ISTITUTO NAZIONALE DI FISICA NUCLEARE

Sezione di Milano

PRODUCTION OF NUCLEAR HYDROGEN Nu2 OR HYDRICITY BY HIGH TEMPERATURE NUCLEAR REACTOR

Mauro Bonardi[1,2,3], Flavia Groppi[1,4], Simone Manenti[1]

[1]*INFN-Sezione di Milano, Dip. Scienze Fisiche Università di Milano, LASA, Via F.lli Cervi 201, 20090 Segrate, Milano, Italy*
[2]*International Committee of the American Nuclear Society, at http://www.ANS.org*
[3]*Scientific Secretary of Interdivisional Group of Radiochemistry and Radiation Chemistry applied to environment, health and industry (GIR) of the Italian Society of Chemistry (SCI), at http://wwwGIR.mi.infn.it*
[4]*A.N.P.E.Q., Italy*

Abstract

Hydrogen in both gaseous or liquid form is an *energy vector* also called *hydricity*. Hydrogen gas in very rare in the atmosphere and in geological sites, thus it must be produced with advanced technological devices, mainly by water splitting. This clean energetic utility can be produced efficiently by dedicated high or very high temperature nuclear reactors of *Generation IV*, instead of using the traditional methods based on steam reforming of either light hydrocarbons or coal.

The advantages are evident due to the *high power density* of a nuclear power plants and the almost complete lacking of green-house gaseous emissions, heavy metals and radioactive ones, typical of traditional thermoelectric power plants. This method for producing NuH2 is clean, safe and environmental friendly. There are being discussed the main *thermochemical* and *pyrochemical* routes for water splitting in H_2 and O_2, by using at least three designs of nuclear reactors of *Gen IV*, that are described in some details.

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INTRODUCTION

After 20 years of moratoria, more than 40 new nuclear power plants (NPPs) are in construction all over the world and in Europe too. They add to the presently operating 440 NPPs, that do produce surprisingly alone, well 16% of total world electricity, a vector of energy that is expected to double in the next 25 years. In Italy about 10 000 hydro-PPs (600 of which with capacity larger than 1 MWe) do not reach the 12% of the total electric demand, while photo-voltaic units (PV) do contribute for the 0.0015% of the total (*i.e.* 5 GWe.h on a total of 342 000 GWe.h consumption on 1997-2006 period, ~ 18% of which is produced in France by NPPs and imported from France and indirectly from Switzerland, Austria and Slovenia).

According to OECD estimates the number of NPPs will double in the next few decades, reaching a world electric capacity of 30-35% of the total, like is presently in the 22 OECD Countries. Italy would have the short-medium term goal to cover a 25% of internal electricity demand by NPPs, that means either **8 EPR**s of AREVA-Siemens consortium [1] or **12 AP1000**s of Westinghouse-Toshiba [2].

1.1. Nuclear Energy vs. Solar Energy

In other papers we showed that the availability of natural nuclear fuels $^{(234),235,238}U$ and ^{232}Th is largely sufficient for several *thousand of years* – especially if fast- and thermal-breeder reactors would be implemented in *Gen IV* NPPs – making nuclear energy a *potentially inexhaustible resource* [3]. Moreover our opinion is that this kind of energy is very safe, clean and characterized by low cost and negligible rad-waste by-production, which can be efficiently transmuted by dedicated nuclear power transmuter plants (NTR) or accelerator driven systems from transmutation (ADS or ADT) [4,5,6,7,8].

As a short digression, unlikely the *inexhaustible* and attracting *nuclear energy of the Sun* has - on the average - a small *surface power density* in Italy (145-155 W.m^{-2} at North latitude, 160-170 at Center and 180-190 at South) [9], with a further decrease during PV conversion, that for Si panels reaches the 11% in the more optimistic case [10,11,12], making the extensive use of solar energy an utopia. This is also due to the low *theoretical photosynthesis quantum yield* $\phi^{max} = 0.125$ (*i.e.* mol of CO_2 fixed or O_2 evolved per mol of photons absorbed, where ϕO_2 may be somewhat higher than ϕCO_2), which operationally has a mean value of 0.081 only [13]. This means that the theoretical number of **8** moles of photons (*i.e.* 8 einstein) are necessary either to fix by plants and some bacteria (*i.e.* containing *chloroplasts*) 1 mole of CO_2 or to produce 1 mole of O_2 by *chlorophillian photosynthesis*, but in practice more than **12** moles of photons are necessary (practical efficiency ~ 8%). Apart a limited use for supplying low temperature utilities, obviously the more updated use of solar concentrators and heat storage in molten salts (*e.g. solar thermodynamic* ARCHIMEDE project, Priolo Gargallo, Sicily), cannot change the available surface power density values. Thus, even increasing of 10^3 folds the solar plants

capacity, the total amount of energy obtained would be negligible in any case.

Conversely, it would be a serious possibility the use of *gaseous* or *liquid hydrogen* as a clean *energy vector* for the next future. The world heavy chemical industry already does produce ~ 50-60 Mtonnes of hydrogen annually, but in order to supply all the present consumption of energy it would be necessary to increase this quantity of 100 folds up to 5 Gtonnes annually.

