

## French research strategy to use nuclear reactors for hydrogen production

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### Abstract

*The demand for hydrogen, driven by classical applications such as fertilisers or oil refining as well as new applications (synthetic fuels, fuel cells,...) is growing significantly. Presently, most of the hydrogen produced in the world uses methane or another fossil feedstock, which is not a sustainable option, given the limited fossil resources and need to reduce CO<sub>2</sub> emissions. This stimulates the need to develop alternative processes of production which do not suffer from these drawbacks.*

*Water decomposition combined with nuclear energy appears to be an attractive option. Low temperature electrolysis, even if it is used currently for limited amounts is a mature technology which can be generalised in the near future. However, this technology, which requires about 4 kWh of electricity per Nm<sup>3</sup> of hydrogen produced, is energy intensive and presents a low efficiency.*

*The French Atomic Energy Commission (CEA) thus launched an extensive research and development programme in 2001 in order to investigate advanced processes which could use directly the nuclear heat and present better economic potential. In the frame of this programme, high temperature steam electrolysis along with several thermochemical cycles has been extensively studied. HTSE offers the advantage of reducing the electrical energy needed by substituting thermal energy, which promises to be cheaper. The need for electricity is also greatly reduced for the leading thermochemical cycles, the iodine-sulphur and the hybrid sulphur cycles, but they require high temperatures and hence coupling to a gas-cooled reactor. Therefore interest is also paid to other processes such as the copper-chlorine cycle which operates at lower temperatures and could be coupled to other Generation IV nuclear systems.*

*The technical development of these processes involved acquisition of basic thermodynamic data, optimisation of flow sheets, design and test of components and lab scale experiments in the kW range. This will demonstrate the technological viability of the processes investigated and contribute to their optimisation. The extensive research programmes needed are led in the frame of the Hydrogen Production Project of the Generation IV Very/High Temperature Reactor System which helps mutualise the costs.*

*In addition, techno-economical studies are underway to evaluate the potential production cost and help the selection of one or two processes for which demonstrators will be built in the next few years.*

## Introduction

Today the world is facing tremendous energy challenges. There is a demographic explosion, which even in the most conservative scenario will drive the energy demand to high levels whilst at the same time fossil resources are becoming scarcer, and more particularly oil which bears most of the weight in the transportation area. Global warming is also becoming a major concern as the last Intergovernmental Panel on Climate Change concluded that anthropogenic greenhouse gases (GHG) are responsible for most of the observed temperature increase since the middle of the twentieth century. To address these difficulties, the first step is to look for ways to save energy whenever possible. Then, the part of GHG free sources – renewable energies (wind, solar, hydraulic, biomass,...) as well as nuclear energy – has to be increased in electricity production. Lastly, since the part of electricity in the final consumption of energy is less than 20% world wide, GHG free sources of energy have to look for other markets such as transportation, whether directly (electric cars) or indirectly via hydrogen (fuel cells,...) and/or process heat.

Hydrogen is produced currently from fossil fuels (less than 5% is produced by splitting water), and production is increasing steadily, mostly because of its use for refining crude oil and the more demanding standards of purity required. This alone is already stimulating interest in producing hydrogen by sustainable means. Moreover, the hydrogen market is bound to expand soon: hydrogen has been identified as a leading candidate to replace petroleum as part of the transition to a sustainable energy system, and major efforts are being conducted worldwide to develop the technologies and supporting activities required for this transition. A nearer term solution is to use the hydrogen produced together with a carbon source (biomass, coal, waste, CO<sub>2</sub>, ...) to make synthetic fuel. If the carbon feedstock is itself CO<sub>2</sub> free, such as biomass, the fuels obtained can be GHG free and sustainable. Recycling of CO<sub>2</sub> with the reverse water gas shift reaction could also prove to be interesting as the higher energetic cost required by this reaction could be offset by the elimination of the need for sequestration. A longer term and more hypothetical development could be the direct use of hydrogen to power cars, which would even further reduce CO<sub>2</sub> emissions. Hydrogen could also be used in the iron and cement industries as a reducing agent and also help these CO<sub>2</sub> intensive industries to significantly decrease their GHG emissions.

