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Hydrogen economy for a sustainable development: state-of-the-art and technological perspectives

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Abstract

Sustainable energy is becoming of increasing concern world-wide. The rapid growth of global climate changes along with the fear of energy supply shortage is creating a large consensus about the potential benefits of a hydrogen economy coming from renewable energy sources. The interesting perspectives are over-shadowed by uncertainties about the development of key technologies, such as renewable energy sources, advanced production processes, fuel cells, metal hydrides, nanostructures, standards and codes, and so on. The availability of critical technologies can create a base for the start of the hydrogen economy, as a fuel and energy carrier alternative to the current fossil resources. This paper will explore the rationale for such a revolution in the energy sector, will describe the state-of-the-art of major related technologies (fuel cell, storage systems, fuel cell vehicles) and current niche applications, and will sketch scientific and technological challenges and recommendations for research and development (R&D) initiatives to accelerate the pace for the widespread introduction of a hydrogen economy. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Hydrogen; Fuel; Energy carrier; Fuel cells; Storage; Fuel cell vehicles

1. Introduction

Emissions of carbon dioxide (CO_2), the main greenhouse gas (GHG) from human activities, are the subject of a world-wide debate about energy sustainability and the stability of global climate.

Carbon dioxide is essential for life on our planet. Animals and plants, volcanoes, oceans and forests control, in a delicate system of complex equilibrium, the CO_2 concentration in the atmosphere: too low or too high values can lead to global climate changes and, then, to a cooling or warming of the earth.

Before the industrial era, the natural concentration of CO_2 was estimated to be about 280 ppm; since then there has been an increase of about 30%, going up to 370 ppm. This increase, and consequent temperature rise, has been mostly ascribed to the utilisation of fossil fuels [1,2].

Even though it is very difficult to assess the global climate changes related to the increase of the GHG concentration, it is agreed that a "*sustainable*" level must absolutely remain below 550 ppm, which is considered by scientists as the maximum acceptable value in order to minimise the effects of global climate changes (Fig. 1). Stabilisation of CO_2 concentration at this value requires some 50% reduction of the emissions in 2050.

Unfortunately, forecasts for energy demands are not so encouraging, due to both the population growth rate and energy predictions of future consumption (see Fig. 2) [2]. In fact, all the socio-economic scenarios shown in Fig. 2 — from an accelerated technologic development and economic growth (line A) to medium development (line B), to an improbable scenario of strong environmental constraints and de-materialised societies all around the world (line C) — agree on a significant growth rate of energy consumption and world-wide population (in billion).

The Intergovernmental Panel on Climate Change (IPCC), using the scenarios shown in Fig. 2, predicted that, without a strong policy against the greenhouse effect, for increased energy demand, mostly from fossil fuels, CO_2 emissions will treble in this century, going from 7.1 Gt of carbon (GtC) in 1990 up to more than 20 GtC in 2100 (Fig. 3). The consequent concentration of CO_2 in the atmosphere will reach about 700 ppm [3].

From this perspective, the Kyoto Protocol of December 1997 represents the first international common action towards GHG emission controls, but, even if difficult to achieve, it is just the initial step on a long road. Consequently, public awareness and concern over mitigation of GHG emissions and climate change control are significantly growing with increased expectations of concerted decisions [4,5].

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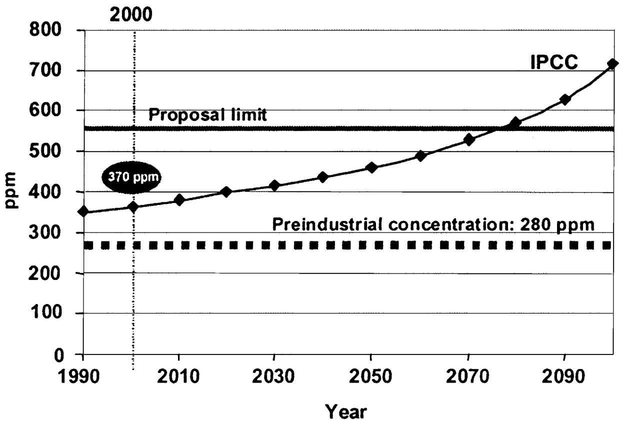


Fig. 1. Projections of atmospheric concentration of CO_2.

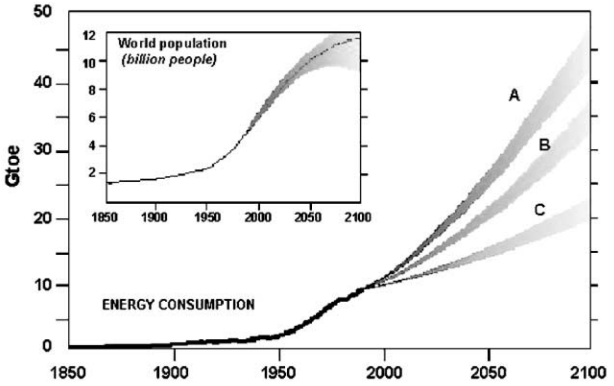


Fig. 2. Scenarios for energy demand and population growth.

