Pergamon

PII: S0360-5442(96)00136-3

Energy Vol. 22, No. 2/3, pp. 161–168, 1997

0360-5442/97 $17.00 + 0.00

HYDROGEN PRODUCTION FROM NATURAL GAS, SEQUESTRATION OF RECOVERED CO_2 IN DEPLETED GAS WELLS AND ENHANCED NATURAL GAS RECOVERY

K. BLOK,[†] R. H. WILLIAMS,[‡] R. E. KATOFSKY[‡§] and C. A. HENDRIKS[†¶]

[†]Department of Science, Technology and Society, Utrecht University, Padualaan 14, NL-3584 CH Utrecht, The Netherlands and [‡]Center for Energy and Environmental Studies, Princeton University, Princeton, NJ 08544, U.S.A.

(Received 30 September 1995)

Abstract—If fuel cells are introduced for vehicular applications, hydrogen might become an energy carrier for transport applications. Manufacture via steam-reforming of natural gas is a low-cost option for hydrogen production. This study deals with the feasibility of combining the production of hydrogen from natural gas with CO_2 removal. When hydrogen is produced from natural gas, a concentrated stream of CO_2 is generated as a by-product. If manufacture is carried out near a depleted natural gas field, the separated CO_2 can be compressed and injected into the field and securely sequestered there. The incremental cost of the produced hydrogen (for CO_2 compression plus transport, injection and storage) would typically be about 7% relative to the case where the separated CO_2 is vented. Moreover, CO_2 injection leads to enhanced natural gas recovery as a result of reservoir repressurization. Though the extra natural gas is somewhat contaminated with CO_2, it is a suitable feedstock for hydrogen production. Taking credit for enhanced natural gas recovery reduces the penalty for sequestration to a net incremental cost of typically 2%. These cost penalties are much lower than those typical of CO_2 removal schemes associated with electricity production. Attention is required for optimum plant siting in order to keep CO_2 transport costs low.

INTRODUCTION

Many studies have been devoted to CO_2 removal, including recovery of CO_2 from an energy conversion process and storage outside the atmosphere.[1–4] Most attention has been focused on recovery of CO_2 from power plants. Typical costs for such recovery processes may range from 50–200 US$ per tonne of carbon avoided. Sequestration in depleted natural gas fields is a feasible, secure option for long-term storage of the separated CO_2, as long as the pressure of the reservoir does not exceed the original pressure of the natural gas field.[4–6] On average, about twice as much carbon can be stored in depleted gas fields as CO_2 than was present in the original natural gas.[4] Most analysts expect that costs for underground storage would be lower than those for recovery, ranging from 2–20 US$ per tC avoided. Costs for transport of CO_2 typically are 3–15 $ per 100 km per tC-avoided. Costs for CO_2 recovery, transport, plus sequestration lead to an overall 25–100% increase in the cost of electricity production.

At present, the least-costly option for production of hydrogen is via steam-reforming of natural gas. We examine CO_2 removal and sequestration associated with this way of producing hydrogen. Hydrogen could come to play important roles in the energy economy if there are opportunities to use hydrogen that give it higher value than the energy feedstocks from which it is derived. Natural-gas-derived hydrogen is now used as a feedstock in the production of chemicals such as ammonia. A promising future application is for fuel-cell vehicles. Hydrogen-fuel buses based on the use of the proton exchange membrane fuel cell are approaching commerical readiness; the City of Chicago purchased three in 1995. These fuel cells could plausibly begin to enter automotive markets before 2010.[7,8] In automotive applications, the market would give hydrogen a value both because fuel-cell cars would be as much as three times as energy-efficient as internal combustion engine cars of comparable performance and because they would emit no air pollutants. Moreover, because of the high efficiency of fuel-cell cars, their fuel-cycle CO_2

[†]Author for correspondence; fax +31-30-2537601, e-mail: blok@chem.ruu.nl.
[§]Present address: Arthur D. Little Co., 20 Acorn Park, Cambridge, MA 02140-2390, U.S.A.
[¶]Present address: Institute for Prospective Technology Studies, World Trade Center, Isla de la Cartuja, E-41092 Sevilla, Spain.

emissions per km, when operated on natural-gas-derived hydrogen, would only be about 35% of the emissions for cars with gasoline-fired internal combustion engines.[9,10]

In this study, we examine the feasibility of reducing CO_2 emissions further by recovering the carbon dioxide from hydrogen production and storing it outside the atmosphere. Both the technical feasibility and the economic viability are examined. We consider storing the separated CO_2 in depleted natural gas fields, both without and with enhanced natural gas recovery. We begin by describing several schemes for the production of hydrogen from natural gas, with and without sequestration of CO_2. Subsequently, we discuss the basis for our technical and cost calculations and present the results.