The French context has also to be taken into account. More than 80% of electricity is produced by nuclear power plants and the hour to hour variations of the electricity demand have to be absorbed by having some plants operating at intermediate power over significant periods of time. This situation presents the double drawback of not taking full benefit of an expensive investment as well as having to take careful steps when going back to full power in order to preserve the fuel cladding. Hydrogen production during off peak periods could help regulate the electricity demand and operate the nuclear plants in base load (Floch, 2007). This then requires hydrogen production means that are flexible and not investment intensive, as they would be used only on a part time basis. Another growing part of the French electricity supply comes from intermittent renewable energies (wind, solar...), and the impact of their intermittent nature could be lessened by storing the excess electricity produced via hydrogen production. Lastly, biomass is a very important carbon feedstock in France, which, as mentioned above, could be transformed, with the help of hydrogen addition, into a GHG free fuel for transportation.

CEA's strategy is hence to focus on processes which could be coupled to nuclear plants or renewable energy sources and thus be able to produce hydrogen in a sustainable way, by splitting the water molecule using GHG free electricity and/or heat.

To assess the economic competitiveness of the hydrogen production processes studied, alkaline electrolysis is taken as the reference: the current leading processes which use fossil fuel as reactant and heat sources are not sustainable and the unavoidable price increases in years to come aggravated by carbon taxation will inexorably drive the costs of production of these processes higher than that of alkaline electrolysis.

The French R&D focuses on three baseline processes which require high temperatures: high temperature steam electrolysis (HTSE), and the sulphur-iodine (S-I) and hybrid sulphur (HyS) thermochemical cycles. Alternative cycles able to operate at lower temperatures (Cu-Cl in particular) have also been investigated. All these cycles are being assessed from a feasibility point of view and some technical hurdles remain. Laboratory-scale experiments are ongoing and will give important

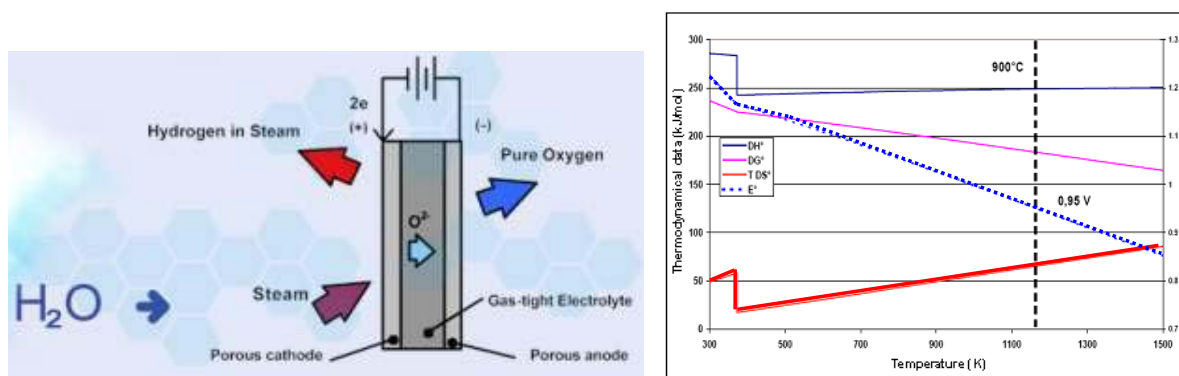
information about the viability of these processes and ways to optimise them. Another crucial aspect is the technico-economic assessment and the projected production price which should compete with that of alkaline electrolysis. The roadmap for France involves selecting one or two of these processes in the next few years and building large scale demonstrators, probably in the frame of international co-operation. This area of research strongly benefits from the Gen-IV hydrogen production project.

## High temperature steam electrolysis

This process, illustrated in Figure 1, has been carried out by the team of Doenitz in the 80s (Doenitz, 1982, 1985, 1988a, 1988b). They showed that the main drawback of water electrolyzers is their high electricity consumption. Electricity is known to be an expensive form of energy. As a result, electrolytic hydrogen is currently more expensive than steam-reformed hydrogen by a factor of at least two.