1.1. Options for reduction of GHG emissions

Reduction of GHG emissions requires substantial modification in conversion and utilisation of different energy sources, which can be only achieved by adopting one or more of the following solutions:

- efficiency improvement, with reduction of fossil fuel consumption;
- use of low-carbon or carbon-free energy sources (natural gas, renewable, nuclear);
- separation and sequestration of the CO_2 produced from fossil fuels.

1.1.1. Improvement of total efficiency in energy cycles

Increased efficiency when using fossil fuels is the main option in the short-term, even though the amount of CO_2 to be eliminated is not very high. Quantitatively high potentialities exist in the following areas.

- Civil applications (residential and commercial).
- Generation of electric power, for instance by using incentives for co-generation. Large improvements in efficiency are possible, considering the present average efficiency of 38% (Italian generation mix) is compared with 58–60%, currently achieved with new combined cycle plants.
- Transportation sector, by means of more efficient mobility solutions and, in the medium-term, by the reduction of fuel consumption obtained with technological innovation (direct injection engines, electronic control of combustion, alternative cleaner fuels, hybrid electric configurations, lightweight materials) and limitation of vehicle performance characteristics.

1.1.2. Low carbon or carbon-free fuels

A shift towards fuels with a lower content of carbon, such as natural gas, is necessary to meet the Kyoto requirements. US Energy Information Agency (EIA) and some European studies foresee that gas demand will double by 2020, as a "natural" market evolution. Of course, an international

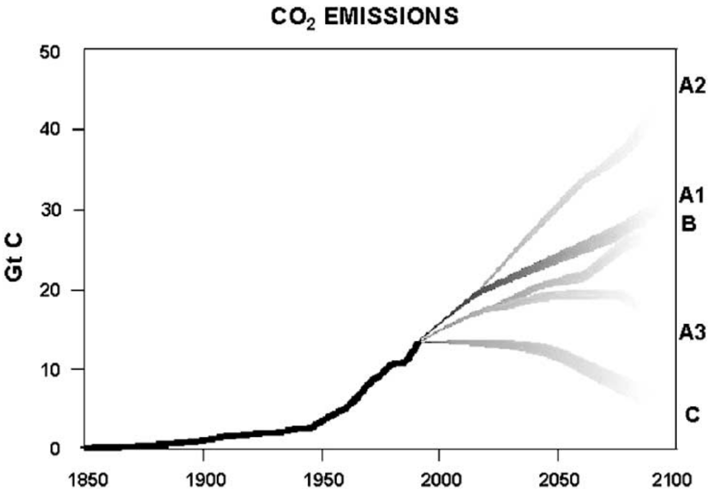


Fig. 3. Projections of CO_2 emissions (the Cases A, B and C in Fig. 2 are further divided in to six scenarios of energy systems: three Case A scenarios (A1, large availability of petroleum and gas; A2, return to coal; and A3, non-fossil future), a single Case B scenario (middle course, intermediate economic growth and technological improvements), and a single Case C scenario (ecologically driven, with renewable energy sources and new nuclear power).

agreement for GHG emission reduction will probably cause a steeper increase in the demand for cleaner fuels and, particularly, for natural gas [6,7].

However, in the long-term, the increase in natural gas utilisation will show limited results, due to:

- the unavoidable CO_2 production, even if less than with other fuels (14.6 kg C/G J with respect to 24.1 kg C/G J for coal);
- the logistics of transportation from production areas to the market, that will require new high pressure or liquid natural gas (LNG) technologies.

The other important option is to adopt renewable energy sources, whose role will become more and more important in future energy scenarios. Currently, hydropower, almost to its maximum potential, has a good share of electricity production, as well as "traditional" biomass. Interest and initiatives in developing other kinds of renewable energy sources (geothermal, wind, photovoltaic, thermal solar and biomass from ad hoc cultivation) are growing, but environmental problems exist and the costs are still high. Consequently, their contribution to future energy scenarios is quite uncertain, even if significant capacity growth for electricity generation is targeted in the US (an increase of 10% by 2015) and in European Union (from 6 to 12% of total production by 2008–2012) policies. Different scenarios, developed worldwide, reflect this uncertainty, forecasting a total contribution ranging between 20 and 50% by 2020 [8].

1.1.3. Carbon sequestration

Efficiency improvement, low carbon fuels and renewable energy sources will not be sufficient to stabilise atmospheric CO_2 concentration, since fossil fuels will continue to be the largest part of the energy demand (from 50 to 70% in 2050 and 18 to 50% in 2100, in IIASA–WEC scenarios) [2].