SCHEMES FOR THE CONVERSION OF NATURAL GAS TO HYDROGEN

The first step in the conventional manufacture of hydrogen from natural gas involves reforming the natural gas feedstock with steam at high temperature, to produce a gaseous mixture consisting mainly of carbon monoxide and hydrogen. Subsequently the gaseous product of the reformer is processed in shift reactors operated at much lower temperatures. In these reactors, the carbon monoxide reacts with steam to produce hydrogen plus CO_2. Subsequently, the hydrogen and CO_2 are separated using pressure swing adsorption (PSA) units, the CO_2 is vented to the atmosphere, and the purified hydrogen (up to 99.999% pure) is compressed (e.g. to 75 bar as an input pressure for a long-distance transmission line). Carbon dioxide leaves the hydrogen production plant in two streams: in a diluted stream as a component of the reformer stack gases (about 30% of the total CO_2) and in a concentrated stream that is separated from the hydrogen in the PSA unit (about 70% of the total).[11] A schematic overview is given in Fig. 1, scheme (a).

Alternative schemes with CO_2 sequestration can be designed. Scheme (b) shown in Fig. 1 is a straightforward extension of scheme (a); instead of venting the concentrated carbon dioxide stream to the atmosphere, it is compressed and injected into a depleted natural gas field.

Scheme (c) in Fig. 1 is an alternative option in which extra natural gas is recovered from the depleted field, as a result of gas reservoir repressurization from CO_2 injection; we call this option enhanced gas recovery (EGR). This extra gas contains a substantial fraction of CO_2 (although the co-produced CO_2 is only a small part of the CO_2 injected). In scheme (c), we assume that this extra gas is mixed with natural gas extracted from a conventional natural gas field and used for hydrogen production.

INPUT DATA FOR THE CALCULATIONS

Here we present the technical and cost data for the hydrogen plant, CO_2 handling, and enhanced natural gas recovery that provide the basis for our calculations.

Hydrogen production

A hydrogen production plant has been described by Katofksy,[11] based on natural gas that is 94.7% methane, 2.8% ethane, 0.2% carbon dioxide and 2.3% nitrogen/argon (higher heating 39.7 MJ/m^3). Cost figures for the hydrogen production plant are updated from this earlier study.[9,10] For an overview, see Table 1. For the EGR option, the natural gas input is contaminated with CO_2. Since the contamination on average is less than 4% (v/v) we assume that this does not substantially affect the characteristics of the plant as given in Table 1.

Compression, transport and injection of carbon dioxide

The CO_2 exiting the hydrogen production plant would be available at a pressure of about 1.3 bar.[11] For transport of the CO_2 a pressure of 80 bar is required. From P–H diagrams of CO_2 Hendriks[4] derives that the electricity required to attain this pressure is 281 kJ$_e$/kg, assuming a five-stage compression and an isentropic compressor efficiency of 85%. Above 80 bar, the CO_2 is a liquid; the electricity requirement for compression above this pressure can be calculated as 0.204 kJ$_e$/kg/bar,[12] assuming an isentropic pump efficiency of 70%.

The water content of CO_2 as it leaves the hydrogen plant is 0.6% by weight. After the third and fourth compression stage, further water removal takes place in a knock-out drum. At 55 bar the solubility of water in CO_2 is at a minimum and reaches a level of 0.06% by weight. Further drying of the CO_2