According to some projections, in the 2050 it would be possible to produce 50% of energy vectors as *hydricity* and the other 50% as *electricity* by using NPPs. This kind of hydricity is normally called nuclear hydrogen or NuH2 [10,11,12].

2   HIGH TEMPERATURE GAS COOLED NUCLEAR REACTORS

Regarding the *Gen IV* reactors, these are a set of designs currently being researched. Most of these designs are generally not expected to be available for commercial construction before 2030, with the exception of a version of the Very High Temperature Reactor (VHTR) called the Next Generation Nuclear Plant (NGNP) to be completed by 2021. Research into these reactor types was officially started by the *Generation IV International Forum* (GIF) [14] based on eight technological goals. The primary goals are to improve nuclear safety, improve proliferation resistance, to minimize rad-waste and natural resource utilization, and to decrease the cost to build and run such NPPs. An Integrated Nuclear Energy Model is central to standardized and credible economic evaluation of *Gen IV* nuclear energy systems. The innovative nuclear systems considered within *Gen IV* require new tools for their economic assessment, since their characteristics differ significantly from those of current *Gen II* and *III* NPPs.

The *Gen IV* reactors can be classified in two main types, some based on the *UOT* process, others on *Partitioning and Transmutation* (P&T) of fuel [15,16,17,18,19,20,21,22,23,24]. They are devided in: **A**) Thermal reactors: Very-High-Temperature Reactor (VHTR); SuperCritical-Water-cooled Reactor (SCWR); Molten Salt Reactor (MSR); **B**) Fast reactors: Gas-cooled Fast Reactor (GFR); Sodium-cooled Fast Reactor (SFR); Lead-cooled Fast Reactor (LFR). Some of these reactors, whose details and schematics can be found at GIF web-site [14], have the capability to produce hydricity if medium- and high-temperatures are reached (*i.e.* VHTR, GCR and LFR).


3   NUCLEAR HYDROGEN, NuH2 OR HYDRICITY

The world chemical industry does produce ~ 50-60 Mtonnes of hydrogen annually, basically by *steam reforming* of light hydrocarbons C_nH_m to CO and H_2 and high temperature conversion of coal though the *Fisher-Tropsch gasification*, followed by *water gas shift* reaction to CO_2 (Dussan's reaction). The mixture of H_2 and CO (*i.e.* city gas,

syngas) is used since a long time for heating and lighting houses and cities and for production of methanol, that is - together with H_2 itself - a suitable feed for fuel cells for supplying motorveicles, buses, motorcycles, aircraft, PCs and many public and home utilities.

Today, most of H_2 is used for production of NH_3 and then nitrates as explosives, polymers, pharmaceuticals and ammonium salts as fertilizers. Besides, a large part of H_2 is used for catalyzed hydrogenation of heavy unsaturated hydrocarbons for production of lubricants and syn-gasoline as well.

Several methods have been proposed for preparation of even larger amounts of hydricity by heating water in a gas-cooled high temperature nuclear reactor (*i.e.* Nuclear Hydrogen or NuH2), without any use of fossil fuels, which lead to the undesired co-production of CO_x, NO_x, SO_x, volatile organic compounds (VOCs) and residues containing *heavy* and *radioactive metals* (HMs), or even worse any use of electrolysis of water, which is thermodynamically very unfavorable [10,11,12].
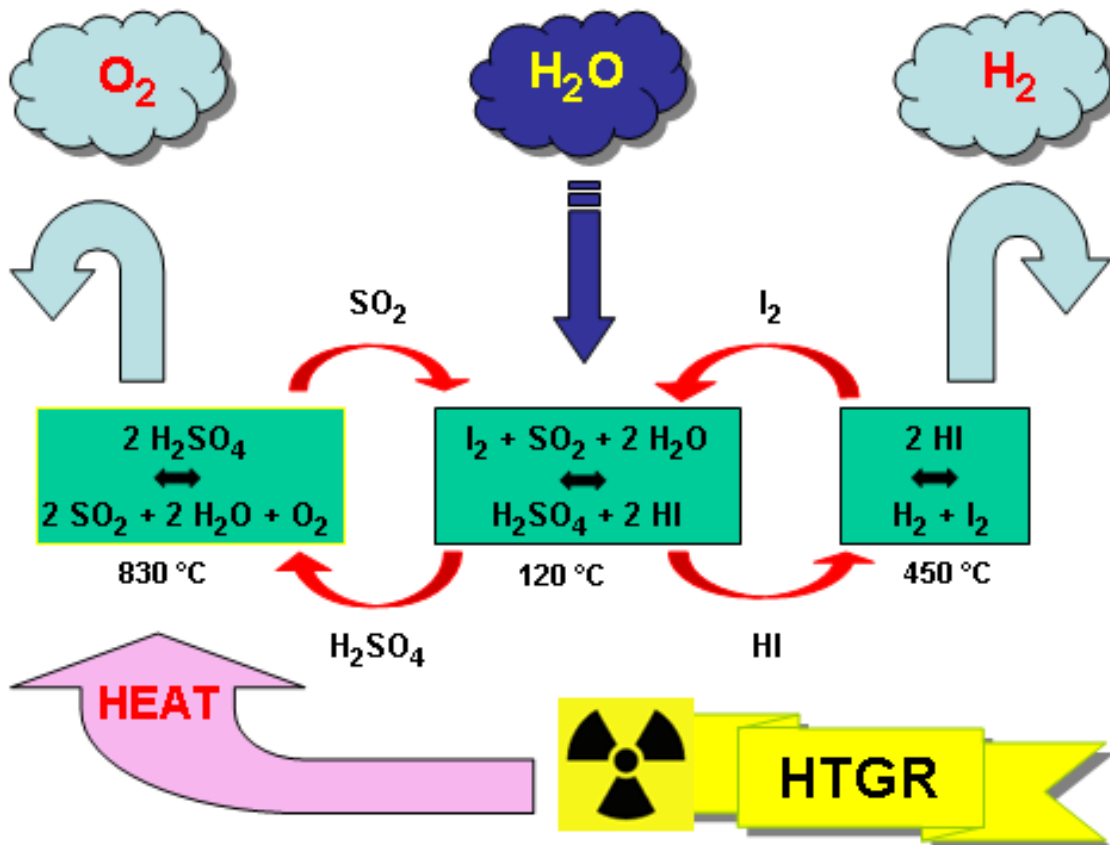