So, it is very important to develop another technological option, *carbon sequestration*, both indirect (absorption by ecosystem) and direct (CO_2 capture in fossil fuel cycles) [9].

1.1.3.1. Improvement of absorption capacity by the ecosystem. Forestation and a proper use of the land can lead to a good reduction of the GHG concentration, by increasing CO_2 absorption capacity of plants and the land itself.

According to IPCC studies, a global program could absorb, by 2050, 60–87 GtC, equal to 12–15% of total emissions from fossil fuels in the same period.

1.1.3.2. CO_2 capture and sequestration ("decarbonation" of fossil fuels). CO_2 capture and sequestration allow the decoupling of fossil fuels from CO_2 emissions, making them comparable, from an environment point of view, with renewable energy sources.

There are different options for CO_2 separation (pre- and post-combustion), but only the "pre-combustion" one can be adopted in sectors different from power generation, like transportation and residential.

1.2. Why hydrogen?

Hydrogen is the lightest, the simplest, and one of the most abundant elements in nature. It always comes combined with other elements and has a variety of good properties.

- Both production and utilisation of hydrogen can be emission-free.
- It can be obtained from a variety of feedstocks (fossil, renewable energy, nuclear).

These characteristics make hydrogen an ideal candidate for a future sustainable energy system using renewable energy, as primary source, and hydrogen and electricity as fuels and energy carriers. In particular, hydrogen:

- can be produced from fossil fuels by conversion, with CO_2 separation. This one can be considered the cleanest way to continue using those fuels, which will continue to have an important role in our societies;
- can be produced from other sources (renewable, nuclear) without CO_2 emissions;
- can be utilised in different applications (transportation, electricity production, etc.), without producing any pollutant but water steam.

Hydrogen not only can be produced with "zero emissions" from fossil fuels, but also can be obtained from several sources, in particular, by renewable energy and biomass. Production from fossil fuels could be considered as a "technological bridge" towards new processes like photoelectrochemical, biological and "new" nuclear, expected for the second-half of the century. Actually, the development, in the next few decades, of technologies for distribution and utilisation of hydrogen, will be the basis for the introduction of those CO_2-free production technologies.

Of course, the introduction of hydrogen as a fuel and energy carrier presents, besides unquestionable advantages, several problems in developing the required technologies (Fig. 4) for the following:

- production,
- transportation,
- storage,
- utilisation.

This paper will survey the present state-of-the-art of hydrogen technologies [10,11], will explore the scientific and technological challenges and limitations, and will sketch the short and medium to long-term research and development (R&D) needs for a 21st century vision for a future hydrogen-based economy.

2. Status of the art

2.1. Production

Hydrogen can be obtained from different sources. It is possible to produce it from water, both by conventional

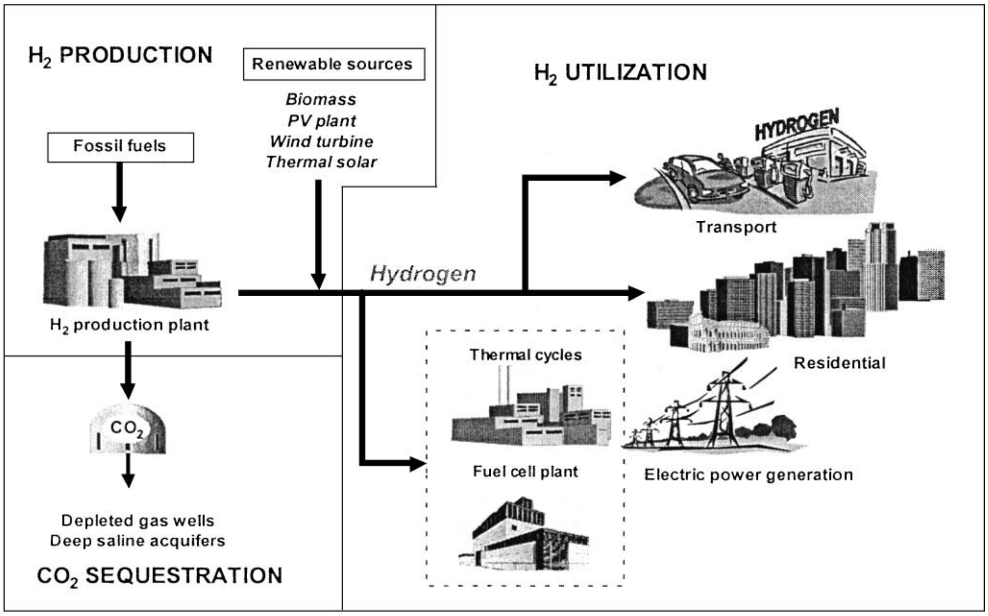


Fig. 4. Technological pathway for hydrogen economy development.

electrolysis and advanced high temperature processes; nuclear or solar energy can be used as heat source for H_2 producing processes, biomass and coal can be gasified to obtain hydrogen, as well as fossil fuels (Fig. 5).

