Nuclear Systems for Hydrogen Production: State of Art and Perspectives in Transport Sector

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Abstract: A technologically feasible transition towards a realistic and sustainable hydrogen economy (i.e. on large scale and without carbon dioxide emissions) could be made through the use of nuclear energy. In fact, nowadays hydrogen production methods without the employment of fossil fuel represent only a low share of the total production; but the use on large scale of hydrogen produced by "carbon-based" sources is neither environmentally nor economically meaningful.

In the present paper, besides a deep evaluation of the state of art of hydrogen production methods *via* nuclear source, it has been proposed an energy scenario analysis (based on the hydrogen produced by the thermo-chemical Iodine-Sulfur process fed by High Temperature Gas Reactor (HTGR) included in a symbiotic nuclear fuel cycle), focused on the China region, that would meet the sustainability criteria in both the energy and environmental domains for transport sector.

Keywords: Nuclear Energy, Hydrogen Production, Scenario Analyses, High Temperature Gas Reactors.

INTRODUCTION

The beginning of the twenty-first century has been characterized by two major problems: the sharp increase in global energy consumption and the environmental sustainability of economic growth in developing countries, in particular with regard to China and Asian countries.

The economic and industrial growth in developing countries has led to a sharp increase in the consumption of fossil fuels, thus the quantity of world reserves available is shrinking at very high speed with the possibility of their lack in the coming decades (despite the slowdown in consumer spending which has been followed by since the crisis of 2008-2009). In addition, the large use of fossil fuels has resulted in a sharp increase in the emissions of gases that contribute to global warming, a source of major climatic changes on a global level.

Taking these issues into account it is clear that it is necessary for governments of all nations to adopt policies aimed at reducing the consumption of fossil fuels and increase the use of alternative sources for the generation of electricity and in the transport sector, the sectors whose emissions have the greatest impact on the greenhouse effect and consequently the environment.

In 2011, following the accident at Fukushima has dropped the use of nuclear energy, to stop power plants in several different nations; nuclear power provided 4.9% of primary energy worldwide, in France 41.2%, Japan 7.7%, while the United States 8.3%.

One of the possible solutions to the energy problems of the twenty-first century could be the increased use of nuclear energy for electricity generation and for industrial production of hydrogen in order to use it as, for example, fuel in the transport sector, or, more generally, as a more environmental-friendly energy carrier.

As known, hydrogen would also allow a better exploitation of nuclear installations:

- Firstly because the energy, produced at a "constant" rate (while the demand is, by definition, variable), may be partly accumulated in the form of hydrogen, subsequently consumed following the users' requests
- Secondly due to the fact that adopting new reactors with an higher core coolant output temperature (e.g. up to 900/1000 °C for HTGR) it would be possible to implement more efficient hydrogen production systems without needing of transforming the heat produced inside the reactor core into electricity before using it for hydrogen production (so obtaining a substantial improvement in the global energy efficiency)

In this way, nuclear energy can penetrate more deeply into the energy market.

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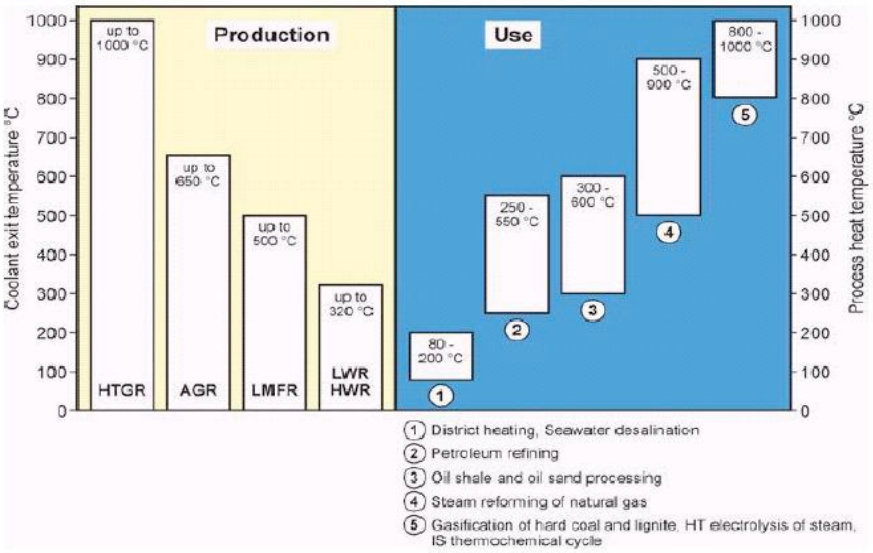

Figure 1: Temperature requirements of some industrial processes and heat capacity of nuclear reactors [4].

1. HIGH-TEMPERATURE GAS REACTORS (HTGR)

In general terms, almost all of the electricity generated by nuclear is achieved using Light Water Reactors (LWR) and steam turbines that exploit the Rankine cycle with efficiencies lower than 35% (having inlet temperatures to the turbines below 350°C, with obvious penalty in terms of performance). However the HTGR, due to their characteristics [1-3], may offer some advantages. The Gas Turbine Modular Helium Reactor, GT-MHR (one of the possible HTGR), could reach an efficiency nearly 50% adopting a regenerative Brayton cycle. In addition to the improved efficiency of the cycle, the HTGR reactors have the considerable advantage to have a high core coolant outlet temperature (around 950°C) that meets the requirements for many industrial applications (Figure **1**), including the production of hydrogen without the CO_2 emissions.

1.1. Historical Evolution of HTGR

The technological development of HTGR [5] began in the early '60s, with the construction in the UK of the Dragon reactor (1963÷1976, Figure **2**), and is continued for more than half a century. Several reactors were designed and built over the years (e.g. AVR-10, FSV, THTR-300, etc.), the most recent creations have been the HTTR (Japan) and the HTR-10 (China).