FIG. 1: Production of hydrogen (and oxygen) by thermochemical decomposition of water in high temperature nuclear reactor, by using the **sulphur-iodine (S-I) cycle.**

Amongst them, the most promising is the sulfur-iodine cycle (S-I cycle), which is a series of thermochemical processes based on Bunsen's reaction and used to produce H_2. The reactions that produce H_2 are as follows, Eqs. (1-3):

$$I_{2(v)} + SO_{2(g)} + 2\ H_2O_{(v)} \leftrightarrow 2\ HI_{(v)} + H_2SO_{4(l)} \qquad \text{(Bunsen's reaction, at 120°C)} \quad (1)$$
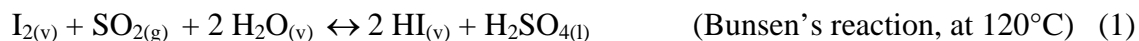
$$\Delta H_R{}^{\varnothing}(298.15\ K) = -41.08\ kJ < 0 \qquad \Delta G_R{}^{\varnothing}(298.15\ K) = +51.47\ kJ > 0) \qquad (1')$$

The HI is then separated as a vapor by distillation. Note that concentrated H_2SO_4 may react with HI, giving I_2, SO_2 and H_2O (backward reaction, *i.e.* ΔG is > 0), but removing the desired product will shift the equilibrium to the right.

$$2\ H_2SO_{4(v)} \leftrightarrow 2\ SO_{2(g)} + 2\ H_2O_{(v)} + O_{2(g)} \text{ (at 830-850 °C in vapor phase)} \qquad (2)$$
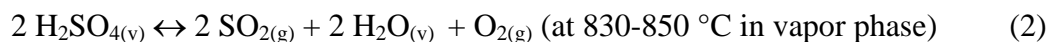
$$\Delta H_R{}^{\varnothing}(298.15\ K) = +629.7\ kJ > 0 \qquad \Delta G_R{}^{\varnothing}(298.15\ K) = +486.1\ kJ > 0 \qquad (2')$$

The H_2O, SO_2 and residual H_2SO_4 must be separated from the O_2 (that is a valuable by-product) by condensation. The decomposition reaction of Eq. (3) now produces H_2 and recycle the I_2:

$$2\ HI_{(v)} \leftrightarrow I_{2(v)} + H_{2(g)} \qquad \text{(at 400-450°C in gaseous phase)} \qquad (3)$$
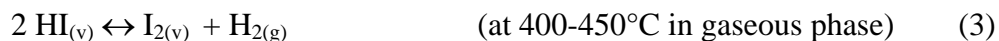
$$\Delta H_R{}^{\varnothing}(298.15\ K) = +9.48\ kJ > 0 \qquad \Delta G_R{}^{\varnothing}(298.15\ K) = +15.93\ kJ > 0 \qquad (3')$$

By double integration of Eqs. (4) and (5), it is possible calculating and optimizing the $\Delta G(T)$ and T from the knowledge of the behavior of tabulated $C_P{}^{\varnothing}(T)$ of different components, *i.e.* to calculate $\Delta H(T)$, by using for example the 5 parameters Eq. (6) [25].

$$\Delta H(T) = [\partial\ (\Delta C_P(T))\ /\partial T]_P \qquad (\textit{Kirchoff's Eq.}) \qquad (4)$$

$$[\partial\ (\Delta G(T)/T)/\partial T]_P = -\Delta H(T)/T^2 \qquad (\textit{Gibbs-Helmoltz's Eq.}) \qquad (5)$$

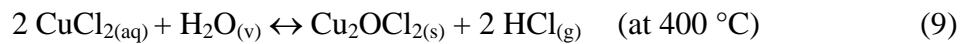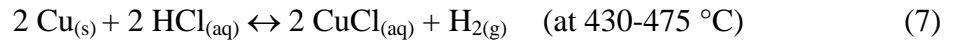$$C_P{}^{\varnothing} = \alpha + \beta\ T + \gamma\ T^2 + \delta\ T^3 + \varepsilon\ /\ T^2 \qquad (\textit{Shomate's Eq.}) \qquad (6)$$

Iodine and any accompanying H_2O or SO_2 are separated by condensation, while the H_2 product remains in the gas phase. Then H_2 can be either compressed, adsorbed as *metal hydride* or into *nanotubes*, or *liquefied* by iso-enthalpic Joule-Thompson free expansion, after pre-cooling with liquid N_2 at 78 K (- 195 °C) and than *stored* at 20 K (- 253 °C). The S and I compounds are recovered and reused, hence the consideration of the process as a catalytic cycle (FIG. 1). All reactions are *endoergonic* at room temperature.