In the long-term, hydrogen will be produced from renewable energy sources, once related technologies are completely developed to meet the required energy demand, while in the short-term, the only viable option is the production of hydrogen from fossil fuels. Currently, most hydrogen produced world-wide (about 500 billion N m^3 per year) is derived from fossil fuels, mainly natural gas. Production technologies (steam reforming, partial oxidation, gasification) are already commercial, but they are capable of further improvement from both energetic and environmental points of view.

The main problem to solve is the separation and sequestration of the CO_2 produced during H_2 production, by storing it in safe locations. Different solutions are under evaluation in different countries (oceans, depleted natural gas or oil wells, saline deep aquifers, rock caverns, etc.), including also processes (like natural gas pyrolysis) which produce no CO_2 at all. The development of technologies for the capture and sequestration of CO_2 must be part of large R&D programs, involving mainly separation (membranes, absorption and adsorption processes, cryogenic processes, etc.) and transportation technologies.

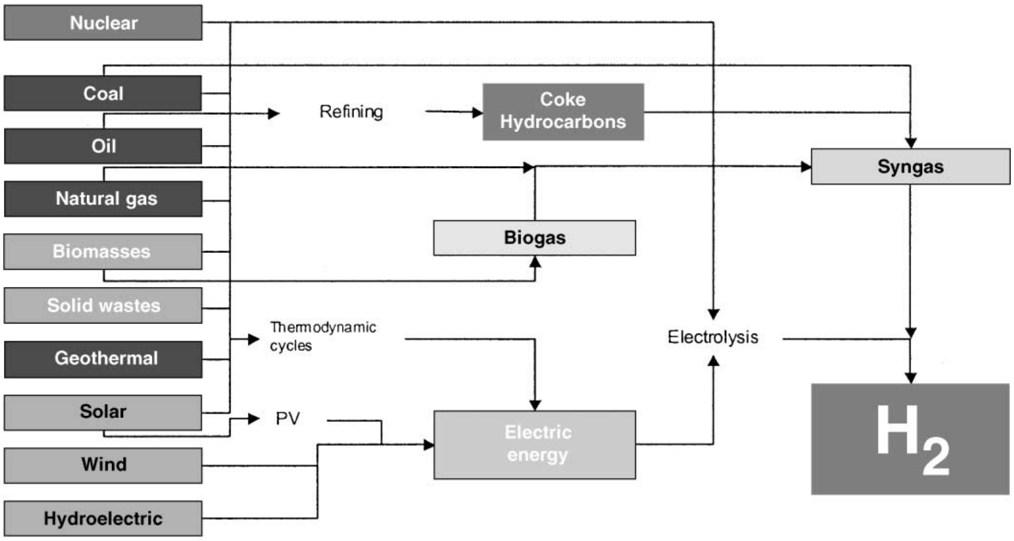


Fig. 5. Different H_2 production techniques.

Hydrogen production processes from biomass (gasification of solid biomass, fermentation of liquid organic wastes, photosynthesis-based processes) are under development, but still require a strong effort in terms of R&D and demonstration activities. On the other hand, hydrogen production by water electrolysis is an established, though expensive process. Hydrogen production costs could become acceptable if electricity were to become very cheap or if new high efficiency processes were to be developed (e.g. high temperature electrolysis).

Actually, a bundle of new technologies is under development for example, photoelectrolysis, in which photoelectrochemical cells produce hydrogen directly from water without producing electric energy as a separate step, or the utilisation of algae or bacteria for water splitting, or the use of bacteria themselves to fix CO_2 in the atmosphere.

2.2. Transport, distribution and storage

A widespread use of hydrogen as a fuel and an energy carrier will imply its large availability for a variety of applications close to the points of use. Adequate infrastructures will then be requested to transport and distribute the hydrogen, and also to store it to match time and space in the displacements between availability of the primary feedstocks, the production and end use. The hydrogen currently can be transported and distributed in liquid and gaseous form in special tanks. The gas can also be transmitted at high pressure in dedicated pipelines.

In the future, the transport and distribution of hydrogen can be envisaged as part of a system of "energy carrier" networks, including electricity and natural gas. The gaseous hydrogen transport and distribution system might look like current natural gas pipelines with significant technological innovations: new materials for the ducts able to avoid hydrogen leakage, and different working pressures and flows to overcome the reduced energy content of gaseous hydrogen. Consequently, the development of hydrogen-based infrastructures has technological and economic constraints that must be solved [12,13].