The Dragon reactor was the first to use TRISO type fuel (TRIstructural-ISOtropic coated fuel particle) still in use today. The American reactor FSV (Fort Saint Vrain, 1976÷1989, Figure **2**) proved the effectiveness of the prismatic core design, obtaining a net thermal efficiency of 39% for the generation of electricity, while using an indirect cycle with steam turbine. However, some technical problems, including difficulties in the circulation of the coolant, did increase significantly the

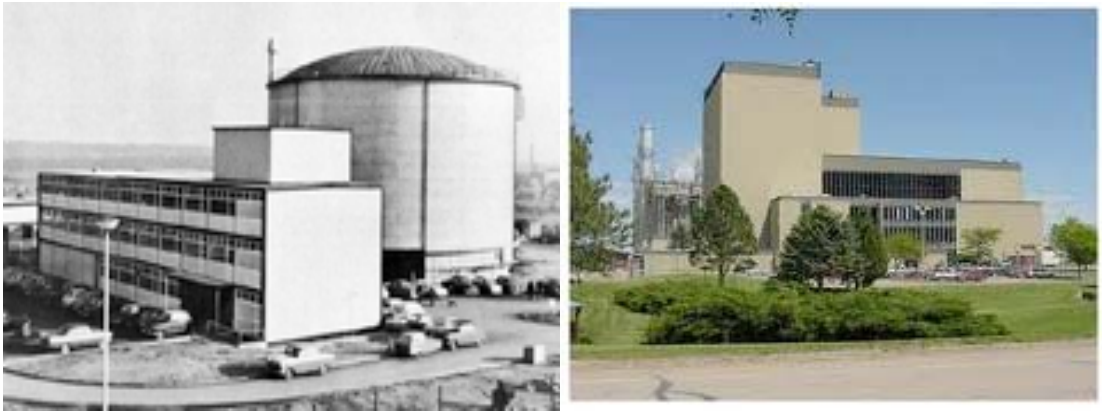

Figure 2: Dragon (on the left) and FSV Reactor (on the right).

costs of the system, making uneconomical investing in the reactor. Another great commercial plant that used as fuel thorium was the German THTR-300 (Thorium High Temperature Reactor, 300 MW_t, 1986÷1989), with the configuration of the pebble-bed kernel type (which has a core with fuel elements in continuous charging and arranged stochastically).

As already mentioned, the latest operating reactors are located in Asia, the HTTR (High Temperature Engineering Test Reactor, 30 MW_t, Figure **3**) with prismatic kernel configuration in Japan and the HTR-10 (High Temperature Reactor, 10 MW_t) with a pebble-bed configuration in China. The Japanese reactor has an outlet temperature of the coolant (helium) of 950°C which allows the use of heat at a temperature even higher than 850°C for industrial thermal processes. This feature allows the reactor to be potentially used for numerous industrial processes and not only for the generation of electricity.

Figure 3: View of the reactor HTTR [6].

In the first half of the '90s a research team led by General Atomics (GA), and also funded by the U.S. DOE has designed the GT-MHR (Gas Turbine Modular Helium Reactor) [7]. The design is based on a 600 MW_t reactor, with prismatic core and an output temperature of the coolant of 850°C, combined with a gas turbine for electricity generation. The thermal efficiency of the system is close to 50% and the GA estimates the cost of the electricity to be competitive with other power sources [8].

Furthermore, South Africa has developed the PBMR reactor (Pebble-Bed Modular Reactor, Figure **4**), 400 MW_t with coolant output at 900 °C, designed for the generation of electricity, the production of hydrogen and for further heated industrial processes [9].

Since 2001, the international consortium for the development of Generation IV innovative nuclear power plants (Generation-IV International Forum, GIF) has selected six nuclear systems that can be built and become operational by 2030÷2040 and will enable the production of energy in a sustainable manner, both from an environmental and economic point of view, with particular attention to safety, non-proliferation and the minimization of nuclear waste [10, 11]. Among the six selected systems, one, the Very High Temperature Reactor (VHTR) is an enhanced version of the "classic" HTGR.

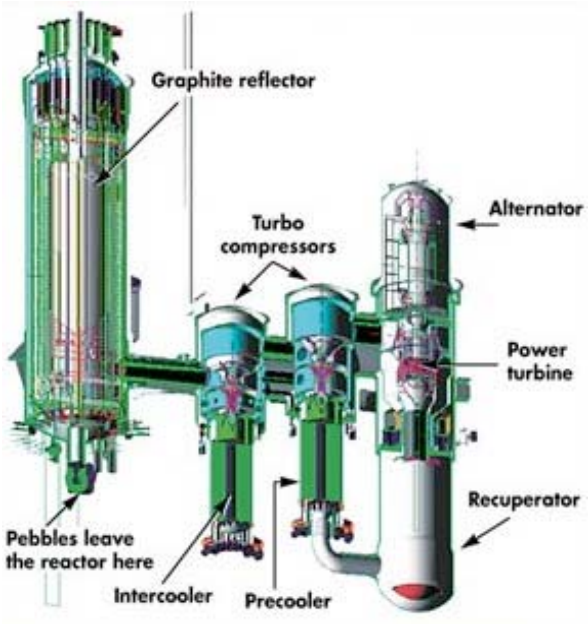

Figure 4: Schematic drawing of the PBMR system [10].

Since the VHTRs have the potential characteristic of being efficient both in the production of electrical energy that hydrogen (as well as useful for other industrial applications), the DOE has placed particular emphasis in the supply chains proposed by GIF on the development of these reactors. This led to the design and financing of the NGNP program (Next Generation Nuclear Plant) for demonstration and validation (up to the pre-commercial level) of the high efficiency in the generation of hydrogen and energy [12].

1.2. Characteristics of Modern HTGR

As regards safety, the use of graphite and ceramic materials in the construction of the core, allows HTGR to withstand high temperatures, even in case of accidents and breakdowns. Moreover, the low power density proper to this type of system, helps to limit the maximum temperature reached during an accident.

From the neutronic point of view, in the case of an abnormal temperature rise of the core, the negative

temperature coefficient of the fuel would lead anyway at shutdown of the reactor (in a similar way as provided for all the classical "western" designed reactors).

In case of switching off, the radioactive isotopes decay heat can be removed from the core by solely thermal conduction without the need for auxiliary safety systems. The characteristics of the coolant (helium) also contribute to the safety of the reactor: in fact this element is a noble gas with optimal chemical, thermal and nuclear properties. Some of these features directly mitigate the potential consequences in case of an accident with loss of the cooling fluid [13].

Currently, the type of fuel used in nuclear reactors consists mainly of low-enriched uranium (<20%) and everything suggests that it will continue to be used in this form in the next few years.

In an alternative way, the use of thorium as a fuel [14] is very interesting for some countries with large reserves of this element and, more in general, in view of a long-term perspective (being the thorium much more abundant than uranium). All HTGR reactors mentioned above (HTTR, HTR-10, GT-MHR and PBMR) are able to use this alternative fuel.

More generally, the HTGR support multiple types of fuel and several fuel cycles are being studied [15-18] (some foresee the use plutonium from decommissioned nuclear weapons) [2] and reduce the amount of minor actinides (elements that contribute most to the long-term hazards of nuclear waste) during the production of electricity [3]. In fact, the fuel in the form of microparticles (the so-called Coated Particle, CP) used in HTGR has shown (experimentally, in facilities suitable to simulate high neutron fluences [19]) a capacity of burnup (i.e. of energy produced per unit mass of fuel) greater than 700 GWd/t: this is a key feature to get a fuel cycle with consumption of trans-uranic elements (TRU deep-burn fuel cycle); it should also be highlighted that, among other things, the high burnup reduces the amount of exhausted material and therefore the costs of reprocessing [20, 21].

