AN UPDATE ON CANADIAN ACTIVITIES ON HYDROGEN

Alistair I. Miller
Atomic Energy of Canada Limited

Abstract

While Canada is one of the charter signatories of the International Partnership for the Hydrogen Economy, its national programme of R&D is still being defined. With awareness of what the national program will likely include and on the premise that nuclear energy will be the main primary source of energy, Atomic Energy of Canada Limited has evolved a vision of the way forward.

AECL's perspective is based on Canada's existing strengths in hydrogen technology and on the vital importance of significant deployment of hydrogen as a fuel ahead of the commercialisation of Gen IV reactors. Based on Canada's strong position in technology for fuel cells, low-temperature electrolysis, hydrogen storage, and relatively inexpensive electricity, we are promoting phase-out of coal-fired electricity generation between 2010 and 2030 and phase-out of oil-fuelled vehicles between 2020 and 2040. The electricity generation and the initial launch of hydrogen for vehicles will largely depend on Gen III+ reactors. Hydrogen production based on a mix of nuclear and wind generation looks interesting. Distributed generation using low-temperature electrolysis is particularly suited to early deployment of hydrogen fuelling when demand is small.

For hydrogen production after about 2030, AECL is evolving a Gen IV SCWR reactor from its Gen III+ Advanced CANDU® Reactor (ACR®). In the context of our SCWR design, we plan to collaborate with the USDOE's Argonne Laboratory in the development of the relatively low-temperature (~515°C) Cu-Cl thermochemical cycle. We are also working on ways to apply electrical heating to provide higher temperatures for high-temperature electrolysis and I/S technologies.

AECL, in partnership with others, is also developing several existing technical strengths:

1) heterogeneous catalysis for PEM fuel cells.

2) low-temperature electrolysis cells adapted for variable current loads; and is considering resumption of work on.

3) plasmolysis of hydrogen sulphide.

Introduction

The key question is "How do we severely curtail CO_2 emissions, worldwide?" If what we propose does not accomplish this, failing to address the key question becomes part of the problem. The envisaged role for hydrogen is as a non-polluting energy carrier for the transport sector. Its use would avoid both local pollution – with the possible exception of some NOx if it were utilised in internal combustion engines (ICEs) – and CO_2 emissions. The reduction in CO_2 emissions is, of course, only accomplished if the hydrogen production process emits little or no CO_2. Hydrogen production by steam-methane reforming (SMR) would not accomplish this unless the co-produced CO_2 were to be effectively sequestered.

Atomic Energy of Canada Limited's (AECL) vision of hydrogen production has two phases and a variation:

1) For the early stages of hydrogen fueling of the transport sector, hydrogen would be produced by low-temperature electrolysis (LTE) close to the point of fueling. The electricity would be produced from sources that emitted little or no CO_2.

2) When the hydrogen fueling market has grown large enough, centralised hydrogen production will become economic, either using thermochemical processes (such as sulfur-iodine (S/I)), high-temperature electrolysis (HTE), or SMR with sequestration.

3) The variation on Phase 2 is the continuing use of LTE for centralised production including AECL's NuWind© concept in which electricity from nuclear reactors and wind turbines is combined.

The Nadir of SMR-Produced Hydrogen

Unlike oil, where prices are unified worldwide, the price of natural gas has always shown large regional variations. Recently, North American prices have been in the 10 to 14 ^{i}/GJ. At 60 $/bbl, the energy content of oil is valued at about 11 $/GJ. So it appears that oil and natural gas are now being priced in North America as interchangeable energy sources – not unreasonably given the growing extent of capacity for dual-fuelling that now exists in industry. Given the limited availability of new supplies of natural gas within North America, one can reasonably assume that this link will continue.

The market for natural gas in North America is tied together by pipelines. Beyond the reach of this pipeline network, markets for natural gas outside North America have lower, usually much lower, prices. It has often been argued that liquefied natural gas (LNG) would effectively cap the natural gas price in gas-deficient markets like North America at around 5 $/GJ. So far, this has not happened because of the limited capacity of the handful of LNG terminals in North America. However, in a world of inexorable growth in demand for oil from emerging economies, oil prices seem quite likely to remain closer to 60 $/bbl than to 40 $/bbl or less. Note that the actual cost of production in the Middle East, which is as little as 1 or 2 $/bbl, is irrelevant. So it is our judgment that growth in North America's capacity to import LNG is likely to bring the value of LNG into line with that of oil, worldwide, rather than to bring down the price of natural gas in North America.