This S-I process is a chemical heat engine; heat enters the cycle in high temperature endothermic chemical reactions (2) and (3), and heat exits the cycle in the low temperature exothermic reaction (1). The difference between the heat entering the cycle and the heat leaving the cycle exits the cycle in the form of the heat of combustion of the H_2 produced. In conclusion the net reaction is the decomposition of water and oxygen is a valuable by-product of the process.

3.1   Hybrid thermochemical-electrochemical cycles

Apart the S-I cycle developed by General Atomics and Argonne-NL in USA, there are several *hybrid termochemical-electrochemical* cycles to be cited for completeness, combined between a medium-high temperature termochemical step and an electrolytic step at low temperature. There are several variant of the Fe-Ca-Br adiabatic cycle (UT-3) at 1 033 K of ANL, USA and Japan, the process hybrid-sulfur (HyS) combined with electrolysis of Westinghouse-USA and Savannah-River-NL, USA, the reverse Deacon cycle (Mg-Cl) with electrolysis and the process Cu-Cl at low temperature (Cu-Cl) of ANL, USA and AECL, CA combined with electrolysis too (Eqs. 7-10) [26,27]. All these methods do not utilize fossil fuels and do not have undesired combustion products, apart water vapors, and the net reaction is always: $2\ H_2O \leftrightarrow 2\ H_2 + O_2$

$$2\ Cu_{(s)} + 2\ HCl_{(aq)} \leftrightarrow 2\ CuCl_{(aq)} + H_{2(g)} \quad \text{(at 430-475 °C)} \tag{7}$$

$$2\ Cu_2OCl_{2(aq)} \leftrightarrow 4\ CuCl_{(aq)} + O_{2(g)} \quad \text{(at 500-530 °C)} \tag{8}$$

$$2\ CuCl_{2(aq)} + H_2O_{(v)} \leftrightarrow Cu_2OCl_{2(s)} + 2\ HCl_{(g)} \quad \text{(at 400 °C)} \tag{9}$$

$$2\ CuCl_{(s)} \ \leftrightarrow\ CuCl_{2(aq)} + Cu_{(s)} \quad \text{(ambient-T \textit{electrolysis})} \tag{10}$$

3.2   Advantages and drawbacks in nuclear hydricity production

Finally, in the gas-cooled very high temperature reactor of *Gen IV* it would be possible to dissociate water by *direct pyrolysis* at temperatures higher than 1 000 °C up to 1 600 °C, either directly or on a metal catalyzer (Fe, Co, V) or a refractory metal oxide. Indeed the use of high temperature cycles have the further advantage of increasing the *efficiency* of the *Brayton thermodynamic cycle* usually adopted (*i.e.* two isobaric and two isoentropic transformations). There are three designs of these reactors suitable for NuH2 production: the **VHTR**, the **GFR** and the **LFR** [14].

Besides, it is evident the main advantages of the nuclear methods for hydricity production are based on the elimination of any fossil fuel feed and relative green-house effect by-products, together with high toxic residues like toxic and even radioactive

elements, coming from natural decay chains, which are concentrated in both *bottom-ash* and *fly-ash* fractions of thermoelectric-PPs.

Unfortunately, inside this optimistic scenario might be a really unexpected drawback to be considered: there are some researchers claiming that the physiological losses of hydrogen of about 10% (during production, storage and transportation), could increase the H_2 concentration in the troposphere and then be hazardous for the *ozone hole* in the stratosphere [28]. In case of extensive use of hydricity, this potential hazard would be even more relevant than the more known effect due to green-house emissions like CFC, BFC, methane and sulphurated dielectric gases like SF_6.