1.3. HTR-PM

Currently in China, near the city of Rongcheng in Shandong Province, the company Huaneng Shandong Shidao Bay Nuclear Power is manufacturing two 250 MW_t High Temperature Pebble-bed Modular reactors (HTR-PM, Figure **5**) that will power a single 210 MW_e steam turbine power with an efficiency around 42% (Figure **5**); the cost of this plant is about 300 million Euros. Table **1** shows the design parameters of the reactor. These reactors are part of the $ 5 billion project Rongcheng Nuclear Power Industrial Park that aims to build 18 units of 210 MW_e (corresponding to 36 reactors of 250 MW_t) [22].

Compared to the HTR-10, the helium temperature output from the core and the power density is increased by using a larger amount of heavy metals (7 g instead of 5 g) for each fuel element. The fuel inside a pebble runs out after approximately 15 steps within the core with an average burnup of 90 GWd/t_U. During operation, in the annular cavity, which separates the bars of the core and the Reactor Pressure Vessel (RPV), flows helium at 250°C in order to limit the RPV temperature.

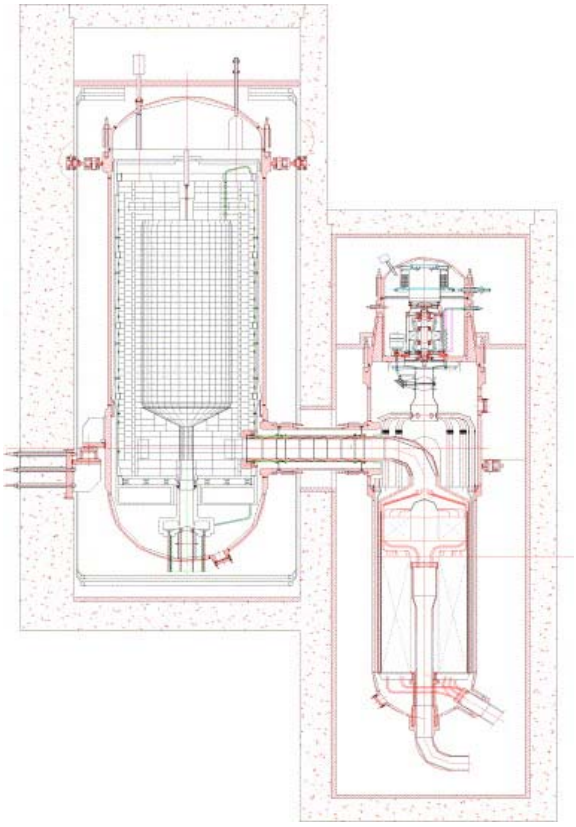

Figure 5: HTR-PM section.

The design of the annular type was initially proposed to increase the output thermal power of the PBMR reactor. In this case the core is a central column of pebble graphite without fuel, so the fuel is moved radially outwardly leading to an efficient exchange of conductive heat to the outside, this ensures that in case of a depressurization of the cooling fluid the fuel temperature does not exceed the design limit of 1600°C.

Table 1: HTR-PM Design Parameters [13]

Parameter	Value
Electric power estimated	210 MW_e
Plant thermal power	2 x 250 MW_t
Life cycle	40 years
Core's average power density	3.22 MW/m^3
Power generation efficiency	42%
Helium pressure in the primary circuit	7 MPa
Temperature helium input/output	250/750 °C
Type of fuel	TRISO (UO_2)
Weight of each element in heavy metal	7g
Enrichment of fresh fuel	8.9%
Effective diameter of the core	3 m
Equivalent height of the core	11 m
Fuel elements for core	420,000
Average burnup time	90 GWd/t_U
Type of steam generator	Single pass helical coil
Steam pressure	13.24 MPa
Steam temperature	566 °C
Water inlet temperature	205 °C
Water flow to the turbine inlet	673 t/h
Turbine type	High pressure condensation turbine

1.4. GTHTR300

By partnering with a number of industries, the Japan Atomic Energy Agency (JAEA) has designed a family of plants called GTHTR300, marketable in the near future and with the capability to produce in a competitive way electricity, hydrogen or both to adapt to the demands of the market.

The system combines a basic GTHTR300 (600 MW_t reactor) and a gas turbine for the generation of electricity [23]. The outlet temperature of the helium in this case is 850°C, operating efficiency is 45.6%. The studies related to the design of the plant began in 2001 and were completed in 2004. The technologies and components needed for the system have been tested on a 1:3 scale model. The passive safety and the cost of electricity of 3.5 c$/kWh have been confirmed by further analyses [24].

Subsequently has been designed the GTHTR300+, a plant for the production of electrical energy with helium at 950 °C at the reactor outlet and an efficiency of 50%. Then the GTHTR300C and the GTHTR300H, two systems for the cogeneration of hydrogen (Table **2**). The GTHTR300C uses technologies common to the whole family of plants and consequently would be used as a prototype, while the GTHTR300H is designed for maximum hydrogen production possible by using the electrical energy produced to support the IS process and the necessary operations to keep in function the nuclear reactor.

In Figure **6**, the design of the JAEA's GTHTR300C is shown. The cogeneration of hydrogen is achieved by inserting a helium-helium heat exchanger (IHX, Intermediate Heat Exchanger) in series between the reactor and the gas turbine.

This design offers several advantages, e.g. the helium cooling circuit passes only once through the turbine, so eliminating the need to separate the circulation of fluids. The ratio of the thermal power generation and electric power can be varied through different types of operations, studied by JAEA, without compromising the efficiency of electricity production.

A secondary loop allows the transport of helium from the IHX to the chemical section of the plant, where hydrogen is produced at a safe distance from the nuclear section: this design feature greatly simplifies the safety measures. In Figure **7** is shown a sample diagram (based on the IS process, better detailed in a later section) is shown.

The JAEA would like to build a prototype of the GTHTR300C production system by 2020 to verify its reliability and demonstrate the technology to begin marketing the GTHTR300 family. Later in the future, the Nuclear Energy Vision 2100 document (prepared by the Office for Strategic Research of the JAEA) highlights how the construction of 120 new HTGR reactors (72 GW_t) from 2030 onwards for the production of hydrogen would meet the national energy demand and achieve the goal of reducing the CO_2 emissions (compared to 2000 emission levels) by 50% in 2050 and 90% in 2100.