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i. All dollars are U.S.

The developers of new projects for oilsands extraction in the northern part of Alberta, Canada appear to agree with our expectation of high natural gas prices since they are contemplating use of substitute energy sources to natural gas for their considerable energy needs and for hydrogen to upgrade the bitumen that is produced to synthetic crude. This is totally reasonable: at 12.5 $/GJ for natural gas, SMR hydrogen costs about 2 300 $/tonne H_2 or 2 700 $/tonne if a realistic cost of sequestration[ii] is included. *One concludes that higher prices for natural gas mean that hydrogen produced by SMR is no longer the low-cost process of choice*. This is true today in North America and is likely to become the case in other markets.

The New Competitiveness of Electrolytic Hydrogen

The cost of hydrogen production by both electrolysis and SMR is dominated by the cost of their energy inputs. Around 300 $/t H_2 is associated with capital costs and operation of an SMR while electrolysis cells costing 300 $/kW require produce capital costs of about 400 $/t H_2. Electricity from Generation III+ nuclear reactors (such as Westinghouse's AP-1000, AECL's ACR-1000©, or the European EPR) is expected to cost 3 to 5 ¢/kW.h – 1 500 to 2 500 $/t H_2. This is without credits for co-production of oxygen (300 $/t H_2) and heavy water (120 $/t H_2 net of production costs). On this basis, the total cost of electrolytic hydrogen would be comparable to that from an SMR.

These costs are for continuous production of hydrogen by electrolysis. Studies by AECL [1] have shown that the economics for electrolytic hydrogen production are substantially improved if they are operated intermittently when the price of electricity to the grid is relatively low. While this increases the cost of the electrolysis installation and introduces costs for hydrogen storage, the savings in electricity costs easily offset these. Figure 1 gives an example of the revenues that would have occurred with varying levels of electricity conversion using actual hourly prices paid for electricity in Alberta in 2003 and valuing hydrogen at 2 000 $/t. There is more revenue from sale of any mixture of electricity and hydrogen than from sale of only electricity or from total conversion of the electricity to hydrogen and sale of that hydrogen.

Figure 1.   Value of revenue from sales of hydrogen and electricity, Alberta 2003

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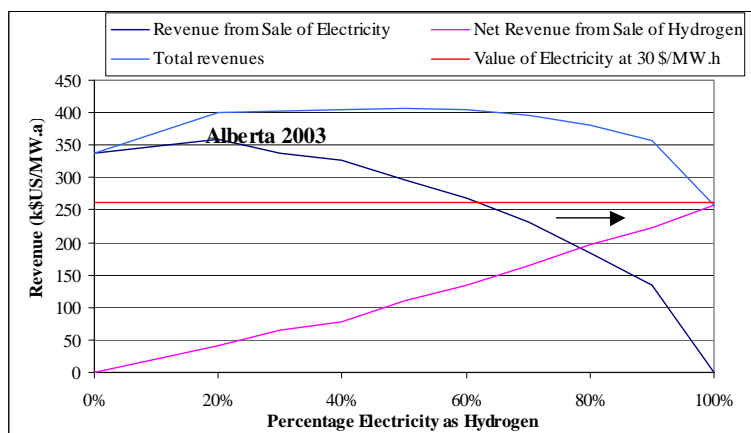
ii.   400 $/t H_2 for CO_2 sequestration occurs with a coast for separation, transport ans sequestration of 50 $/t CO_2. (About 7.5 tonnes of CO_2 is producted for every tonne of H_2.) Including the effects of collateral CO_2 releases from the energy used in CO_2 sequestration, we calculate that the likely cost may be closer to 70 $/t CO_2.

If one were to assume 50% conversion of electricity to hydrogen, a 1 000-MW(e) reactor would provide hydrogen fuel for around 400 000 cars with PEM fuel cells operating a typical (for North America) 20 000 km/a. The balance of the electrical output would be sold to the electricity grid at times of highest demand. Not only does this extend the nuclear power market beyond electricity but it also opens up nuclear's share of the electricity market beyond their normal base-loaded part.