This drives the need to develop a high efficiency steam electrolyser for carbon-free production of hydrogen. From the thermodynamic viewpoint of water decomposition, it is more advantageous to electrolyse water at high temperature (800-1 000°C) because the energy is supplied in a mixed form of electricity and heat. In addition, the high temperature accelerates the reaction kinetics, reducing the energy loss due to electrode polarisation, thus increasing the overall system efficiency. Typical high temperature electrolyzers such as the German HOT ELLY system achieve 92% in electrical efficiency while low temperature electrolyzers can reach at most 85% efficiency (Doenitz, 1990). Another interesting feature is that they can be used quite efficiently in an autothermal mode as the need for heat at high temperatures is fairly limited.

Figure 1: Schematics of HTSE



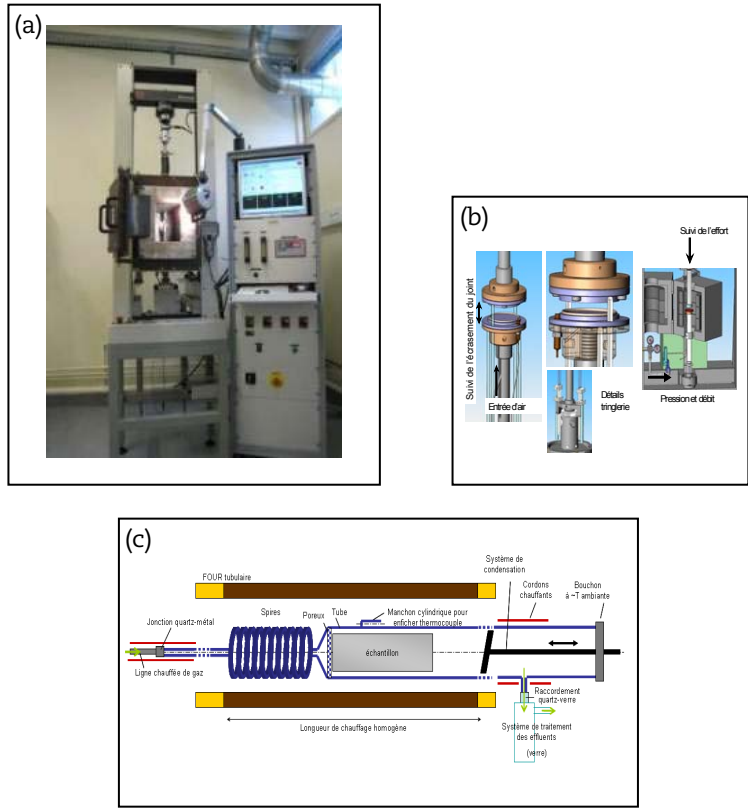
Since 2004 the CEA is engaged in a major programme of development of this technology (Le Naour, 2009). All aspects of the electrolyser are studied, through components and modelling, from materials to the plant. Two work paths are conducted in parallel.

The first focus was to invest in analytical tools (see Figure 2) to study one by one the breakthroughs identified from solid oxide fuel cell (SOFC) feedback and bibliography. Manufacturing capabilities of planar or 3-D cells were optimised to achieve cells with a high level of electrochemical performances, but also reliable and exhibiting a long durability. Specific devices have been developed to study the mechanical behaviour of these cells in HTSE environment. Similarly, a system has been developed for evaluating the effectiveness of coatings interconnectors against the evaporation of chromium that could pollute the electrodes. Always by way of example, solutions for glass or metallic seals are also characterised through a specific equipment dedicated to this purpose. All these devices are now operational and enable better understanding of the phenomena and evaluation of new solutions to address the key issues of high temperature steam electrolysis.