Table 2: Design Parameters for the GTHTR300 Plants [13]

	GTHTR300+ Power generation	GTHTR300C Cogeneration	GTHTR300H H_2 Generation
Reactor thermal power	600 MW_t/module	600 MW_t/module	600 MW_t/module
Reactor lifetime	60 years	60 years	60 years
Power generation efficiency	50%	47%	38%
Net electricity output	300 MW_e	174 MW_e	34 MW_e
H_2 plant effective heat rate	N/A	220 MW_t	505 MW_t
H_2 conversion efficiency	N/A	43%	41%
H_2 Production	N/A	0.64 milion m^3/day 58 tons/day	1.41 milion m^3/day 126 tons/day
Total plant efficiency (net)	50%	45%	40%

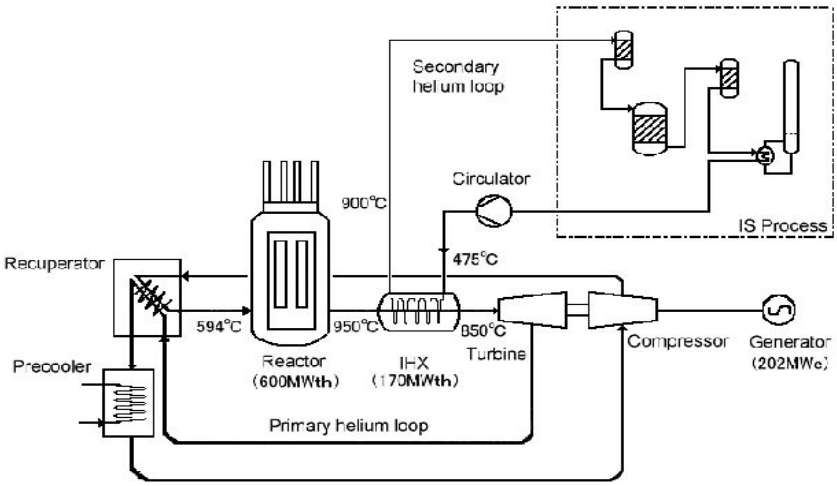

Figure 6: GTHTR300C functional diagram [25]

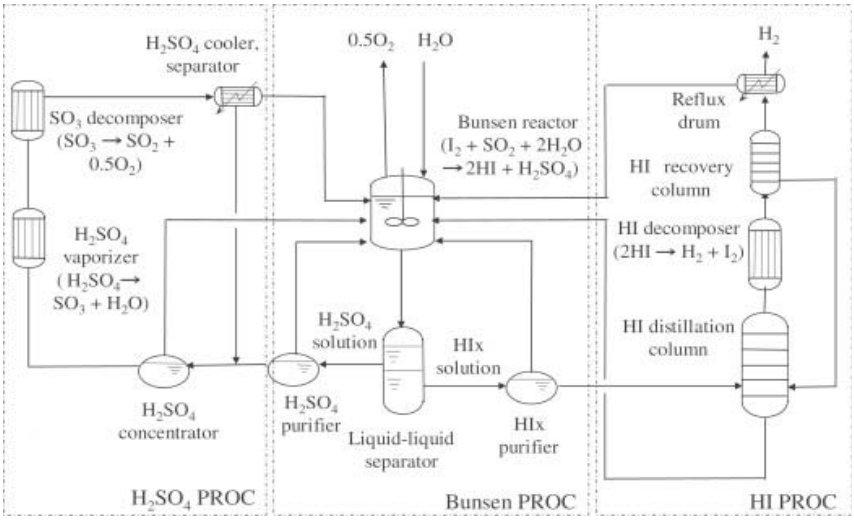

Figure 7: Diagram of the thermochemical IS process [26].

2. PROCESSES FOR HYDROGEN PRODUCTION

2.1. Water Electrolysis

The electrolysis of water is a commercial method for H_2 production that uses electrical energy. Approximately 4% of worldwide hydrogen is produced in this way. Emissions of greenhouse gases generated by this process are related to the source used to produce electricity, with an HTGR reactor these would be close to zero.

By using the existing power grid to transmit the energy needed to the process the need for construction of infrastructure to support the plant would be considerably lower.

Because this process would use only the electricity generated by HTGR reactor as input energy, the nuclear and the chemical plant would be completely separated.

The biggest production unit uses 2 MW_e to generate 458 Nm^3/h of H_2 (40 kg/h) and 242.5 Nm^3/h of O_2 occupying an area of 4x14 m^2 (including the spaces required for maintenance). To improve the performances and costs of large-scale production new high pressure units and polymer electrolyte membranes are being studied.

The total cost for a state of the art plant is therefore about 450 $/kW, and the price for large scale production of hydrogen is estimated at 2.6÷3.1 $/$kg_{H2}$ (based on the electricity price in the United States varies between 0.04 to 0.05 $/kWh); the cells life cycle for this system is at least 10 years [27].

A 100÷300 MW_e HTGR can provide the energy needed by 50÷150 units of the biggest electrolysers for a theoretical combined production of 24250÷73750 Nm^3/h of hydrogen (with high index of purity) and 12125÷36375 Nm^3/h of oxygen. The production volume can be increased by multiplying nuclear reactors units.

The efficiency for electric production of a direct cycle HTGR, with core coolant outlet temperature of 950 °C is around 50%. Given an additional loss due to the transformation from AC to DC is achieved a total efficiency close to 40%, based on the higher calorific value, for hydrogen production through electrolysis with a HTGR.

The production of electricity through HTGR has an estimated cost of 3.5÷4.0 c$/kWh [28]. The life cycle of the nuclear plant is at least 40÷60 years long. Based on these parameters, the ultimate cost of the hydrogen produced by hydro-electrolysis with an HTGR, is estimated to be 2.3÷2.6 $/$kg_{H2}$. Since the primary cost of the electrolysis process is due to electricity, it is highly desirable to produce hydrogen at the times when the electricity demand is low, being its price considerably lower. Furthermore, the stored hydrogen can be used to produce electricity through fuel cells or hydrogen combustion turbines, to compensate possible peaks in the current demand.

Since large units of electrolysers are already available on the market, the future challenges in the development of this technology lies in the creation of GEN-IV VHTR plants with high efficiency (~50%) for energy production.

2.2. Electrolysis of Steam

The electrolysis of steam can be obtained through a isothermal thermodynamic process in which the energy required is provided in the form of electricity. The increase of the temperature at which the process takes place decreases the amount of electrical energy required, with approximately the same gradient. In the extreme case in which the temperature exceeds 2000 °C the water molecules start to separate into hydrogen and oxygen, through thermolysis and the need for electricity tends to zero. Moreover, the electrolysis of steam at high temperatures (the so-called high temperature electrolysis or HTE) may be more efficient from the economic point of view when compared to electrolysis at low temperatures, as it is energetically less expensive to provide energy as heat. Thanks to high temperatures there is an increase in electrochemical reactivity and electrical conductivity of the conductors which contributes to an increase in the efficiency of the process.

The design of the actual electrolytic cells provides operations from 800 to 900 °C. At high temperatures, the efficiency can reach 50% (greater than 40% obtainable by the electrolysis of water at low temperatures). However, achieving high performance and a long life cycle for the electrolysis unit is proving to be a challenge. Although the HTE is not yet a commercial reality, in recent years significant progress has been made to demonstrate this system of hydrogen production.