Adding Wind Turbines alongside Nuclear-Generated Electricity

The main drawback to generating electricity from wind and other fickle sources is their intermittency and the need to provide a back-up source of electricity generation. This is a major problem and many studies of actual and proposed wind deployment have examined it. One excellent study by the national electricity generator (ESB) in Ireland examines this problem in detail [2]. The study's conclusion is that wind generation has little scope for reducing CO_2 emissions from the current or future Irish electricity-generating mix. A solution often proposed to address wind's intermittency is to convert the electricity generated into hydrogen using electrolysis. But a simple examination of the economics of electrolysis shows that this is not economically competitive.

In favourable locations, wind turbines can average around 33% of nameplate capacity and produce electricity at about 4 ¢/kW.h – comparable to that from Generation III+ nuclear. This would lead to a capital cost of 1 200 \$/t H_2 (allowing for three-fold oversizing to match wind's low capacity factor) and an electricity cost of 2 000 \$/t H_2. The hydrogen storage cost is hard to calculate because wind (in middle latitudes) has large seasonal variability – as much as three-fold more in peak winter months compared to summer lows. However, underground cavern storage (similar to that used for natural gas) does not introduce a large cost element. Even so, the costs of electricity and capital do not lead to a competitive price for hydrogen produced in this way.

Because the electrolysis installation is already so large when all wind-generated power is converted to hydrogen, the concept of adding yet more electrolysis capacity to allow sale of a mixture of electricity and hydrogen is also unattractive.

To deal with this, AECL has developed the NuWind© concept. In this concept, a mix of electricity from nuclear and wind sources is either sold into the high-value peaks of the electricity market or converted to hydrogen. When the price of electricity is low, any wind-produced electricity is used for electrolysis alongside the electricity generated from the nuclear source. Crucially, the electrolysis installation is designed to handle a about a 40% range of current densities – which is estimated to increase the capital cost by 10%. This also leads to a modest increase in electricity consumption per tonne of hydrogen. Details can be found elsewhere [1]. Figure 2, which combines electricity price data from Alberta in 2003 with actual wind data from a typical mid-latitude site shows that the cost of additional hydrogen coming from the wind source is indistinguishable from that of the nuclear-based hydrogen over a broad range of fractional conversion of the electricity and up to 20% of the electricity being generated by wind (i.e. a wind farm with a nameplate capacity 60% of that of the nuclear).

**Figure 2.   Cost of hydrogen generation with nuclear alone and incremental cost
of additional hydrogen generation from wind, Alberta 2003**
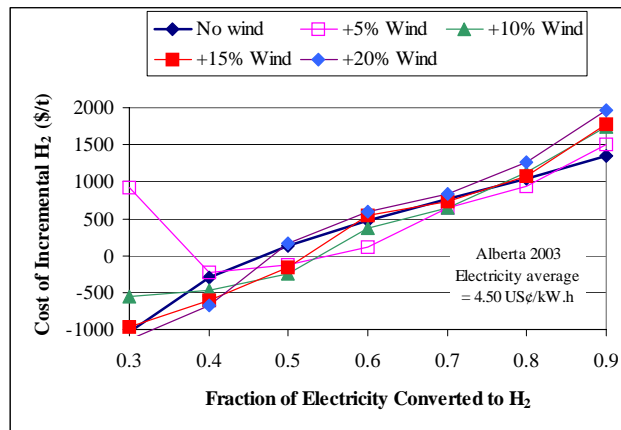
Competitiveness of Hydrogen from Conventional Electrolysis with Advanced Concepts

The most important aspects of hydrogen production by LTE are (1) that it is currently competitive with SMRs using natural gas in the North American context; (2) that it can be deployed immediately; and (3) that it is a near-zero CO_2-emission technology.

Even before the cost of CO_2 capture and storage is included, intermittency of hydrogen generation gives LTE cost superiority over SMR-generation at today's natural gas prices where electricity prices float with market demand. Contrary to the intuitive expectation that electrolysis can only compete where electricity prices are low, it is important to realise that high average electricity prices favour this approach. High prices lead to higher revenues from the electricity product without affecting the cost of electricity generation.