In the second work thrust, different designs of electrolyser stacks (as shown in Figure 3) have been developed to reach high surface and volume power. Due to the ultimate integration of this technology, where the electrochemical mechanisms, thermal, fluidic and mechanical properties are intimately linked on a few cm<sup>3</sup> of materials, a stack design provides an optimal compromise between these properties. We have thus developed architecture where the cells are not in close contact with

**Figure 2: Analytical tools for separate effect studies**

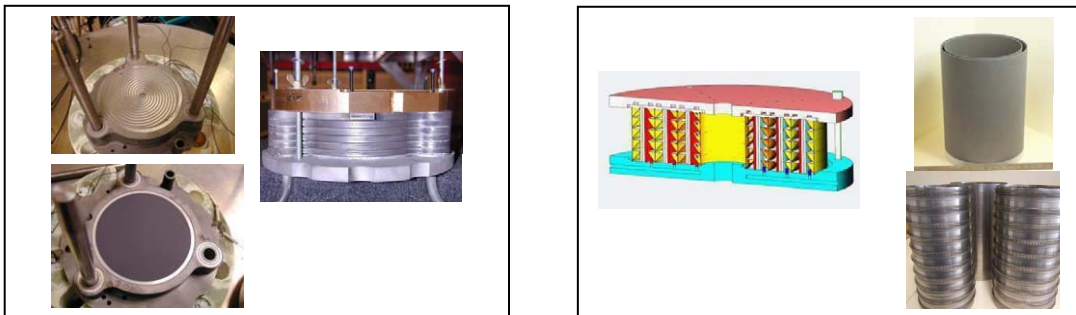
(a) MATTAHDHAUR device for four points bending tests on cells, (b) BAGHERRA device for seals characterisation and contact studies, (c) Specific equipment for dechromisation of interconnect metal studies



**Figure 3: Advanced architectures of stacks**

(a) SOFC derived architecture

(b) Innovative coaxial design



the interconnectors to minimise mechanical stress. Similarly, an alternative design increases the volume capacity to minimise the number of modules to implement in a electrolyse plant linked to a nuclear reactor and then simplify the global plant. Today, we have achieved performances on three to six cells stacks of these different designs operating under electrolysis conditions with production levels of around 0.1 L<sub>H<sub>2</sub></sub>/h.cm<sup>2</sup> (4 litres H<sub>2</sub> per hour for the whole stack).

Flow sheets and technico-economical evaluation are also in progress for different coupling solutions between a Gen-II or -IV nuclear plant and the HTSE technology (Rivera-Tinoco, 2008). We start this year the design of a lab scale plant able to test in 2011 our solutions of stacks in a complete system (target of scale: 2 000 l/h H<sub>2</sub>). This pilot will be used to study the behaviour of the

system, and in particular to the autothermic operating mode with all the loops of gas output heat recovery. Finally, an installation based on a 5 MW heat power source is also underway to test in 2013 a high temperature electrolysis system under representative industrial conditions.

## Sulphur-iodine cycle

This process, shown in Figure 4 was proposed by General Atomics (O'Keefe, 1982; Brown, 2003) and first demonstrated by JAEA (Kubo, 2004). It is now being tested in San Diego in an international collaborative effort between Sandia National Laboratories (SNL), General Atomics Corporation (GA) and the French Commissariat à l'Énergie Atomique (CEA). This project is being conducted as an International Nuclear Energy Research Initiative (INERI) project supported by the CEA and the US DOE Nuclear Hydrogen Initiative. The objective of initial testing of the sulphur-iodine (S-I) laboratory-scale experiment (equipment shown in Figure 5) was to establish the capability for integrated operations and demonstrate hydrogen production from the S-I cycle. The first phase of these objectives was achieved with the successful integrated operation of the SNL acid decomposition and CEA Bunsen reactor sections to produce the required heavy and light acid phase product streams, and the subsequent generation of hydrogen from that material in the HI decomposition section. This is the first time the S-I cycle has been realised using engineering materials and operated at prototypic temperature and pressure to produce hydrogen. Although this operation successfully produced and separated the expected heavy and light acid phases, control of the feed flow rates to the Bunsen reactor must be improved in the next phase of testing to optimise the composition of the output phases. The heavy phase material subsequently processed in the GA's HI decomposition section produced hydrogen at the expected rates. Several subsystems within the HI skid were successfully demonstrated during this integrated experiment with a maximum hydrogen production rate of ~50 litres per hour (Moore, 2008). Based on these initial results, several process and hardware improvements were identified that will be implemented before resuming integrated testing. The next stage of tests will focus on integrated operations for longer run times.