In 2008, the INL (Idaho National Laboratory) has completed a test of 1000 h for a demonstrator of this

technology at a laboratory scale. During the experiment was obtained a peak production of 5.7 Nm^3/h with the absorption of 18 kW. In 2009, a unit of 10 planar cells was maintained operative for 2500 h. Currently the research is focusing on the fundamental mechanisms of degradation of the electrodes and the electrolyte through the simulation at atomic level of their interaction.

There are several options for integrating a nuclear reactor in the HTE. The system can use a reactor that supplies the water vapor needed by the electrolyser and one to obtain the necessary electric energy. Alternatively, a single system can simultaneously generate the steam and electricity needed for the process as shown in Figure **8**.

The GA has estimated the performance of a plant for the combined production of hydrogen through HTE with a conceptual study. Heat loss associated with the piping were ignored and so the processing losses from AC to DC; the pressure losses in the components are estimated to be around the 1%. The electricity is cogenerated with an efficiency of 50.5% from a DC gas turbine utilizing helium. The 10 units for the HTE require a total of 18.7 MW_e, of which 0.41 MW_e can be provided through a process of heat recovery. To support the process 3.59 MW_t of hot steam are provided by the reactor [12].

The overall efficiency of the process is estimated to be:

$$h = \left(\frac{21.3}{[3.59 + (18.7 - 0.41)/0.505]} \right) \times 100 = 53.5\% \text{ (HHV)}$$

The GA estimates that a large commercial plant would consist of four reactors of 600 MW_t HTGR cogenerating steam and electricity to support the plant for hydrogen production, based on HTE technology, consisting of 292 units made up of 8 modules of planar solid oxide electrolytic cells (SOECs). Assuming that the plant reaches the technological maturity, the GA estimates that the final cost of hydrogen will be around 2.22 $\$/kg_{H2}$ [12].

2.3. Steam Reforming of Methane by Nuclear Power

Steam reforming of methane (SRM) is the most economical and developed method in the world for hydrogen production. Conventionally, the heat necessary for the reaction is supplied by the combustion of additional methane, this method is not sustainable from an environmental point of view, especially in the (likely) scenario of a growing demand for hydrogen.

The temperatures necessary for the reaction are between 800 °C and 850 °C, obtainable by a HTGR reactor. Using a nuclear reactor to provide heat to the reaction would save at least 37% of the methane and reduce the CO_2 emissions by the same percentage [29].

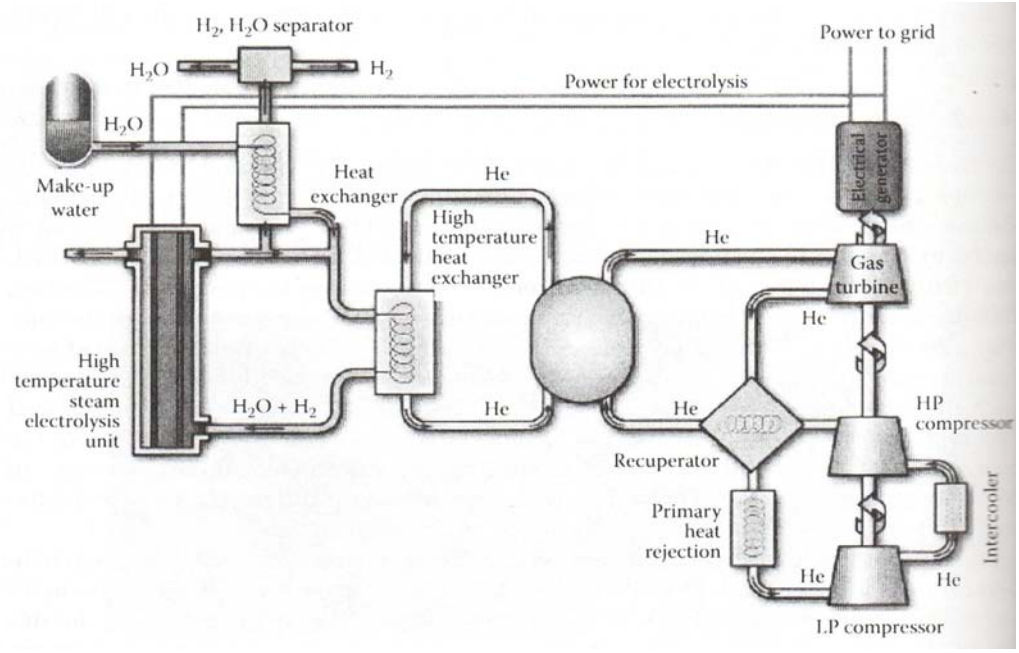

Figure 8: Plant for the steam electrolysis [13].

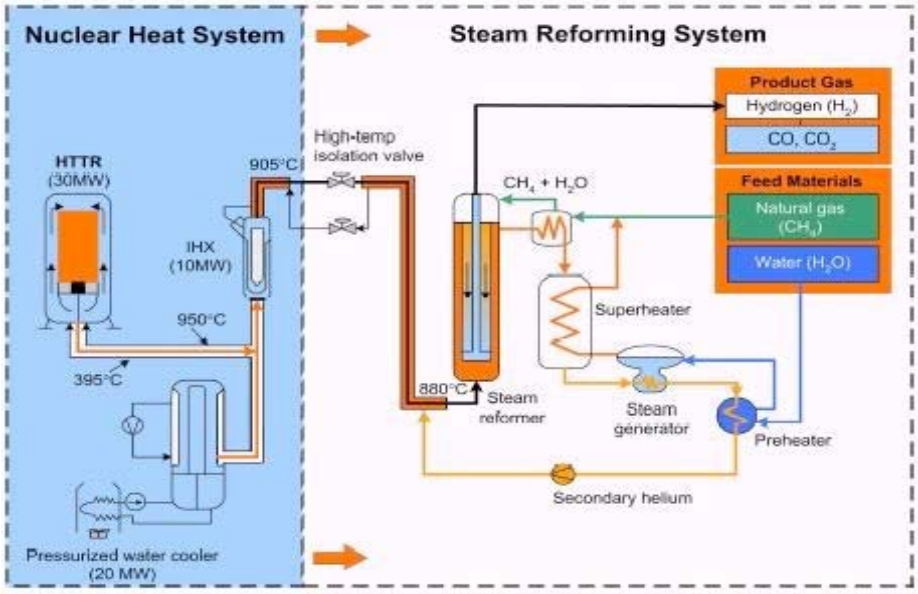

Figure 9: Diagram of SRM by nuclear power [30].

The JAEA has designed and built the reactor HTTR also with the aim to evaluate the process of SRM based on the use of nuclear energy. The scheme of the built combined system is shown in Figure **9**.