In about twenty years time, new high-temperature production processes may be ready for deployment. This, however, is about as early as their widespread deployment will be possible *and it is probably ten years after hydrogen-fueled vehicles will begin to come into widespread use*. So, with its immediate availability and intrinsic scalability, LTE is the natural forerunner of either HTE or thermochemical processes. A typical 2 000 MW(th) reactor operating with 50% conversion efficiency to hydrogen would fuel 1.1 million vehicles using PEM fuel cells. Though some high-temperature reactor technologies envisage building reactors an order of magnitude smaller, these high-temperature processes are definitely large-scale technologies and multiple sources will presumably be linked by a network of hydrogen pipelines. Their economic competitiveness – currently unknowable – will determine whether they will ultimately displace LTE though it seems plausible that LTE will persist in places where the demand is smaller and pipelines are unavailable. Nonetheless, LTE using off-peak electricity sets a fairly demanding target for HTE and thermochemical processes. If the latters' conversion efficiencies are found to fall much below 50%, LTE will probably offer superior economics through production of electricity (at around 50% conversion efficiency from 950°C reactors) and conversion to hydrogen using LTE.

Canadian Work on Technology Associated with Hydrogen Production

With financial support from Natural Resources Canada, AECL is carrying out a modest program of R&D on high-temperature hydrogen-production processes. The main focus is on the copper-chlorine system, which is principally being studied by the USDOE's Argonne National Laboratory (ANL) [3]. AECL is assembling a consortium of Canadian university researchers to develop the electrochemical step in this cycle in which cuprous chloride is disproportioned into copper and cupric chloride. A particular attraction of this cycle is the relative low temperature (530°C) required for heat input, a temperature that is within reach of AECL's Mark 2 SuperCritical-Water Reactor (SCWR). SCWRs are envisaged as the next stage in the incremental development of CANDU reactors after the ACR.

AECL also has a small program examining SO_3 decomposition using resistance heating of catalysed surfaces. This is a common step of a group of hydrogen-production processes including S/I and the Westinghouse process.

PEM Fuel Cell Development

For many years, Canada has been a notable leader in the development of PEM fuel cells. Sharing of intellectual property among Canadian fuel-cell developers in the last few years has led to a surge in progress. Ballard, who are leading developers of PEM fuel cells and have numerous collaborative agreements with major automobile manufacturers, can now achieve repeated cold starts from -20°C, retain 95% of the original power output after 2 000 hours of operation, and achieve a cost of 100 $/kW (assuming mass production). Ballard is now forecasting competitive PEM-engined vehicles by 2010.

AECL is working with several Canadian PEM developers to apply its heterogeneous catalyst expertise (developed for processes to produce and purify heavy water) to PEM technology. Early work suggests that a significant reduction in platinum loading may be achievable.

Other Routes to Hydrogen Production

With large natural gas production, Canada must dispose of large amounts of hydrogen sulfide (H_2S). The Claus Process is the standard technology used to convert H_2S to sulfur and water. This is wasteful since a small energy input (10% of that needed to dissociate water) is required to dissociate H_2S into sulfur and hydrogen.

In the early 1990s, AECL and Shell Canada collaborated to develop technology originating in the Russian Kurchatov Institute to apply an RF plasma to dissociate H_2S. Proof-of-principle was achieved and AECL is now considering resumption of development since it has potential for significant hydrogen production from much smaller inputs of electrical energy than is needed for electrolysis.

CO_2 Abatement is Urgent

A massive reduction in CO_2 emissions associated with energy supplies is the World's Number One environmental priority. The capacity for CO_2 (assisted by other greenhouse gases) to cause climate change worldwide cannot be quantified precisely though overwhelming qualitative evidence of warming is accumulating in, for example, the melting of previously permanent Arctic sea-ice cover.