Beside this integrated experiment, CEA is focusing its efforts on specific areas of the process in order to optimise the flow sheet. Efforts concern all three parts of the S-I process. For the Bunsen section, extensive R&D is under progress to optimise the stoichiometry of the reaction: currently there is an excess of water (16 moles of  $H_2O$  when only two are stoichiometrically required) and an excess of iodine (9 moles of  $I_2$  when only one is required) in order to ensure a complete reaction, a good separation of the two acid phases and also to prevent side reactions. On the sulphuric acid section, work is in progress to optimise the flow sheet and, within the frame of a European programme, on the process heat reactor which is a key component. The most intensive part of the work is on the iodine section: sophisticated measurement apparatuses have been developed to get a more accurate knowledge of the liquid-vapor equilibria in the water-iodine-iodhydric acid ternary system (Doizi, 2007, 2009a; Larousse, 2009). This will help the design of the reactive distillation column which is considered to be the most efficient option for HI processing (Goldstein, 2005). Collaboration on this distillation column is underway with the Bhabha Atomic Research Centre in India.

Figure 4: Schematics of the sulphur-iodine cycle

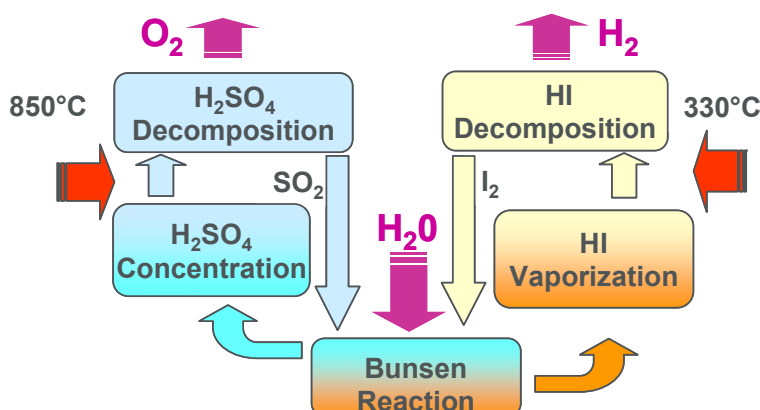
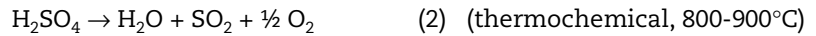
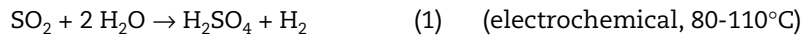


Figure 5: I-NERI S-I loop at General Atomics



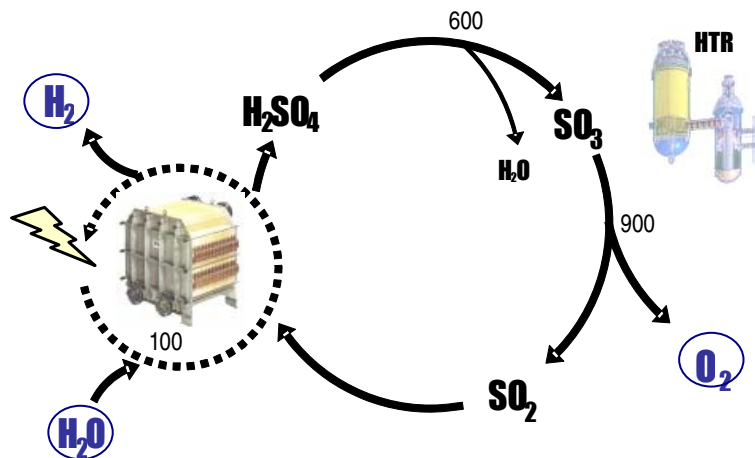
**Hybrid sulphur cycle**

Like the sulphur-iodine cycle, the hybrid sulphur (HyS) process (Westinghouse, 1975) is being developed to produce hydrogen by water-splitting using heat from advanced high temperature nuclear reactors. It is a leading candidate among thermochemical cycles for coupling with a high temperature gas reactor (HTGR). A two-step process shown in Figure 6, the HyS cycle is one of the simplest thermochemical cycles, and moreover the simplest all-fluid one. The two reactions comprising the cycle are:



The sulphuric acid decomposition reaction to regenerate  $\text{SO}_2$  and to produce oxygen, (2), is common to all sulphur cycles, including the sulphur-iodine (S-I) cycle. What distinguishes the HyS process from the other sulphur cycles is the use of sulphur dioxide to depolarise the anode of a water electrolyser, as shown in reaction (1).

Figure 6: Schematics of the hybrid sulphur cycle





The use of  $\text{SO}_2$ -depolarisation reduces the theoretical cell voltage for water electrolysis to 0.158 V at infinite dilution and 0.262 V for an anolyte consisting of 50 wt.% sulphuric acid. In contrast, conventional water electrolysis has a theoretical cell voltage of 1.23 V. Under practical operating conditions, a  $\text{SO}_2$ -depolarised electrolyser (SDE) is expected to require approximately one-third as much electricity as a conventional water electrolyser for the same hydrogen production. An overall thermal efficiency for converting heat to hydrogen with this process of 40-45% (HHV-basis) is deemed possible based on detailed flow sheet analysis. The HyS cycle therefore has the potential for high efficiency and competitive hydrogen production cost. Furthermore, it has been demonstrated at laboratory scale.

At CEA, the studies on this process have started more recently. The two critical components of the process components are the high-temperature decomposition reactor and the SDE. Beside the European project mentioned above on the process heat reactor for  $\text{SO}_3$  decomposition, CEA studies therefore focus on the electrolysis section, with a pilot now in operation in Marcoule (see Figure 7). Indeed, a major challenge for the HyS process is the development of an efficient, cost-effective SDE. Prevention of  $\text{SO}_2$  migration through the separation membrane of the electrolyser, which leads to undesired sulphur deposits, remains a major technical hurdle to overcome. Like for all electrolytic processes, economic competitiveness of the cycle will also depend on the minimisation of electrolysis overvoltage and on components lifetime (membrane, interconnectors).

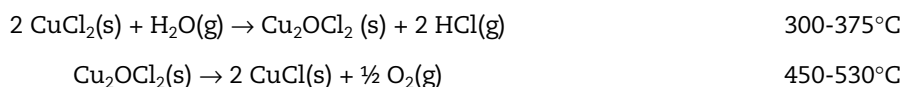
**Figure 7: Hybrid sulphur pilot at CEA/Marcoule**



## Other cycles

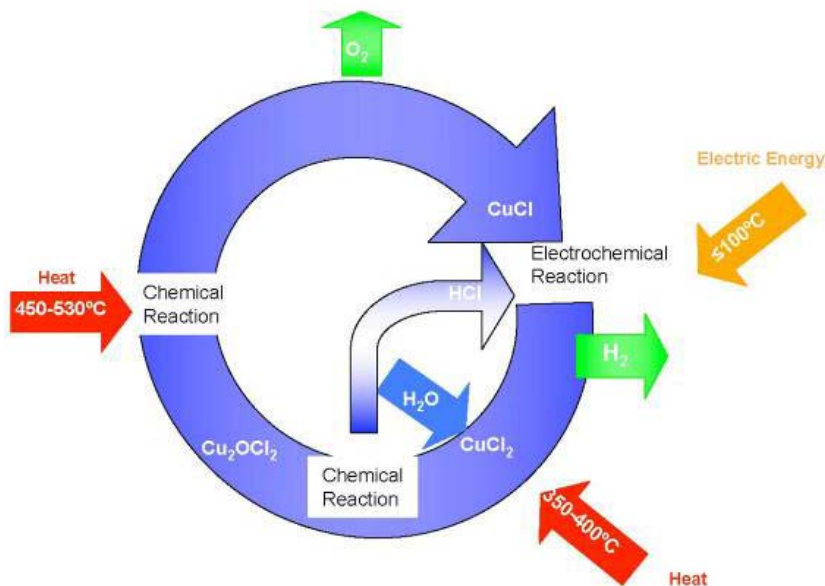
The whole set of possible thermochemical cycles has been considered and ranked based on a list of predefined criteria such as levels of temperatures required, rarity or toxicity of the reactants, number of electrochemical steps,... This led to the selection of a few cycles of interest (Mg-I, Ce-Cl and Cu-Cl). After further evaluation, Cu-Cl (shown in Figure 8) was retained. This cycle presents the advantage of dealing with only moderate temperature reactions ( $< 530^\circ\text{C}$ ), which offers the possibility of coupling it with other Gen-IV systems such as the Sodium Fast Reactor and the Supercritical Water Reactor.