3.3   The Very High Temperature Reactor (VHTR) of *Gen IV* features

The VHTR is designed to be a high-efficiency system (FIG. 2), which can supply electricity and process heat to a broad spectrum of high-temperature and energy-intensive processes. The reference reactor is a 600 MW_{th} core connected to an intermediate heat exchanger to deliver process heat.
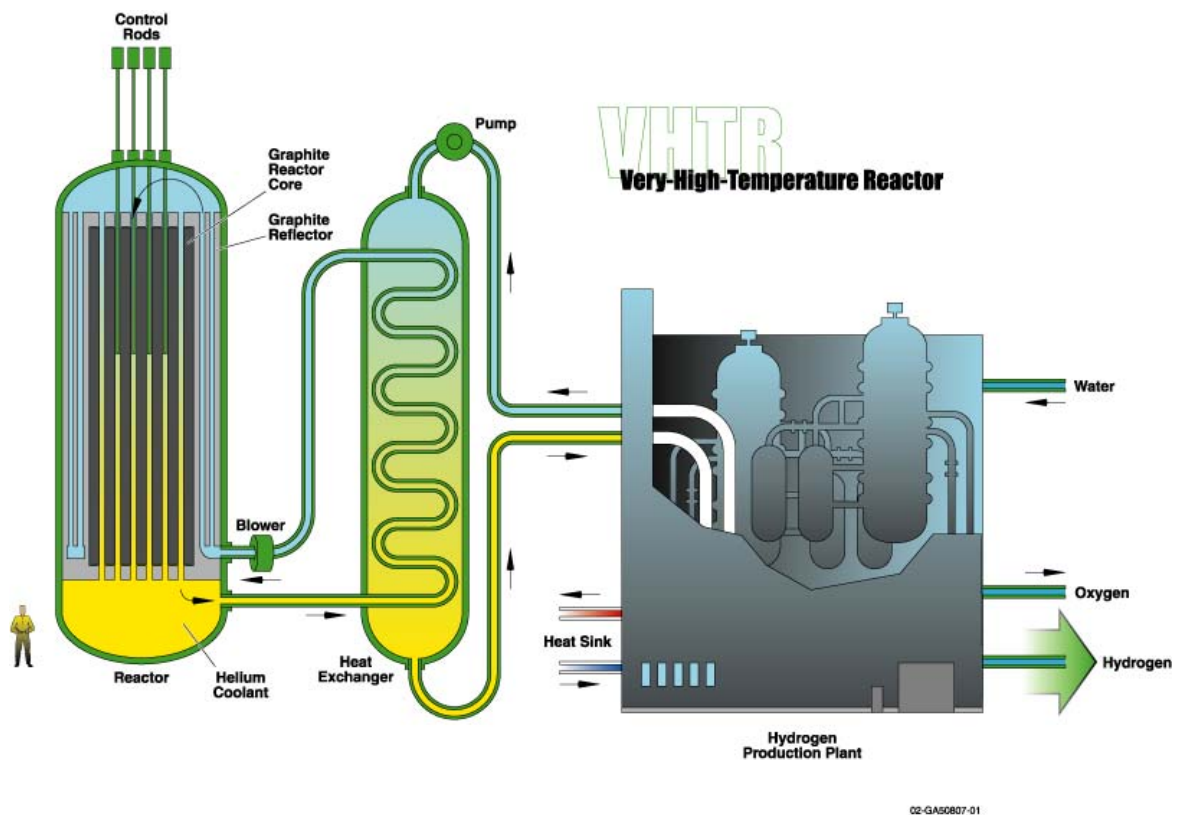
FIG. 2: Schematics of very high temperature gas cooled fast reactor (VHTR) of *Gen IV*, which can be adopted for NuH2 production.

The reactor core can be a *prismatic block core* or a *pebble-bed core* according to the fuel particles assembly. Fuel particles are coated with successive material layers, high temperature resistant, then formed either into fuel compacts embedded in graphite block for the prismatic block-type core reactor, or formed into graphite coated pebbles (**PBR**).

The reactor supplies heat with core outlet temperatures up to 1 000 °C, which enables such applications as hydrogen production or process heat for the petrochemical industry. As a nuclear heat application, hydrogen can be efficiently produced from only heat and water by using thermochemical iodine-sulfur (S-I) process, or high temperature thermochemical-electrolytic process or with additional natural gas by applying the steam reformer technology.

Thus, the VHTR offers a high-efficiency electricity production and a broad range of process heat applications, while retaining the desirable safety characteristics in normal as well as off-normal events. Solutions to adequate waste management will be developed. The basic technology for the VHTR has been well established in former High Temperature Gas Reactors plants, such as the US Fort Saint Vrain and Peach Bottom prototypes, and the German AVR and THTR prototypes. The technology is being advanced through near- or medium-term projects lead by several plant vendors and national laboratories, such as: PBMR, GT-HTR300C, ANTARES, NHDD, GT-MHR and NGNP in South Africa, Japan, France, Republic of Korea and the United States. Experimental reactors: HTTR (Japan, 30 MW_{th}) and HTR-10 (China, 10 MW_{th}) support the advanced concept development, and the cogeneration of electricity and nuclear heat application.

3.4 The Gas cooled Fast Reactor (GFR) of *Gen IV* features

The high outlet temperature of the helium coolant used in the GFR system (FIG. 3) makes it possible to deliver electricity, hydrogen, or process heat with high efficiency. The reference reactor is a 1.2 GWe helium-cooled system operating with an outlet temperature of 850 °C using a direct *Brayton cycle* gas turbine for high thermal efficiency.