One of the major obstacles to the diffusion of hydrogen as an energy carrier is the lack of safe, efficient and cost-effective storage systems, suitable for the various stationary and mobile applications. The choice of hydrogen storage system is always a compromise among various criteria, which at the present stage of R&D of related technologies, must still be clearly identified. The criteria can be based on external (transport and distribution infrastructure, connection between the network and the final users, handling safety) and internal (size, charge–discharge kinetics, efficiency, cycle life, safety in use, life-cycle environmental impact of storage materials) factors and, finally, on the economics, which includes not only the foreseen costs of the storage systems, but also the needs for R&D. For example, the use in vehicles, one of the most challenging and intriguing future applications, has the mandatory need of high energy storage systems, which must cope with the low energy density and boiling point of hydrogen. In addition, hydrogen storage selection has a sure impact on the refuelling time, cost, infrastructure and energy efficiency. Table 1 compares the energy content of various fuels in various states with respect to gasoline.

Various methods are currently proposed and being investigated for the direct (without any reformation process, the more appealing for the efficiency and emission standpoint), or indirect (through a transport medium or intermediate fuel carriers) storage of hydrogen. The storage of hydrogen then owns many scientific and technological challenges, which are being faced in various ways: hydrogen can be stored physically by changing its state conditions (temperature, pressure, phase), and chemically or physico-chemically in various solid and liquid compounds (metal hydrides, carbon nanostructures, alanates, borohydrides, methane, methanol, light hydrocarbons). The storage in solid media are safer and, potentially, more efficient than compression or liquefaction, due to leak-proof status, higher charging efficiency and lower self-discharge. The

Table 1
Energy density for various fuels relative to gasoline (adapted from [11])

Fuel	State at ambient temperature and pressure	Specific energy (per unit mass) relative to gasoline	Energy density in liquid state (per unit volume) relative to gasoline
Hydrogen	Gas	2.70	0.27–0.33[a]
Methane (natural gas)	Gas	1.13 (0.97)[b]	0.68 (0.57)[b]
Ethane	Gas	1.07	0.76
Propane	Gas (liquid)[c]	1.05	0.73
Gasoline	Liquid	1[d]	1[d]
Ethanol	Liquid	0.60	0.68
Methanol	Liquid	0.45	0.51

[a] The higher number refers to hydrogen density at the triple point.

[b] The number in brackets refers to a typical composition of natural gas.

[c] Normally stored as a liquid at moderate pressure.

[d] The reference values for gasoline are 44.4 MJ/kg and 31.1 MJ/l.

most common methods currently investigated include the following.

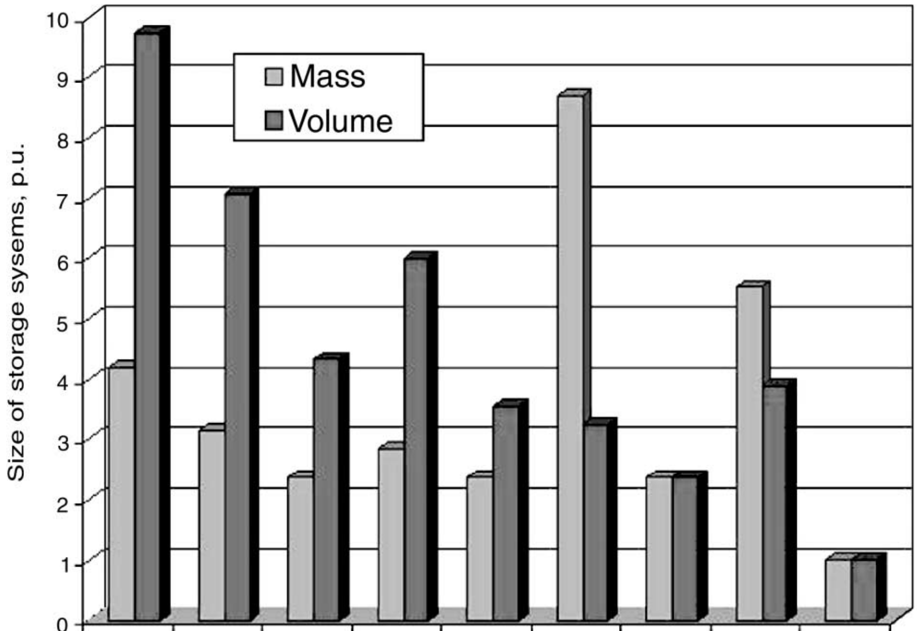
1. Storage in gaseous form.
   1.1. High pressure gas tanks in different materials
2. Storage in liquid forms or media.
   2.1. Liquefied hydrogen in cryogenic "dewar"
   2.2. Liquid compounds plus reformer
3. Storage in "solid" form, more properly in solid media in chemical, absorbed or adsorbed form.
   3.1. Metal hydrides
   3.2. Carbon structures
      3.2.1. Nanotubes
      3.2.2. Fullerenes
   3.3. Glass microspheres

The large variety of methods depends on the already mentioned need to compromise different technical, economical and safety criteria. For example, despite the high energy content in a unit mass, at ambient temperature and pressure hydrogen has very poor energy density, which is impractical for real applications. As a consequence, the selection of hydrogen storage methods should take into account such an intrinsic limitation. Fig. 6 demonstrates, with a real calculation, one of the most significant hurdles to be overcome to develop hydrogen storage systems for vehicular applications: storing enough hydrogen for meeting performance requirements. The comparison of the dimensions (mass and volume) of various hydrogen storage systems (in pure units, relative to the gasoline tank properties: 71 kg and 72 l) considers a typical range (560 km) of a conventional mid-size car and includes the dimensions of the storage media.