This process is already being thoroughly investigated. The CEA is therefore focusing its actions on specific points where techniques developed for the sulphur-iodine cycle can bring new insights (Doizi, 2009b; Borgard, 2009). Specifically, CEA has designed an experimental set-up which utilises optical spectrometry (used for liquid-vapour equilibrium measurements of water-iodine-iodhydric acid) to study the speciation of the gaseous phase for the two chemical reactions of the Cu-Cl cycle, namely:



In particular, the first (hydrolysis) reaction is not thermodynamically favored and only takes place if a large excess of water is used. Gas phase composition measurements allow *in situ* study of the reaction to better understand the influence of various parameters such as water stoichiometry, temperature or residence time, which therefore improve the assessment of the viability of this cycle.

**Figure 8: Schematics of the Cu-Cl cycle**



### Techno-economic studies

Research is underway on four different processes, however, a significantly sized pilot (100 m<sup>3</sup> of H<sub>2</sub>/hour) will be done on, at best, two of the advanced processes, due to the high cost of such a demonstrator. The French roadmap (Figure 9) calls for decisions in 2009 and then in 2012. An important aspect of the selection is the cost of production which can be achieved with these different processes. As discussed earlier, the costs of production will be compared to those of conventional electrolysis, which is the sustainable reference process. Our methodology was the following: after acquiring the required thermodynamic data and characterising the materials considered, we established a flow sheet, which gave us on one end the energetic cost and on the other end, helped us design the plant, size the components, evaluate the cost of construction and obtain the cost of capital investment. The cost of operating and maintenance was also evaluated. This estimation is not yet complete for all the advanced processes and will be published at a later stage. However, with the current knowledge and the current flow sheets, we have estimated the energetic costs of the HTSE, S-I, HyS and Cu-Cl processes and compared them to those of conventional electrolysis. The potential advantage of these advanced processes is that instead of electricity, part of the energy can be provided as heat, which is cheaper and this would give them an edge on low temperature electrolysis, providing the investment costs stay low. But, as Table 1 shows, so far, only HTSE presents a significantly lower economic advantage and R&D is still needed on the other cycles to increase their efficiency and bring it closer to their thermodynamic potential.

Another more immediate benefit of these techno-economic studies is their use, via sensitivity studies, to identify the areas in which sizeable gains could be made to help focus R&D efforts.