In their plant, an Intermediate Heat Exchanger (IHX) allows to transfer to the bottomer helium loop a 10 MW_t of thermal power. Obviously, some technical safety features are taken into account, such as the higher pressure of the secondary cycle to prevent contamination deriving from possible leaks in the primary loop inside the IHX. The reactor provides helium at a temperature of 880 °C at the inlet of the steam reformer, in which the gas circulates outside the catalyst tubes heating by forced convection in countercurrent flow.

Helium enters from below and after being passed through numerous plates comes out at a temperature of 585 °C.

The tubes contains catalytic pellets of Ni/Al_2O_3 through which the gas and the steam necessary for the reaction flow. The inlet mass flow rate is 1290 kg/h of methane and 5160 kg/h of steam. The mixture of the reacting gases is heated up to 450 °C at a pressure of 4.5 MPa before entering the reformer at the top and flowing downward to the catalytic bed. Here the methane and other light hydrocarbons undergo the reforming process.

The final products of the reaction, that reach a maximum temperature of 830 °C, flow upward through the inner tubes, exchanging heat with the gaseous reactants that flow annularly to the tubes in the opposite direction, and leave the reformer at a temperature of 580 °C and a pressure of 4.1 MPa. The exit gas is cooled in a condenser and separated into vapor and non-condensable gases, which include hydrogen, carbon monoxide and residual methane.

The conversion rate of methane to hydrogen is estimated around 68% for the system described above and for a large-scale commercial plant is thought to be up to 80% [31].

The JAEA's test facility has achieved a production capacity of 120 Nm^3/h of hydrogen, using 43.2 kg/h of methane and has served for testing all the key components of the reactor's IHX system [32]. A 400 kW electric heater was used to heat the helium at 880 °C instead of nuclear heat. The gas was used to provide energy to the reforming process, generate steam and preheat the methane and steam prior to their entry into the reformer.

The most important goal for the improvement of the process is to increase the efficiency in the production of hydrogen, and then becomes a priority to improve the conversion rate of methane by decreasing the pressure at which the steam-reforming chemical reactions take place. By calculating the chemical balance of the reaction at 800 °C is obtained a conversion rate of 64% at 4.5 MPa, 81% at 2 MPa and about 92% with a pressure of 1 MPa during the process.

However getting low pressures is a problem since for the efficient cooling of the reactor and its

components is required an helium pressure of at least 4 MPa. This would result in a difference of 2 MPa compared with the pressure of the process gases, unsustainable (at temperatures around 900 °C) for the current metals in which are manufactured the catalyst tubes.

In the case of the system used by JAERI, the tubes were designed to withstand a pressure of 0.5 MPa from the outside and 1 MPa from the inside.

The development of ceramic materials for the catalyst tubes is one of the possible solutions in order to obtain low pressures during the reforming process and consequently a better efficiency.

2.4. Thermochemical Iodine-Sulfur Process (I-S)

The process (Figure **10**) consists of three chemical reactions to dissociate the hydrogen and oxygen in gaseous form; a large amount of heat and a lower electrical energy is used as input energy and the only resource "consumed" is water. Therefore, it is a cyclical process and does not generate greenhouse gases. For these reasons, there is great interest in developing nuclear technology in order to make achievable and sustainable the production of hydrogen through thermochemical process.

The three reactions that produce hydrogen are as follows [6]:

1. $I_2 + SO_2 + 2\ H_2O \rightarrow 2\ HI + H_2SO_4$ (120°C); Bunsen reaction

   - The HI is then separated by distillation or liquid/liquid gravitic separation.

2. $2\ H_2SO_4 \rightarrow 2\ SO_2 + 2\ H_2O + O_2$ (830 °C)

   - The water, SO_2 and residual H_2SO_4 must be separated from the oxygen byproduct by condensation.

3. $2\ HI \rightarrow I_2 + H_2$ (450 °C)

   - Iodine and any accompanying water or SO_2 are separated by condensation, and the hydrogen product remains as a gas.

Net reaction: $2\ H_2O \rightarrow 2\ H_2 + O_2$

In practice, it is required an electric power equal to 20% of the total energy necessary for the process to activate all the components, such as pumps, compressors, etc. indispensable for the system.

Since the '70s many nations (including Japan, France, United States, South Korea and Italy) have invested in the development process.

The JAEA is the organization that has achieved the most important results to demonstrate the feasibility of producing hydrogen through the thermochemical IS process by using the HTGR; it has already successfully performed tests in reduced scale of the process IS and has planned to assess the performance of the system model through the construction of a prototype in reduced scale, followed by the demonstration of hydrogen production with a flow rate of 1000 Nm^3/h using the HTTR+IS system.

The Bunsen exothermic reaction produces two aqueous solutions of sulfuric acid and hydrogen iodide starting from water, sulfur dioxide and iodine. An excess of water and iodine allows the reaction to become spontaneous and a solution rich in hydrogen iodide promotes the subsequent separation steps. However, the excess water and iodine requires a careful control of the flow of the acids in the following steps, in particular in the steps for the decomposition of hydrogen iodide. Numerous studies are also in progress with the aim of reducing the excess reactants in the process in order to minimize the costs and simplify the process.

The sulfuric acid (H_2SO_4) coming from the Bunsen reaction is purified and concentrated before being decomposed in H_2O and SO_3, which then separates into SO_2 and oxygen in a gaseous state (the process requires a temperature of 850 °C). The sulfuric acid decomposition process is well-known and well-proven, the most important issue from the technical point of view is the resistance to heat and corrosion of the decomposers. Many industries, interested in the problem, are manufacturing and testing components to assess the performance of the I-S process.

The hydrogen iodide produced by the reaction Bunsen through numerous passages is concentrated and then decomposed into hydrogen and iodine, which will be used again as a reagent. The steps that involve the hydrogen iodide present great possibilities for improvement and in the diagram in Figure **7** have been incorporated some of the most innovative techniques. Given the concentration of HI several steps are combined in order to reduce the excess iodine and water before the final distillation with the use of cells for electrodialysis and carbonic membranes for osmosis.

Toshiba Corporation has proposed to include in the HI decomposer an absorber of iodine in order to enable the following cogeneration process:

(1) $2HI \rightarrow H_2 + I_2$ (400°C)

(2) $Co + I_2 \rightarrow CoI_2$ (400°C)

(3) $CoI_2 \rightarrow Co + I_2$ (600°C)

(4) $2HI \rightarrow H_2 + I_2$

As has been experimentally observed, absorbing the I_2 produced by the reaction (1) in the presence of the reaction (2) is obtainable a decomposition ratio of HI up to 80% in the reaction (4). The cobalt and iodine are then regenerated in the endothermic reaction (3).