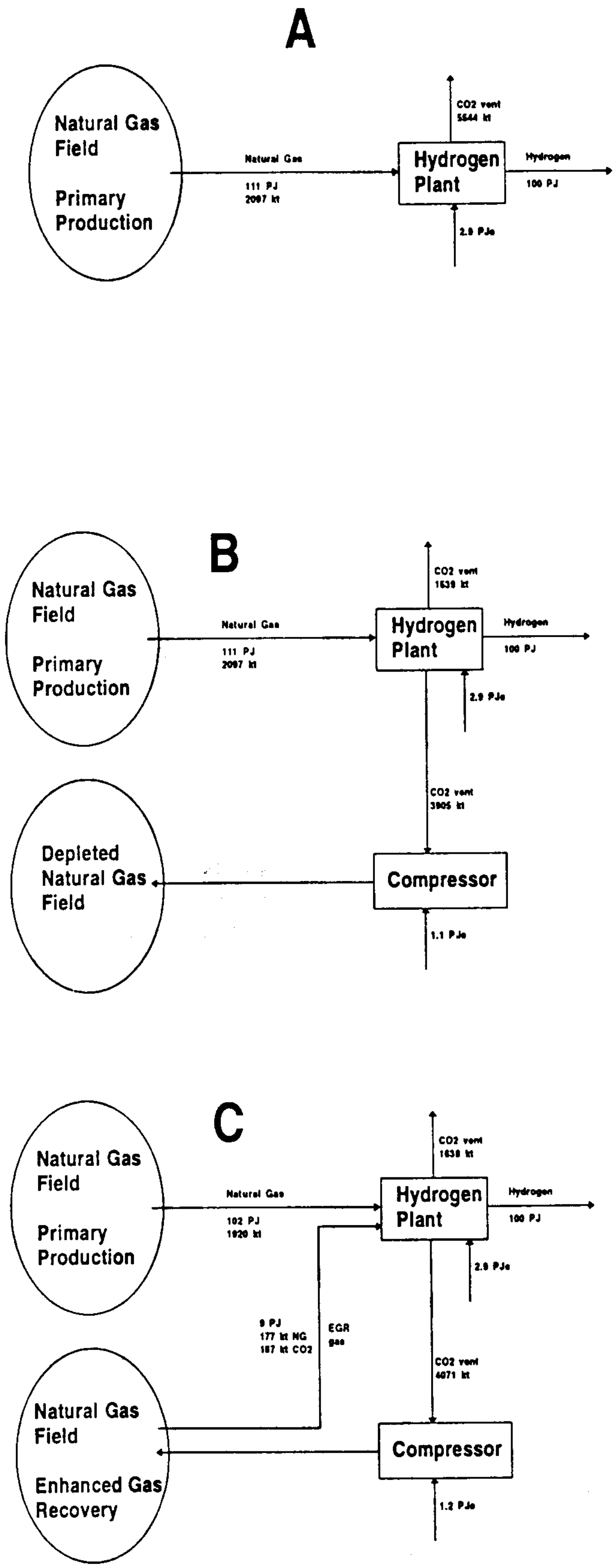

Fig. 1. Schemes for the production of hydrogen out of natural gas, as they are considered in this paper. In these schemes, annual flow rates are given.

Table 1. Overview of the data for hydrogen production used in our calculations. The capital requirement is valid for an annual operation time of 7880 hours per year (90% availability). The figures are valid for a plant with an annual production of 19 PJ per year of hydrogen compressed to 75 bar.

Natural gas requiremment ($GJ/GJ_{product}$)	1.11
Net electricity requirement ($GJ_e/GJ_{product}$)	0.029
Capital requirement ($ per $GJ_{product/year}$)	10.8
Operation and maintenance ($/$GJ_{product}$)	0.61

is possible, but is assumed to be unnecessary for pipeline transport, according to a Shell study.[5] Costs of equipment for the compression of CO_2 including the knock-out drum, are estimated to be 61200 $/(ton/h).[4]

When CO_2 transport is required, there is some freedom to choose the pipeline diameter for a given CO_2 transport rate. Hendriks et al[13] have performed an economic optimization of the pipeline diameter and found for a rate of 500 tonne CO_2 per hour that the cost is minimized if the pipeline diameter is chosen to be 0.5 m. We assume this diameter for a 500 tonne per hour CO_2 flow rate and that pipeline cross sectional area is proportional to flow rate. In the case described by Hendriks et al[13] the pressure drop along the pipeline is 0.12 bar/km. As a base case we assume that 100 km of CO_2 transport is needed. Hendriks et al[13] also derived a formula for the pipeline costs:

$$I = (300 + 1500 \times d^{0.9}) \times L,$$

where I = investment (in fl, we assume that 1 fl = \$0.56), d = pipeline diameter (m), L = pipeline length (m). The author of another study[14] calculates a different optimum pipeline diameter, but comes to about the same costs for CO_2 transportation: about 3 $/ton CO_2 for a distance of 100 km (at a rate of 500 tonne/hour).