Figure 9: French roadmap

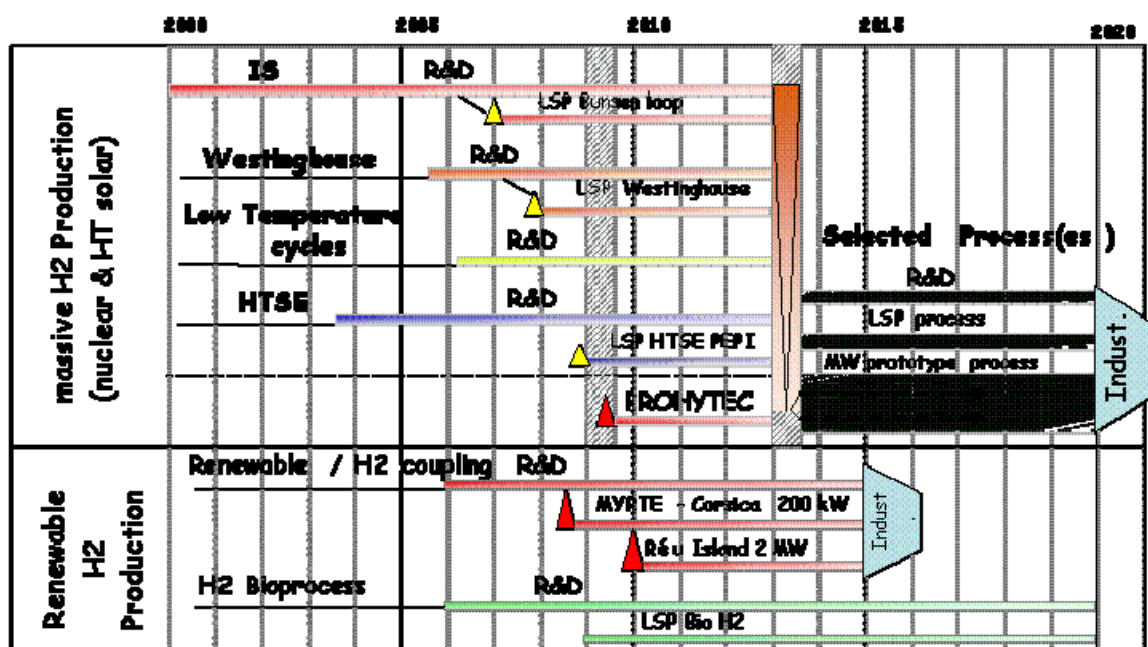


Table 1: Energy consumption costs for the different processes with the current efficiencies – the cost of heat is linked to that of electricity but using the efficiency of the reactor providing the heat ( $\eta = 50\%$  for HTSE, S-I and HyS and  $40\%$  for Cu-Cl)

Electricity unit cost (€/MWh)	Heat unit cost (€/MWh)	Alkaline electrolysis	HTSE	S-I	Hy/S	Cu/Cl
40	$40 \cdot \eta$	2.1	1.6	2	1.8	1.9
50	$50 \cdot \eta$	2.6	2	2.5	2.2	2.4
60	$60 \cdot \eta$	3.2	2.4	3	2.7	2.9

## Conclusions

Dwindling fossil fuel reserves and GHG emissions are driving the need for clean hydrogen production. For the short term, hydrogen will likely be used as an intermediate to produce synthetic fuels from carbon source (biomass, CO<sub>2</sub>, C, waste,...). Hydrogen economy and the use of hydrogen as an energetic vector can also happen but is a longer-term goal.

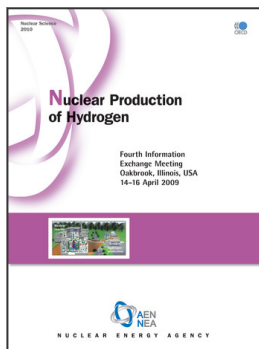
In the French context, hydrogen can be used to regulate electricity production and to use to their fullest the nuclear plant investments. They can also help optimise the use of intermittent renewable energy sources.

The reference process against which we will compare advanced processes is the alkaline electrolysis. This is a mature and flexible process, offering excellent lifetime of cells (20 years currently). Progress can still be made in investment costs, but this represents only 20 to 30% of the production cost. Among the advanced processes, HTSE currently offers the best prospects, providing it can address some key issues such as reliability and lifetime. Thermo-chemical production still represents a major challenge, as its net efficiency has to increase and the investment and operating costs have to decrease. For these processes, the coupling issues (co-existence of nuclear and chemical safety constraints) have also to be addressed.

Careful economic studies are needed to select the most promising processes and to guide the R&D effort to provide the greatest impact. Finally, international collaboration is essential to share the R&D and split costs.

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