The helium exiting the IHX is transported to the chemical plant section to transfer heat to the process, exchangers and decomposers. A total of 21.7 MW_e electric power is used in the purification process of the HI *via* electrolyzers (13 MW_e), for the circulation of the helium in the secondary loop, for the pumps, the circulation of the hydrogen gas and other functions. The process, using 21.7 t/h of water, reaches the production of 26829 Nm^3/h (2.4 t/h) of H_2 and 13515 Nm^3/h of oxygen.

The overall efficiency of the process is defined as the HHV of the hydrogen produced in relation to the totality of the energy consumed, in which the heat input and the equivalent thermal energy provided by electricity during the process are considered.

The net efficiency is greatly affected by how efficiently the electrical energy is produced. In the case of the GTHTR300C plant, the electricity is produced on site through a gas turbine with a direct current efficiency of about 47%, therefore, based on a system diagram like that of Figure **6**, the efficiency of hydrogen production is equal to 44%.

3. EXPLOITATION OF HYDROGEN IN THE TRANSPORT SECTOR

Hydrogen is an ideal energy carrier to be introduced in the transport sector; its combustion at lower temperatures compared to other hydrocarbons limits the production of NOx (although internal combustion engines or gas turbines fuelled by hydrogen usually burn it at low dilution ratio in order to achieve the same output power of other fuels with a calorific value significantly higher than hydrogen; as a consequence the higher H_2 concentration leads to higher adiabatic temperature and higher NOx emission), moreover does not contain sulfur and carbon, therefore does not produce greenhouse gases such as CO, CO_2 and SO_2.

Another system for moving vehicles is the use of electric motors powered by fuel cells that use hydrogen as fuel.

The operating principle of the fuel cells is based on the ionization of the molecules of the fuel or the comburent to obtain an electromotive force through an electrochemical reaction. The electrons separated by the ionization are channeled in an electrical circuit supplying an electric current proportional to the speed of the chemical reaction; ultimately ions, electrons and molecules of comburent/fuel combine in a final waste product.

The difficulty in ionizing many molecules limits the choice of fuel elements or molecules with quite weak bonds. The fuels used are molecular hydrogen (H_2) and syngas (a mixture of hydrogen and carbon monoxide) while the atmospheric oxygen is used as comburent because of its abundance and because its reaction with hydrogen generates steam vapor.

Amongst the fuel cells mature from the commercial point of view those which use only hydrogen as a fuel are the PEMFC (proton exchange membrane cells), the AFC (alkaline fuel cells) and the PAFC (phosphoric acid fuel cells).

The PEMFC use an aqueous base, a membrane polymer acids as electrolyte and electrodes coated with platinum; the operating temperature is low, lower than 100 °C. Thanks to these features, the PEMFC allow the use of pure hydrogen as a fuel. This type of cell is currently the most widely used for private vehicles or commercial use (eg forklift for handling of loads). The hydrogen fuel is processed at the anode, where the electrons are separated from the protons on the surface of catalyst platinum-based. The protons pass through the membrane in the direction of the cathode, while the electrons travel the external electrical circuit that connects the anode and cathode, generating electricity. At the cathode, always formed with electrodes in precious metals, protons and electrons are combined with oxygen, pure or extracted directly from the air, producing water as the only waste product.

The AFC utilize an alkaline electrolyte such as potassium hydroxide (KOH) and pure hydrogen as fuel. The working temperature, which was originally between

100°C and 250°C, is decreased up to around 70°C. To accelerate the reactions that occur at the anode and cathode is not necessary to use platinum as catalyst, but may be used various non-precious metals, among which the most commonly used is nickel. Given the low temperature and the speed of the reactions the efficiency in the conversion of fuel into electricity in some applications is up to 60%.

In PAFC anode and cathode consist of catalytic platinum finely dispersed within a structure of carbon or silicon carbide which contains phosphoric acid (which has the function of electrolyte). This type of fuel cell is particularly resistant to poisoning by carbon monoxide, allowing the use of hydrocarbons as fuel; it has a lower efficiency compared to other types of cells (37÷42%); in the case of cogeneration plants, however, reaches 80%.

The most common method for storing hydrogen is the use of pressure vessels, where it is stored in gaseous form at 350 or 700 bar. In this way the gas is stored at room temperature eliminating the need of a plant for thermal insulation present instead in case of using liquid hydrogen.

The development and improvement of the fuel cells technology is being carried out by several private companies and government agencies, often in mutual cooperation. Several prototypes of electric vehicles powered by FCEV have already been made by many manufacturers. In particular, as an example, we may report Honda's FCX Clarity that is already on sale (although in a limited number of copies) in California. In the FCX hydrogen is stored at 350 bar inside a fuel tank of 171 l (equivalent to 3.92 kg of H_2) for a total range of about 240 miles (386 km). The fuel consumption is about 1 kg every 60 miles (97 km) traveled, with only the emission of water vapor [33].

4. ANALYZED SCENARIO: CHINA

The choice of China as the country on which perform a preliminary analysis [30] of the future production capacity of hydrogen from nuclear energy has been determined by several important factors:

- √ It has the largest number of nuclear reactors under construction and design;
- √ It is the largest consumer of coal in the world;
- √ It is the state that emits the greatest amount of carbon dioxide;
- √ It has a strongly growing economy that will lead to a further increase in energy requirements;

To reduce the environmental unsustainability of the national energy situation, the Chinese government aims for 2020 to generate 15% of energy through non-fossil sources and to lower of 40÷45% the CO_2 emissions per unit of GDP compared to 2005 levels.

The Chinese government has decided to use nuclear energy as a third source of domestic energy, investing heavily in the construction of many reactors (over 50), to obtain in future the national energy independence. Currently, the Chinese government has planned to build new nuclear reactors by 2020 to achieve the production capacity of about 58 GW_e by this source, in order to meet the energy needs of coastal areas far away from coal mines and reduce energy dependence of fossil fuels.

The PWR in China are now the most common typology of reactor; their design is mainly based on the AP1000 and CPR-1000. Currently 14 are operating PWR plants with a total capacity of about 11590 MW_e. Another 28 plants are under construction for an estimated power of about 30550 MW_e. China plans by 2015 to increase nuclear capacity to about 40000 MW_e [34].

In addition, as already mentioned, 18 units of HTR-PM with a total capacity of 3800 MW_e are under construction.

The code used for the analysis of the fuel cycle of the scenario under consideration is the Nuclear Fuel Cycle Simulation System (NFCSS), (screenshot of typical results is in Figure **11**), developed by the IAEA to determine, amongst the other parameters, the quantities of materials needed to feed the fuel cycle and the amount of residual nuclear waste [35].

It was therefore suggested that the total power of PWR reactors built in China at the end of 2018 will be about 38580 MW_e corresponding to a thermal power of about 108670 MW_t (assuming conservatively efficiency around 35.5%). Using part of the spent fuel coming out from these reactors, it would be possible to feed HTGR reactors with a total capacity of about 13950 MW_t. If we consider analogous to the HTR-PM plants under construction, you could feed 27 dual reactor units with a power of 500 MW_t.