For injection of CO_2 we assume that new wells have to be drilled. In general more than one well is required and a CO_2 distribution system has to be built on the surface. The costs of the distribution system and the wells strongly depend on the field characteristics (depth, permeability). We assume figures for an average field and wells having an injection capacity of 40 ton/h, for which the estimated cost is $96,000 per ton/h injection capacity.[4] In all cases operation and maintenance costs are assumed to be 3.6% of the investment (including insurance).

Enhanced natural gas recovery

Before natural gas production begins, a natural gas field typically has a pressure up to 400 bar (depending on the depth of the field and the local pressure gradients). The gas field is considered as depleted when the pressure has dropped to 20–50 bar, even though some of the original amount of natural gas remains in the field. If CO_2 is injected, some additional natural gas can be produced as a result of reservoir repressurization. We call this option enhanced gas recovery (EGR).

As far as the authors are aware, there is only one publication in the open literature in which enhanced recovery of natural gas using CO_2 injection is described, namely a report prepared by Shell.[5,6] In that study, various simulations were carried out. Some simulation results were provided by Boutkan.[15] They are valid for a prototype reservoir consisting of 5 layers with varying permeabilities (see Table 2). The initial natural gas content of this reservoir is 70 billion Nm^3 (approx. 50 Mtonnes). The original gas pressure is 350 bar; primary production is stopped at a gas pressure of 30 bar.

The simulation starts from the assumption that at one side of the reservoir, 5500 ktonne of CO_2 are injected per annum. At the other side gas is extracted. Due to the fact that the injected CO_2 travels relatively quickly through the most permeable layers in the reservoir, the natural gas becomes contaminated with CO_2 after a few years. As soon as this happens, the gas production rate is reduced in the simulation to such a level that the CO_2 production rate does not exceed 5% of the injection rate (275 ktonnes per annum). The production levels of CO_2 and natural gas (assumed to consist entirely of methane in the Shell calculations) are depicted in Fig. 2, both for the prototype reservoir and for a reservoir with a higher permeability contrast.

In our calculations, we assume recovery rates that are averaged over the simulation periods. For the

Table 2. Layering in the prototype reservoir for which simulations were carried out.[5,6] The size of the reservoir is length = 4 km, width = 12.5 km.

Layer	Height (m)	Horizontal permeability (μm^2)		Porosity
		Base case	High permeability contrast	
1 (top)	20	0.050	0.017	0.13
2	8	0.015	0.050	0.11
3	20	0.200	0.600	0.14
4	12	0.010	0.003	0.10
5 (bottom)	8	0.002	0.001	0.05