If the complex effect of CO_2 on global warming was not bad enough, its direct effect in acidifying the surface waters of the world's oceans is more definite and more clearly deleteriousive[4] a doubling

of pre-industrial CO_2 levels (from 280 to 560 ppm) will lower pH by around 0.4 units. Lower acidity does its most obvious damage through conversion of carbonate ion (CO_3^{2+}) to bicarbonate (HCO_3^-) thus:

$$CO_3^{2+} + CO_2 + H_2O \Rightarrow 2\ HCO_3^-$$

The dissolved carbonate ion is essential to the building and retention of exoskeletons of corals and phytoplankton, key components of ocean food chains. By 560 ppm CO_2 in the atmosphere, reduction of carbonate is expected to lead to huge disruption of these food chains. (Previous, naturally occurring excursions of CO_2 concentration have been counterbalanced by the higher acidity causing carbonate rock to dissolve. However, this is a slow mechanism dependent on the turnover of ocean water and it has no chance of keeping up with the speed with which atmospheric CO_2 is currently rising – about 100 times faster than past naturally occurring changes.) The rise of atmospheric CO_2 concentration is well-documented and the mechanisms of its accumulation in the atmosphere quite well understood with about half the CO_2 added to the atmosphere quickly dissolving in the well-mixed surface layer of the oceans.

From the ocean's surface layer, a small amount of CO_2 migrates to deeper layers, partly in the form of calcium carbonate in the shells of dead organisms raining down to deeper depths and partly through sinking of dense, highly-saline water in the Thermohaline Circulation. Injections of CO_2 to the atmosphere beyond the capacity of these removal mechanisms accumulate and the CO_2 concentration rises continuously, currently at almost 2 ppm per annum. The removal mechanisms would be in balance with CO_2 addition rate if the addition rate were about 40% of the rate in the year 2000. To place the challenge of stabilisation in context, while CO_2 emissions are stabilizing in many industrially developed countries, large emerging economies are becoming significant contributors as they industrialise rapidly. (In their defence, the per-capita emissions of countries such as China and India are still far below those of developed countries.) Consequently, the overall rate of increase continues to rise.

Atmospheric CO_2 concentration will finally stabilise – either by collective human action, by exhaustion of carbon sources (around an atmospheric CO_2 level of ~2000 ppm), or by the collapse of our technological society. If it is to be stopped by collective human action, the stabilized level will depend on the speed and vigour with which low-carbon energy sources are deployed. Delay raises the ultimate level of stabilisation.

A concentration of 450 ppm CO_2 has recently gained favour as a level with a fair chance of avoiding severe disruptions of the biosphere and the global economy. Given that this is only 70 ppm or about 35 years at current rates above the current level, stabilization at this level is a remote possibility. Most large energy-producing facilities committed today will not have reached the end of their planned lives in 35 years.

Whether 450 ppm is really a "safe" level is unknowable but it should be appreciated that yet higher levels carry ever-greater threats of disruption, possibly including the triggering of runaway feedback mechanisms that would surpass current anthropogenic emissions. Hence the need for urgency in adopting policies that will stop the rise of atmospheric CO_2 at the lowest possible level. Because global energy use is going to at least double as emerging economies industrialise and transportation represents almost 30% of current global energy use and the proportion is rising, energy for transportation must move substantially away from hydrocarbons if we are to stabilise atmospheric CO_2 levels. Currently, hydrogen is the only practicable CO_2-free alternative fuel. That is the simple case for the "Hydrogen Economy".

It is possible that battery storage may ultimately improve to a point where it can make a significant contribution but we simply cannot afford to wait for breakthroughs in battery technology to occur.

REFERENCES

[1]   Miller, A.I. and Duffey, R.B., "Co-production of hydrogen and electricity from wind and nuclear sources (NuWind©) using cavern storage for hydrogen", paper presented at ICENES 2005, the 12th International Conference on Emerging Nuclear Energy Systems, Brussels, Belgium, 2005 August 21-26.

[2]   "Impact of Wind Power Generation in Ireland on the Operation of Conventional Plant and the Economic Implications", ESB, 2004 February.
http://www.eirgrid.com/EirGridPortal/uploads/Publications/Wind%20Impact%20Study%20-%20main%20report.pdf.

[3]   Serban, M., Lewis, M.A. and Basco, J.K., "Source Kinetic study of the hydrogen and oxygen production reactions in the copper-chloride thermochemical cycle", 2004 AIChE Spring National Meeting, Conference Proceedings, 2004, pp 2690-2698.

[4]   "Ocean acidification due to increasing atmospheric carbon dioxide", The Royal Society, Policy Document 12/05, 2005 June, ISBN 0 85403 617 2.
http://www.pml.ac.uk/pml/news/RS%20ocean%20acidification%20report.pdf.

TABLE OF CONTENTS