Several fuel forms are candidates that hold the potential to operate at very high temperatures and to ensure an excellent retention of fission products: composite ceramic fuel, advanced fuel particles, or ceramic-clad elements of actinide compounds. Core configurations may be based on prismatic blocks, pin- or plate-based assemblies. The GFR reference has an integrated, on-site spent fuel treatment and refabrication plant. The GFR uses a direct-cycle helium turbine for electricity generation, or can optionally use its process heat for thermochemical production of hydrogen. Through the combination of a fast spectrum and full recycle of actinides, the GFR minimizes the production of long-lived radioactive waste. The GFR's fast spectrum also makes it possible to use available fissile and fertile materials (including depleted uranium) considerably more efficiently than thermal spectrum gas reactors with once-through fuel cycles.
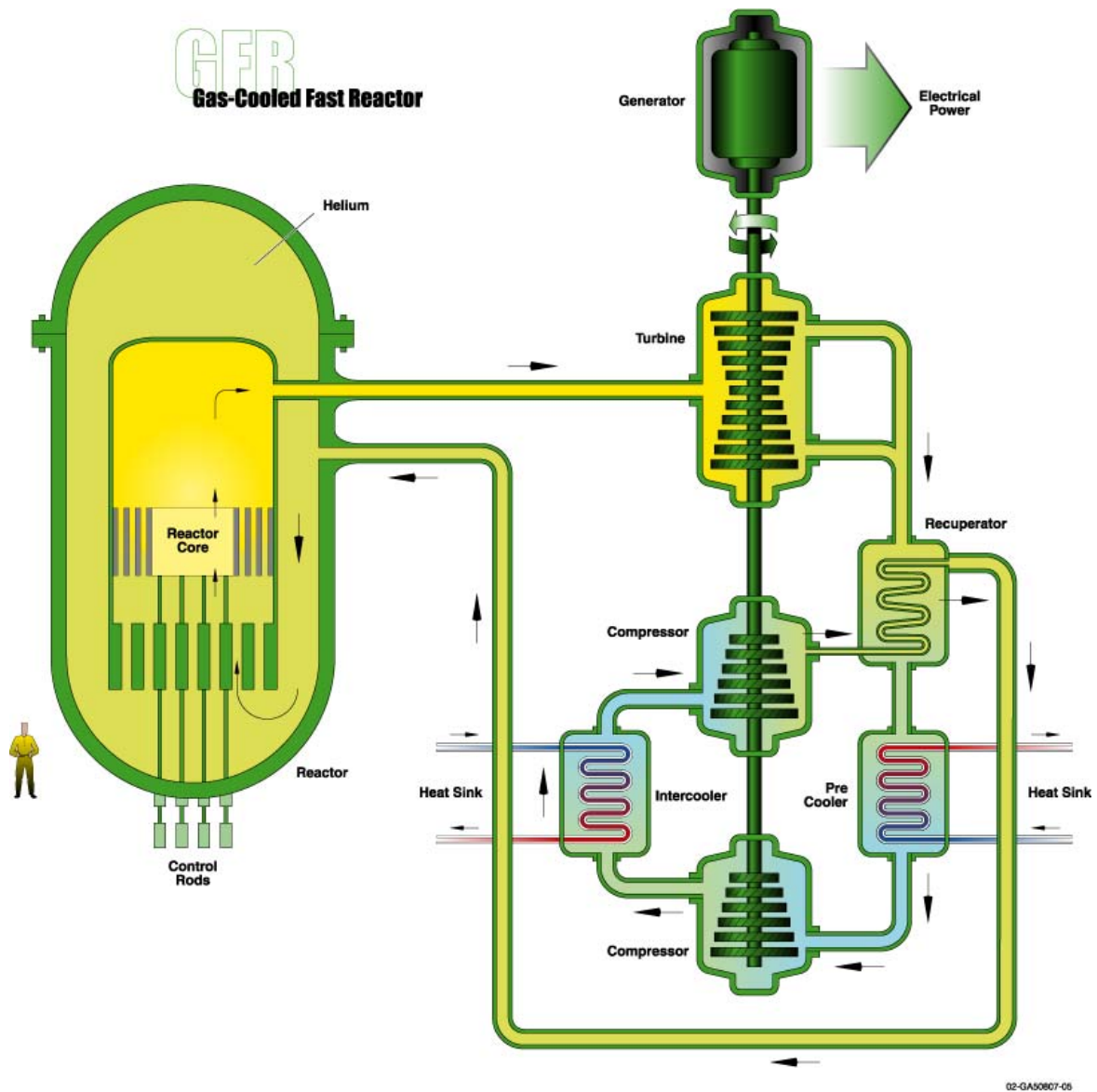
FIG. 3: Schematics of Gas cooled Fast Reactor (GFR)
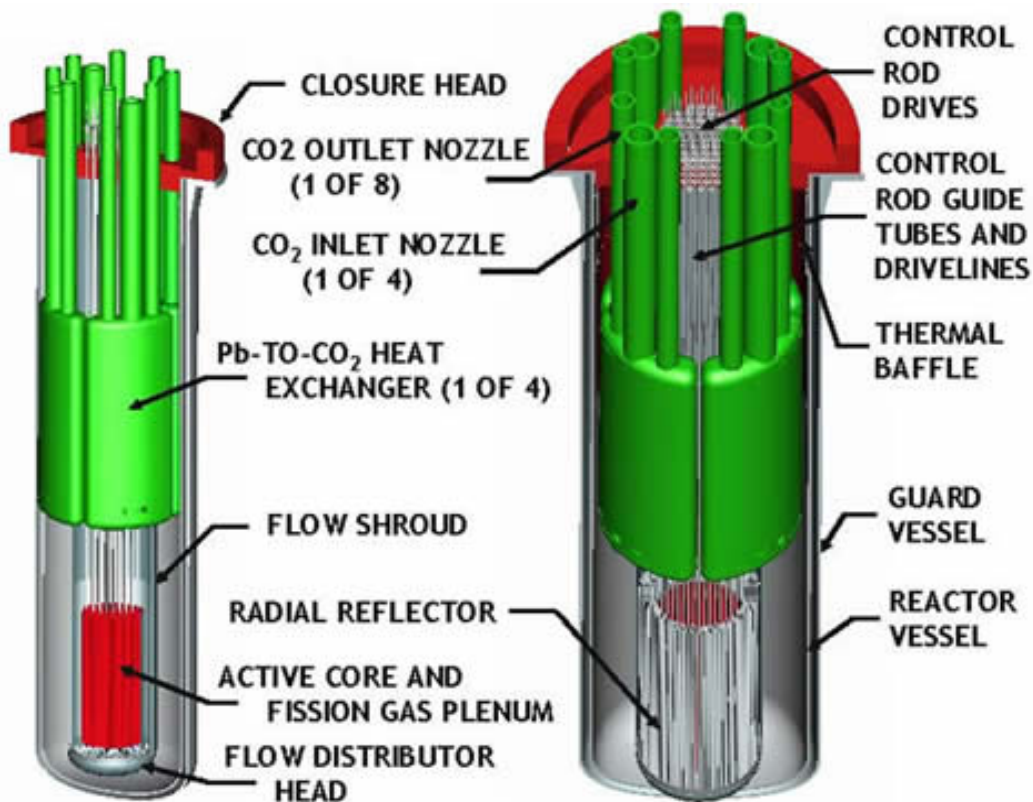of *Gen IV*, which can be adopted for NuH2 production.