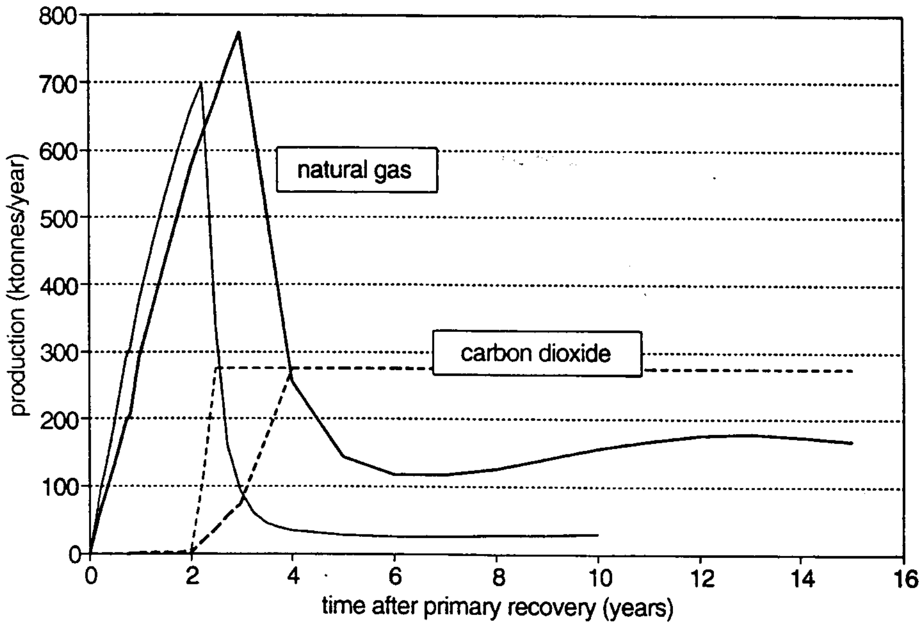

Fig. 2. Production rates of carbon dioxide (dashed lines) and natural gas (solid lines) in the simulations for the prototype reservoir (dark lines) and for the same reservoir, but with a higher permeability contrast (light lines). It can be seen that up to CO_2 breakthrough a considerable amount of natural gas can be produced. In the case of the prototype reservoir, production drops, but a substantial natural gas production rate can be maintained thereafter. In the case of the field with the high permeability contrast the production drops to near-zero after breakthrough. After primary natural gas recovery stops, an amount of gas typically equal to 10% of the primary gas production is still in place. In the enhanced gas recovery cases shown here, the amounts of enhanced natural gas recovery are 6.7 and 2.5% of primary production, respectively.

prototype reservoir, 0.041 tonne of natural gas and 0.044 tonne of carbon dioxide are produced per tonne of CO_2 injected. At this rate, EGR provides 8.4% of the natural gas feedstock requirements for the hydrogen production plant. For a sensitivity analysis we also use data for a reservoir with a higher permeability contrast; in this case, the enhanced recovery amounts to 0.024 tonnes of natural gas (4.6% of the natural gas requirements for the hydrogen production plant) and 0.039 tonne of CO_2 per tonne of CO_2 injected. We assume that these ratios can be maintained over 25 years. We assume that enhanced recovery of natural gas can be carried out using the existing infrastructure, so that no additional investments have to be made for the production of gas (apart from the investments for the CO_2 injection facilities discussed in the previous section).

Other assumptions

All energy values in this paper are on a higher heating value basis. For a base case, we assume a natural gas price of $3/GJ, an electricity price of $0.05/kWh, and a 5% discount rate. The CO_2 emission associated with electricity production is assumed to be 0.153 kg of CO_2 per kWh (world average[16]). The project is assumed to have a lifetime of 25 years. Costs are in 1991 US dollars.

Table 3. Results of the cost analysis for the three schemes (base cases).

Scheme	a	b	c
Capital requirement (million US$)			
Hydrogen plant	1082	1082	1082
CO_2 compressors		30	32
CO_2 pipelines		110	112
CO_2 injection		48	50
Total	1082	1270	1275
Annual costs (million US$)			
Capital costs	77	90	90
O&M Hydrogen plant	61	61	61
O&M CO_2 compressors		1	1
O&M CO_2 pipelines		4	4
O&M CO_2 injection		2	2
Natural gas for hydrogen production	333	333	303†
Electricity for hydrogen production	41	41	41
Electricity for CO_2 compression		15	16
Total	511	547	518
Costs of hydrogen production ($/GJ-HHV)	5.11	5.47	5.18
Avoided CO_2 emission (ktonne/year)	n.a.	3731	3724
Specific CO_2 mitigation costs ($/tC)	n.a.	35	6

†The benefits of enhanced gas recovery are concentrated in the first part of the carbon dioxide injection period and hence the financial benefits are somewhat higher than in the case that the enhanced gas production would be evenly spread over the whole period. We have accounted for this effect by applying levelized discounting. The effect on the cost of hydrogen is small: less than 1%.

RESULTS OF THE CALCULATIONS

For each of the schemes, the average mass and energy flows are indicated in Fig. 1. For the sequestration options, CO_2 emissions for hydrogen production are reduced by 70%. If hydrogen produced via these options were used in fuel-cell cars, the lifecycle CO_2 emissions per km of driving would be about 18% of the emissions for comparable gasoline internal combustion engines, compared to 35% without sequestration.[17,18]

The results of the cost analysis for each of the schemes are presented in Table 3. The cost of hydrogen production is increased 7% when the CO_2 is sequestered in a depleted natural gas field [Fig. 1: scheme (b) compared to scheme (a)]. However, if CO_2 sequestration is combined with EGR, the net additional costs are lower: for scheme (c) the cost of hydrogen is less than 2% more than for scheme (a). Costs of CO_2 removal are $35/tC without EGR and $6/tC with EGR.