In this scenario, these units are exclusively dedicated to the production of hydrogen through IS cycle.

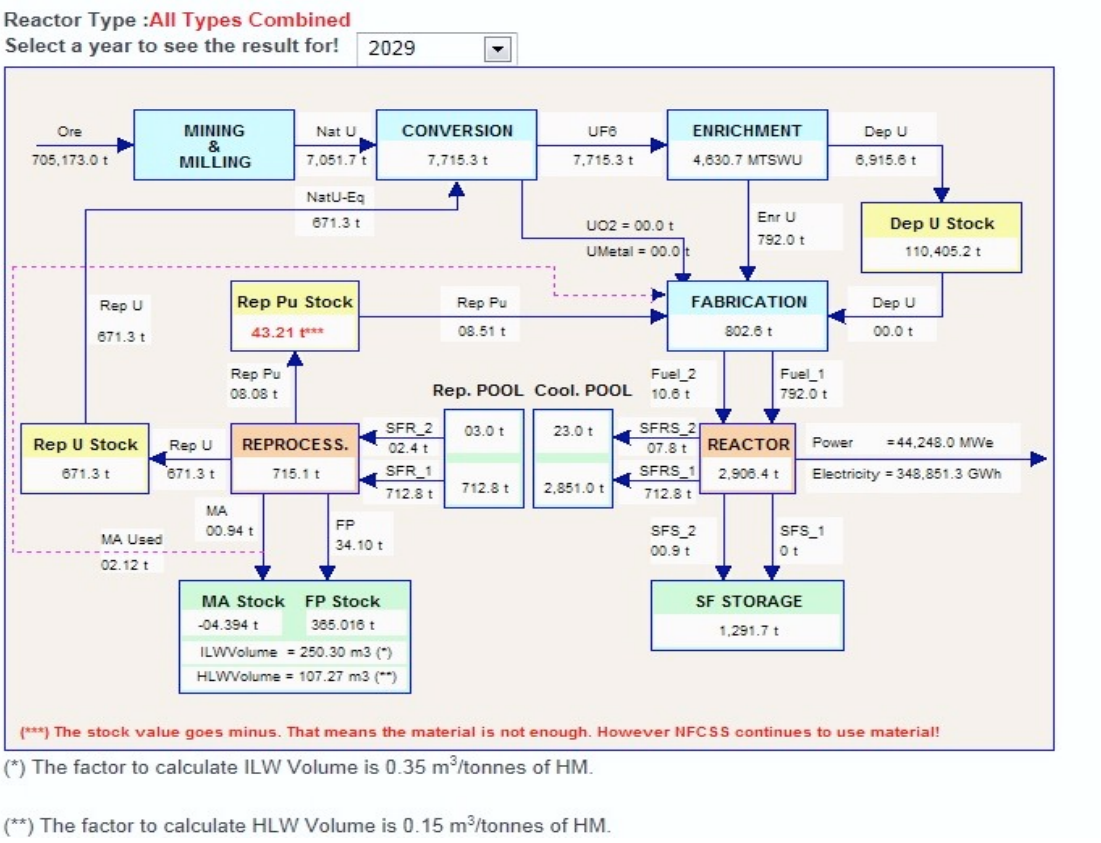

Figure 10: Screenshot of a NFCSS code output.

Table 3: Postulated Commissioning of HTR-PM Units

Year	2018	2021	2022	2023	2024	2025	2026	2027	2028	2029
Installed units	1	2	2	2	2	2	4	4	4	4

The time period studied for scenario analysis goes from 2011 to 2050.

As regards the installation of 27 units HTR-PM (which may be fed by PWR built in China in late 2018) has been hypothesized the commissioning according to the time sequence shown in Table **3**. Increasing the number of units introduced annually is assumed to be due to the maturation of the technology used. As already mentioned, each unit has a power output of 210 MW_e.

The use of Pu-based fuel in the reactors HTGR allows to use the amount of plutonium and minor actinides produced by the PWR reactors after their reprocessing. Results deriving from the code show that PWRs, for the time period of the scenario, would produce about 219312 t of Pu and 27334 t of MA. The use of HTGR reactors in cascade allows to consume these stocks by reducing them in 2050 to about 66092 t of Pu and 16462 t of MA. At the end of 2050, the total volume of waste, amount to about 11500 m^3.

The system for the exploitation of the IS process designed by the JAEA (and illustrated in the previous paragraphs) combined with a reactor of 500 MW_t HTGR would allow to obtain an hydrogen production of about 1.23 kg/s (106.3 t/d) and an electric power of approximately 34 MW_e [13].

In the hypothetical scenario, the 27 HTR-PM units would lead to a daily production of about 2870 t of hydrogen (through a system similar to that designed by JAEA), annually would be achievable a production of approximately 943000 t of hydrogen (assuming a load factor of 90%).

Considering that (as already mentioned) the tank of a FCX Clarity contains 3.92 kg of hydrogen at the time of maximum productivity of hydrogen of the scenario

(hypothetically) it would be possible to supply more than 700000 vehicles.

5. CONCLUSIONS

This work deals with the possibility to create an alternative energy scenario for hydrogen production without the use of fossil fuels, and in particular by means of nuclear energy. The need to study an environmental sustainable scenario arises from a deep analysis of global energy consumption and of hydrogen production methods. Actually, a possible implementation of an hydrogen economy could not be carried forward, in an environmental friendly way, without matching with a nuclear strategy. Nowadays the most popular production methods foresee the use of fossil fuels either for thermal power production or for their chemical content.

Starting from this preliminary overview, a thorough analysis on new generation HTGR is carried on, with the aim of identifying the advantages and drawbacks of such reactors for the hydrogen production purpose. Then the most popular hydrogen production methods coupled to nuclear reactors are deeply studied.

Particularly, a future scenario, based on the hydrogen produced in a thermo-chemical I-S based chemical plant fed by HTGR included in a symbiotic nuclear fuel cycle, has been analyzed for the China region. Considering the emerging and huge Chinese civil nuclear program, it has been feasible to study a hypothetical strategy of hydrogen production for this country *via* nuclear power.

With the purpose of developing such study, it has been used the NFCSS code provided by IAEA, that helped us to evaluate the material flows during the period covered by our simulations. Furthermore, China has been chosen due to, amongst the others, its environmental impacting strategy of economic growth. The code allows us to evaluate the amount of material involved, from the front-end to the back-end of the cycle related to the strategy adopted.

At the end we found that, at the time of maximum productivity of hydrogen, it would be (hypothetically) possible, by the energy infrastructure proposed by us, to supply enough fuel for more than 700000 vehicles without consuming additional mineral resources and/or increasing pollutant emissions (e.g. greenhouse gases) in the environment.

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