3.3 The Lead cooled Fast Reactor (LFR) of *Gen IV* features

The LFR system has excellent materials management capabilities since it operates in the fast-neutron spectrum and uses a closed fuel cycle for efficient conversion of fertile uranium. It can also be used as a burner to consume actinides from spent LWR fuel and as a burner/breeder with thorium matrices. An important feature of the LFR is the enhanced safety that results from the choice of molten lead as a relatively inert coolant. In terms of sustainability, lead is abundant and hence available, even in case of deployment of a large

number of reactors. More importantly, as with other fast systems, fuel sustainability is greatly enhanced by the conversion capabilities of the LFR fuel cycle.

The LFR was primarily envisioned for missions in electricity and hydrogen production, and actinide management. Given its R&D needs in the areas of fuels, materials, and corrosion control, a two step process leading to industrial deployment of the LFR system has been envisioned: by 2025 for reactors operating with relatively low primary coolant temperature and low power density; and by 2035 for more advanced designs. The preliminary evaluation of the LFR concepts considered by the LFR Provisional System Steering Committee (PSSC) covers their performance in the areas of sustainability, economics, safety and reliability and proliferation resistance and physical protection.

The LFR concepts that are currently being designed are two pool-type reactors: the Small Secure Transportable Autonomous Reactor (**SSTAR**) of FIG. 4, developed in the USA and the European Lead-cooled System (**ELSY**) of FIG. 5, developed by the EC.

FIG. 4: Schematics of the Small Secure Transportable Autonomous Reactor (SSTAR), a prototype of lead cooled fast nuclear Reactor (LFR) of *Gen IV*, which can be adopted for NuH2 production.
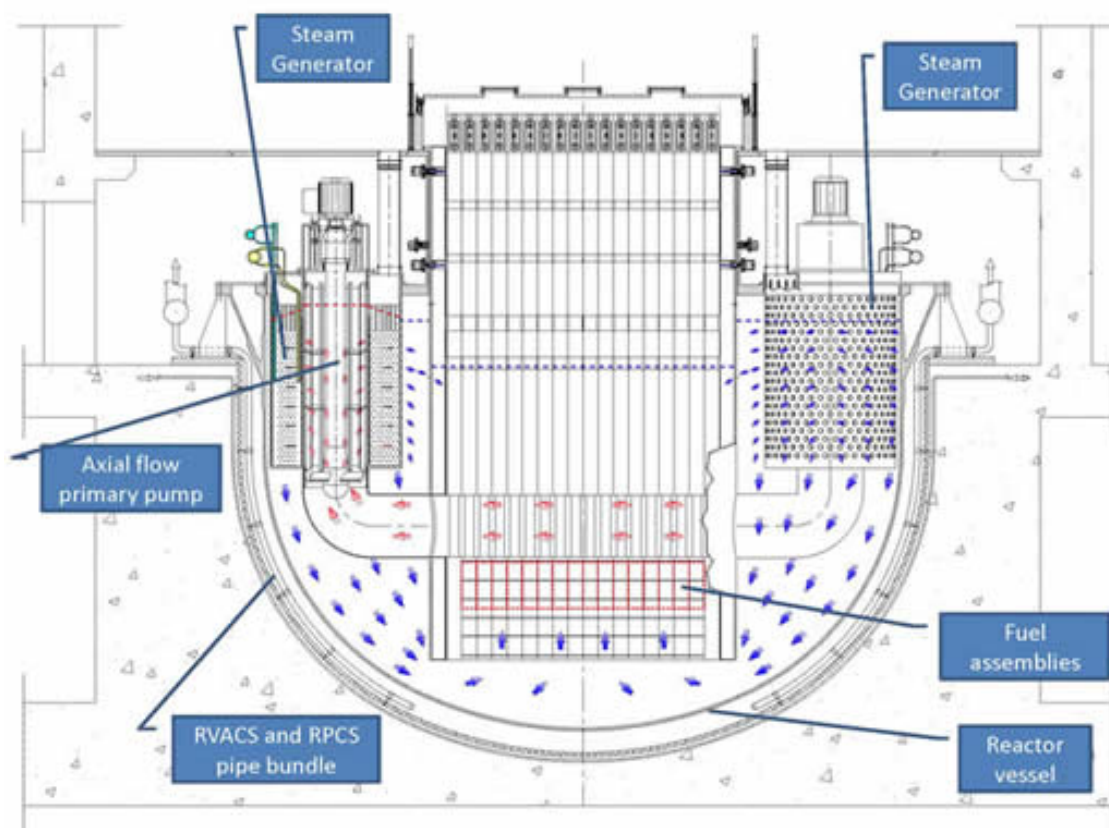
FIG. 5: Schematics of the European Lead-cooled System (ELSY),
a prototype of lead cooled fast nuclear Reactor (LFR) of *Gen IV*,
which can be adopted for NuH2 production.