In Table 4, a sensitivity analysis is presented. In most of the cases, the results are not very sensitive to our assumptions. Cost increases are within a range of about 2% around the base case. However, the cost depends sensitively on the CO_2 transport distance. If the transport distance from the CO_2 production plant to the well is increased from 100 to 500 km, the cost of hydrogen increases by about 9%; the

Table 4. Results of the sensitivity analysis for the three schemes.

	Hydrogen costs $/GJ-HHV			Specific carbon mitigation costs $/tC	
	a	b	c	b	c
Base cases (Table 3)	5.11	5.47	5.18	35	6
Gas $5/GJ (instead of $3/GJ)	7.33	7.69	7.19	35	−14
Discount rate 10% (instead of 5%)	5.54	5.97	5.66	42	12
Transport distance 0 km (instead of 100 km)	5.11	5.35	5.06	23	−6
Transport distance 500 km (instead of 100 km)	5.11	5.94	5.66	82	54
CO_2 injection 3 times more expensive	5.11	5.57	5.28	45	17
CO_2 injection 3 times cheaper	5.11	5.43	5.14	32	3
Higher permeability contrast	5.11	5.47	5.30	35	18

net cost of CO_2 removal is zero for 50 km of transport and increases by about \$12/tC for each 100 km of transport. Thus, attention should be given to identifying the optimal site for the hydrogen production facility. The choice depends, *inter alia*, on the relative costs of transmitting CO_2 and hydrogen and on the size of the production facility. For the plant sizes considered here (see Fig. 1), the cost per 100 km of transmitting hydrogen in a pipeline sized such that the pressure drop is 2.5 bar per 100 km (a pipeline having a 84-cm diameter) is estimated to be \$ 0.05 per GJ of hydrogen produced, assuming that the capital cost for a hydrogen pipeline is 50% more than for natural gas.[18,19] This is less than the cost of \$ 0.12/GJ for transmitting CO_2 100 km (see Table 4), suggesting it is desirable to site the hydrogen plant as close to the natural gas disposal site as possible. The situation is complicated, however, by the fact that in serving distant hydrogen markets without sequestration it would be less costly to transmit natural gas from the natural gas field to a hydrogen production plant located near the hydrogen market. Savings would arise two ways—first from the lower cost of transmitting natural gas compared to hydrogen, and second from the fact that with the hydrogen plant located near the market, the hydrogen could be sold at a pressure lower than the 75 bar level appropriate for a plant located near the natural gas wellhead that would provide hydrogen for a long-distance transmission line. For a case where hydrogen is pressurized to 20 bar at a hydrogen plant sited near a hydrogen market located 1100 km from the natural gas field compared to a 75 bar at a hydrogen plant sited near the gas field that would deliver hydrogen to the distant market at 20 bar, the cost of hydrogen has been estimated for the former case to be less than for the latter case by an amount equal to half the cost of transmitting hydrogen from the natural gas wellhead to the site near the hydrogen market. But in light of the much higher estimated cost of CO_2 transmission per km, the total cost of hydrogen for the seqestration options would still be less when the required CO_2 transmission distance is minimized. This issue requires further study, however, as other different conditions might give different results.

CONCLUSIONS

The major findings of this assessment of hydrogen production from natural gas with sequestration of the separated CO_2 are the following: (i) When hydrogen is produced from natural gas with sequestration of the separated CO_2, CO_2 emissions at the plant would be 70% less than without sequestration. If hydrogen produced with sequestration of the separated CO_2 were used in fuel cell cars, lifecycle CO_2 emissions per km would be less than 1/5 of those for gasoline internal combustion engine cars. (ii) As long as the original natural gas reservoir pressure is not exceeded, storage of the separated CO_2 in depleted natural gas fields is a secure option for which the storage capacity is on average about twice as much carbon as was in the original natural gas. (iii) The cost of CO_2 removal in the production of hydrogen from natural gas combined with sequestration of the separated CO_2 in depleted natural gas reservoirs is much lower than for CO_2 removal schemes for thermal-electric power plants. (iv) If the injection of CO_2 into depleted natural gas reservoirs is used to promote EGR, the net cost of CO_2 removal can be reduced to a very low level-even zero in some instances. (v) For the situations examined, the lowest costs for CO_2 removal would arise when the hydrogen plant is sited at the depleted natural gas field in which the recovered CO_2 would be stored, although further study is needed to ascertain whether this would always be true. (vi) Detailed information on permeabilities and permeability contrasts of natural gas fields and on the state of depletion of various natural gas fields is needed to carry out accurate assessments of this option in specific regions. Exploiting this option requires a planning of natural gas field depletion strategies long before hydrogen production begins.