2.2.1. Pressurised hydrogen gas

The compression of hydrogen in gaseous form at very high pressure is the most commonly used storage method. A pressure of about 200–250 bar (20.7–24.8 MPa) is today available in commercial products, reaching an average hydrogen content of about 1–2 wt.%. Various containment systems have been proposed with different storage pressure, whose choice depends on the specific applications [15,16].

- Large cylinders, spherical containers and long tubes above-ground are normally used in natural gas systems. These systems are normally convenient only for large scale storage (up to 15,000 Nm^3).
- Underground reservoirs (salt or mined caverns, aquifers, depleted gas wells) are used for storing large amount of gas. Since 1971 in Germany town gas has been stored in caverns and, in France, Gaz de France is storing hydrogen-rich refinery by-products in an aquifer. These systems are, of course, applicable for very large storage needs.
- For mobile or small-scale applications, problems arise because of the very low volumetric energy density of conventional gas cylinders. New storage pressure tanks are fabricated that can store hydrogen at extremely high pressures (up to about 70 MPa). New materials and



	H2-gaseous @ 250 bar	H2-gaseous @ 345 bar	H2-gaseous @ 690 bar	H2-liquid (<300mm dia)	H2-liquid (>540mm dia)	Metal hydride AB5-type	Metal hydride Mg-type	Methanol reformer FCEV	ICE
▨ Mass	4,18	3,15	2,37	2,85	2,37	8,68	2,37	5,52	1,00
■ Volume	9,72	7,07	4,32	5,99	3,54	3,24	2,36	3,89	1,00

Fig. 6. Dimensions, relative to a gasoline tank, of various hydrogen storage systems (including containers or medium dimensions), adapted from [14].

configurations have necessarily been developed for realising advanced lightweight high-pressure containers. Composite materials have been used to design and fabricate commercial or prototype vessels with a hydrogen content (wt.%) that can be four to five times higher than that of conventional tanks. The increase of the energy density, being related to a significant increase of the storage pressure, imposes more severe safety and standards requirements. The introduction of special burst fuses and interruption valves significantly improve safety aspects. Compression infrastructure at high pressure and charging efficiency are a cause of further safety and technical issues. Nevertheless, most car companies are using lightweight tanks for storing hydrogen in development of fuel cell vehicles (buses, vans and passenger cars). These systems are also part of the DOE and European Union programs with technical goals of about 4 kWh/kg (12 wt.% of hydrogen) and 0.7 kWh/l [17].

2.2.2. *Liquefied hydrogen*

Hydrogen can be stored in liquid form for both stationary and onboard vehicle applications. The process of hydrogen liquefaction encompasses compression, cooling and expansion phases to modify the physical state of hydrogen from gas to liquid. The simplest liquefaction process is the Linde cycle or Joule–Thompson expansion cycle. In this process, the gas is compressed at ambient pressure, then cooled in a heat exchanger, before passing through a throttle valve where it undergoes an iso-enthalpic Joule–Thompson expansion, producing some liquid. This liquid is removed and the cool gas is returned to the compressor via the heat exchanger. A "dewar"-type container must then be used to maintain the liquid hydrogen at the final temperature of 20 K. Historically, this method has received attention, particularly in Germany due to the well established aerospace industry, where the lightness and compactness of the storage system was one advantage and the cooling of the fuel mixture with cryogenic liquid hydrogen before entering the combustion chamber of thermal engine was another. Super-insulated cryogenic containers have been developed to minimise hydrogen losses from liquid boil-off. In fact, working as a cryogenic liquid at its point, some liquid hydrogen is bound to evaporate at any heat transfer. Fuel losses due to boil-off can reach up to 3% per day. Major concerns of the liquid hydrogen storage method are the amount of energy required (up to 30% of the overall energy content in the storage tank) in the liquefaction process and the complexity of the distribution and fuelling infrastructure.