4   CONCLUSIONS

There are at least three design of nuclear reactors of *Generation IV* (**VHTR, GCR** and **LFR**) that would operate at either very high or high temperature, with the capability of producing nuclear hydricity, NuH2, with high thermodynamic efficiency. Some of these reactors are even modular and transportable along river or sea [29].

Most of these reactors have the capability to transmute any kind of fuel: U and Pu radionuclides, minor actinoids (MAs), long-lived fission and activation products (FPs and CPs), with minimum production of rad-waste to be stored in repository [30,31]. A modern two phase recycling and transmutation fuel cycle is shown schematically in FIG. 6. Today (2010), according to OECD [30] the 440 NPPs operating all around the world in 36 Countries do produce annually a negligible volumic amount of 200 000 m^3 of medium and intermediate level wastes (MILWs) and 10 000 m^3 of high level wastes (HLWs), corresponding to the volume of a basket field. The *Gen IV* will reduce further these quantities by transmutation down to 0.01% of the total [31].

If 1 000 NPPs will operate in the next decades, they will produce 33-35% of total world electricity demand, or they will be able to share about 50% of capacity for producing electricity and 50% hydricity.
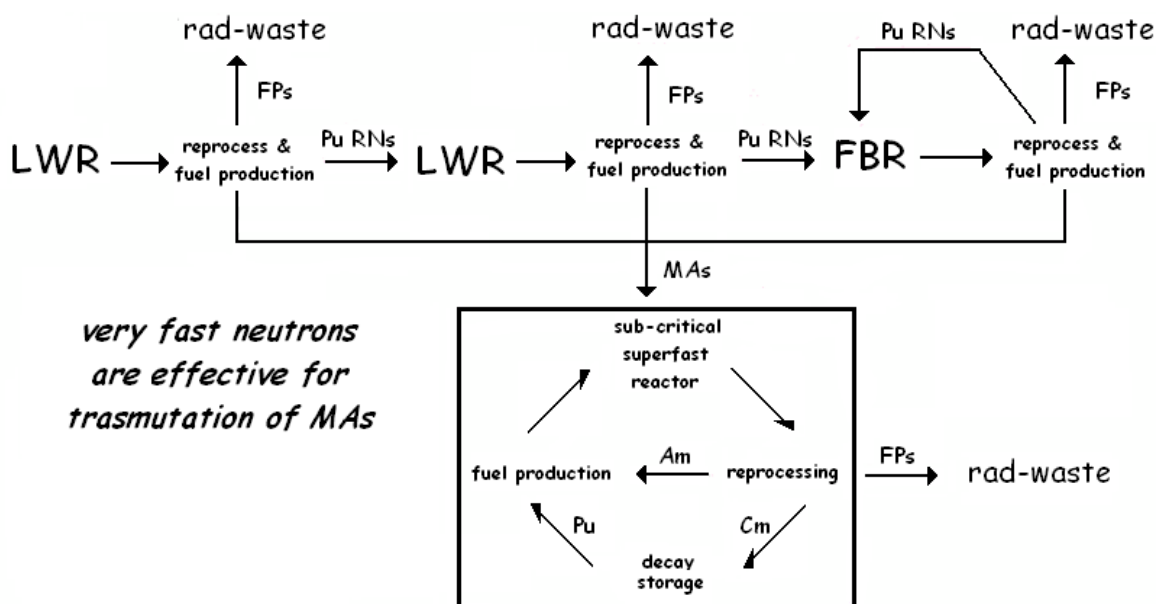
FIG. 6: a fuel recycle of a light water reactor, LWR (in two phases) in which the minor actinoids (MAs) are treated separately from U and Pu RNs. More phases can be added if the spent fuel does not contain high neutron absorbing RNs, whose concentration increases with burnout.

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