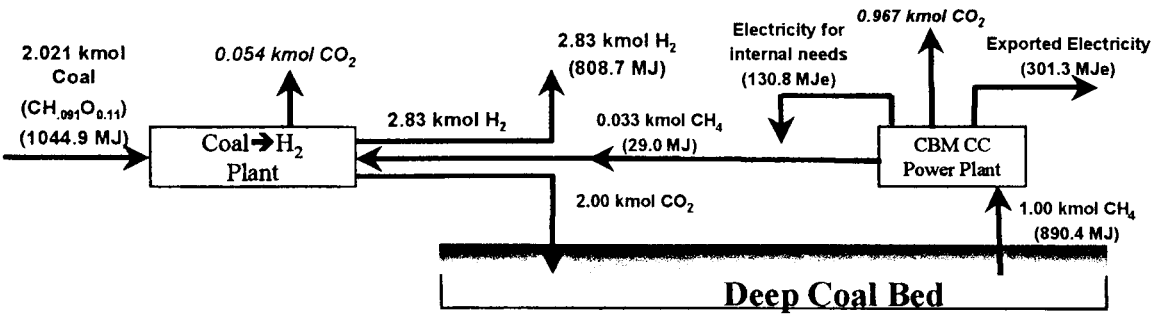

Figure 2: Material and Energy Balances for H_2 Production from Coal and Electricity Generation from CBM in a Combined Cycle Power Plant, Using the CO_2 Separated at the H_2 Production Plant to Recover CBM, with Coal Bed Sequestration of the Injected CO_2 (Case IIa) [Detailed material balances are indicated only for C (e.g., inputs of O_2 and H_2O into the system are not shown).]

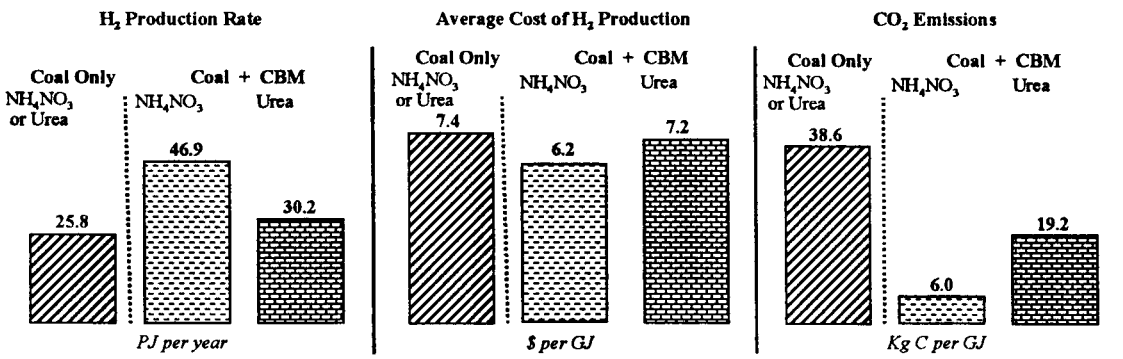

Figure 3: H_2 Production Levels, H_2 Production Costs, and CO_2 Emissions for Alternative Schemes to Produce H_2 from Coal Plus CBM for NH_3 Manufacture [The bars on the left in each set represent the Base Cases, the middle bars are for Case Ia, and the bars on the right are for Case Ib.]

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