2.2.3. *Metal hydrides*

Hydrogen can be chemically bonded to metal or alloys to form metal hydrides. The adsorption of hydrogen in the lattice of more than 80 metals (or alloys) can be achieved at or below atmospheric pressure (the process is exothermic and needs cooling), then the hydrogen is released at significantly higher pressures when heated: the higher the temperature, the higher the pressure. There is a wide operating range of temperatures and pressures for hydrides depending on the alloy. Each alloy has different performance characteristics, such as hydrogen content, desorption rate (and charge–discharge kinetics), cycle life and heat of reaction. The method presents potentially high advantages: the volumetric energy density is three to four times higher than that of a compressed tank but hydrogen can be stored indefinitely provided that no heat is given to the metal hydride, with a much higher safety in real use (no release of hydrogen is allowed in case of accident). Hydrides store only about 2–7 wt.% hydrogen. The heats of reaction for hydrides can range from 9300 to >23,250 kJ/kg of hydrogen, and operating pressures can reach more than 10 MPa. Some hydride release temperatures can also be quite high (>500°C). With this wide range in pressures and temperatures, the construction of the storage unit becomes a challenge. The vessel containing the hydride must be pressurised and contain sufficient heat exchange area to allow rapid heat transfer for charging and discharging the hydride. The metal hydride alloy must also be structurally and thermally stable to withstand numerous charge–discharge cycles. Some hydrides can also be poisoned by carbon dioxide, sulphur compounds, or water.

Metal hydrides can be roughly divided into three categories: (1) $LaNi_5$-based alloys; (2) Mg-based alloys and (3) Ti-based alloys. The $LaNi_5$ and titanium alloys adsorb and desorb hydrogen (an amount ranging between 1.4 and 3 wt.%) at low temperature (room temperature or slightly above), whereas magnesium alloys hydride and dehydrate at high temperature (between 230 and 400°C) with a greater hydrogen content (up to about 7 wt.% for Mg-alloys doped with Ni).

Table 2 summarises the major performance of most investigated metal hydride materials or systems [16,18].

Table 2
Comparison of various metal hydride/systems performance characteristics [18]

Material/systems	Equilibrium temperature ($1 < p < 5$ bar) (°C)	Desorption rate	Amount of adsorbed H_2 (wt.% H_2)	Specific energy (kWh/kg)	Energy density (kWh/l)
MgH_2	290	Unacceptable	7.9	2.6	2.2
$MgNiH_2$	290	Acceptable	4.8	1.6	1
FeTi	0–100	Acceptable	1.2	0.4	1.10
Mercedes (mid-1980s)	0–300	Acceptable	1	0.330	1.10
Toyota RAV4FCEV (1997)	100	Acceptable	2	0.660	2

In conclusion, the metal hydrides are considered a safe and high volumetric energy density technology, but still bulky (for the limited specific energy) and expensive.

2.2.4. Carbon nanostructures

The discovery at the beginning of the last decade of new forms of carbon aggregation with basic particle size in the *nanoscale* range has opened up a variety of scientific and technological speculations and investigations about their potential applications. Fullerenes, *carbon onions*, carbon nanotubes and nanofibres, along with activated carbon, have been developed and proposed also for the storage of hydrogen, through the adsorption at low pressure of compressed hydrogen. This method has been considered as the substantial breakthrough, long awaited, for significantly improving the volumetric and gravimetric energy density of hydrogen storage systems.

Two carbon nanostructures are of major interest for hydrogen storage: nanotubes, one-dimensional structures with a diameter of 1–2 nm and a few micrometer long, in a cylindrical shape and single (single-walled nanotube, SWNT) or multiple walled (multi-walled nanotube, MWNT), looking like a wound sheet of graphite; and nanofibres (graphite nanofibres, GNF), longer than nanotubes (a few cm), but with the same length:diameter ratio. Internally, carbon nanofibres are solid with an ordered stack of nanocrystals, evenly spaced at 0.34–0.37 nm, which are bonded by van der Waals forces in a flexible nanopore structure; they are composed of a nanotube, with a structure of small graphite layers that may be parallel — *tubular nanofibres*, perpendicular — *platelet nanofibres*, or inclined — *herringbone nanofibres* with respect to the nanotube axis. Numerical simulations and molecular dynamics have been applied to define theoretical models for studying the thermodynamic and structural properties of these new classes of carbon-based materials. Fig. 7 shows three sequences of a semi-empirical simulation, demonstrating the transformation of two graphene fragments rolling in to a closed carbon nanotube [19,20].

Fig. 8 shows a schematic section of a carbon nanotube structure with possible localisation of hydrogen.

These materials can adsorb significant amounts of hydrogen at room temperature, but research is still needed to better understand the way they work, to develop reproducible production techniques and to confirm, to a certain extent, contrasting experimental results. Carbon nanostructures have been tested at various operating conditions with pressures from a few bars up to some hundreds of bars, temperature ranging between 80 and 800 K, with percentage of hydrogen adsorption in weight varying from a few percent up to an incredible 60%.