Acknowledgements—The authors wish to express their gratitude to V.K. Boutkan (Shell Research BV) for providing data on his simulations of enhanced gas recovery. One of the authors (RHW) thanks the G.R. Dodge Foundation, the Energy Foundation, the W.A. Jones Foundation, and the Merck Fund for financial support for this research.

REFERENCES

1. K. Blok, W.C. Turkenburg, C.A. Hendriks, and M. Steinberg, eds., Proc. of the 1st Int. Conf. on Carbon Dioxide Removal, *Energy Convers. Mgmt.* **33**, 283 (1992).
2. P. Riemer, "The Capture of Carbon Dioxide from Fossil Fuel Fired Power Plants", IEA Greenhouse Gas R&D Programme, Cheltenham, UK (1993).

3. P. Riemer ed., Proc. of the Int. Energy Agency Carbon Dioxide Disposal Symposium, *Energy Convers. Mgmt.* **34**, 711 (1993).
4. C. A. Hendriks, *Carbon Dioxide Removal from Coal-Fired Power Plants*, Kluwer Academic Publishers, Dordrecht (1994).
5. "Carbon Dioxide Disposal From Coal-Based Combined Cycle Power Stations in Depletion Gas Fields in the Netherlands", Ministry of Housing, Physical Planning and Environment, Publikatiereeks Lucht, No. 91 (December 1990).
6. M. J. van der Burgt, J. Cantle, and V. K. Boutkan, *Energy Convers. Mgmt.* **33**, 603 (1992).
7. R. H. Williams, *Technology Review* 21–30 (April 1994).
8. R. H. Williams, "Fuel Cells, Their Fuels and the US Automobile", paper prepared for the First Annual World Car 2001 Conference in Riverside, California, Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, USA (June 1993).
9. R. H. Williams, E. D. Larson, R. E. Katofsky, and J. Chen, *Energy for Sustainable Development*, **1**, 18 (1995).
10. R. H. Williams, E. D. Larson, R. E. Katofsky, and J. Chen, "Methanol and Hydrogen from Biomass for Transportation, with Comparisons to Methanol and Hydrogen Production from Natural Gas and Coal", PU/CEES Report No. 292, Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, USA (July 1995).
11. R. E. Katofsky, "The Production of Fluid Fuels from Biomass", PU/CEES Report No. 279, Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, USA (June 1993).
12. Fachverband Kohlensäure Industrie, "Eigenschaften der Kohlensäure", Koblenz, Germany (1989).
13. C. A. Hendriks, J. C. M. Farla, and K. Blok, "Verwijdering en opslag van CO_2 bij elektriciteitsopwekking (Recovery and Storage of CO_2 with Power Plants)", Department of Science, Technology and Society, Utrecht University (1992).
14. O. Stovholt, *Energy Convers. Mgmt.* **34**, 1095 (1993).
15. V. K. Boutkan, Shell Research BV, KSEPL, Rijswijk, The Netherlands, Written Communication (2 November 1993).
16. A. Faaij, K. Blok, and E. Worrell, "Worldwide Comparison of Efficiency and Carbon Dioxide Emissions of Public Electricity Generation", Department of Science, Technology and Society, Utrecht University (1994).
17. R. H. Williams, "Fuel Decarbonization for Fuel Cell Applications and Sequestration of the Separated CO_2", in R. U. Ayres et al eds., *Ecorestructuring*, UN University Press, Tokyo (forthcoming, 1996).
18. R. H. Williams, "Fuel Decarbonization for Fuel Cell Applications and Sequestration of the Separated CO_2", PU/CEES Report No. 295, Center for Energy and Environmental Studies, Princeton University, Princeton, NJ, USA (April 1995).
19. D. Christodoulou, "Technology and Economics of the Transmission of Gaseous Fuels via Pipelines", Master of Science in Engineering Thesis, Department of Mechanical and Aerospace Engineering, School of Engineering and Applied Science, Princeton, NJ, USA (April 1984).