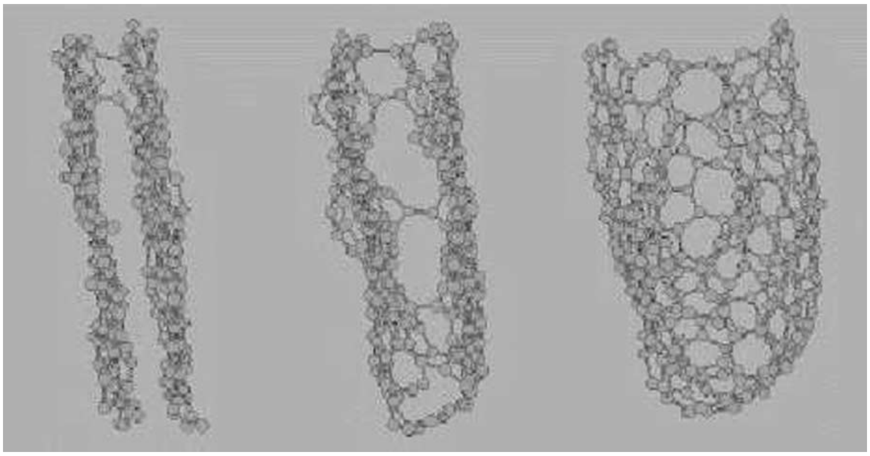


Fig. 7. Three steps, in a time period of 3 ps, of semi-empirical simulation (molecular dynamics) with two graphene fragments rolling into a closed carbon nanotube.

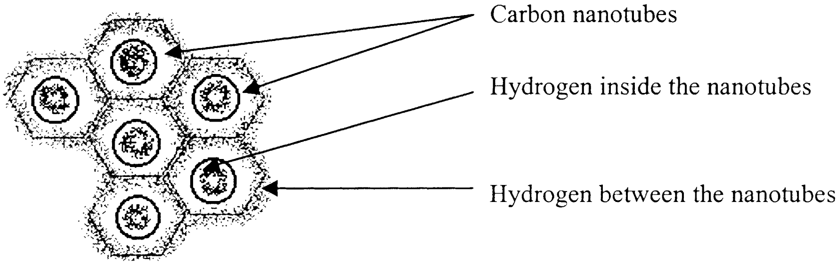


Fig. 8. Pictorial view of possible locations of hydrogen in a structure composed by carbon nanotubes.

Table 3
Carbon structures for hydrogen storage

Material	Temperature (K)	Pressure (bar)	H_2 (wt.%)	Specific energy (kWh/kg)	H_2 (vol.%)	Energy density (kWh/l)
SWNT, high purity [21]	300	0.65	4			
SWNT, high purity [22]	80	70.9	8.25			
MWNT [23]	298–773	1	0.4	0.133	3.2	0.106
Li-MWNT [23]	473–673	1	20	6.66	180	6.0
K-GIC[a] [23]	313	1	5	1.66	60	2
Li-GIC [23]	473–673	1	14	4.66	280	9.32
GNF tubular [24]	298	112	11.26		1.42	
GNF tubular [25]	298	110	10–12			
GNF herringbone [24]	298	112	57.85		13.35	
Graphite [25]	298	112	4.52		0.53	

[a] GIC: graphite intercalation compounds.

Table 3 summarises various experimental results for carbon nanostructures, which show that very high volumetric and gravimetric energy density seem to be possible, not far from the values achievable for gasoline tanks.

Due to the intriguing potentialities of carbon-based storage systems, a great deal of R&D is devoted to optimise production processes and manufacturing technologies up to a mass-production of highly-purified materials, with much higher specific energy.

2.3. Utilisation

Beside its use in the chemical and oil industries hydrogen can be used as fuel both for power generation and or combined heat and power (CHP) production [26]. It can be used for transportation as well, combining in all cases benefits both in efficiency and in reduction of environmental impact.

2.3.1. On-site power generation

2.3.1.1. Fuel cells. There are different fuel cell technologies, at various stages of development and characteristics [27,28]. Most of them, anyhow, can make use of hydrogen as the fuel achieving the highest efficiency, up to more than 60%. They will have widespread use because they present many characteristics that make them a key technology in a future economy based on hydrogen:

- high efficiency and reliability;
- modular structure makes them suitable for a variety of applications;
- excellent performance at partial load and;
- limited or no atmospheric or acoustic emissions.

Moreover, with pure hydrogen it is also possible to reach the best performances of fuel cell systems in terms of simplicity, modularity and environmental protection. Actually, these systems are absolutely zero emission when fuelled with pure hydrogen, giving just water vapour as exhaust gas.

For power generation, the technological options, in the short- and medium-term, can be:

- polymer electrolyte fuel cells (PEMFC) in the power range from few kW up to hundreds of kW for generation and CHP in residential and light industry applications;
- high temperature fuel cells, molten carbonate fuel cell (MCFC) and solid oxide fuel cell (SOFC) for power from some hundreds of kW to tens of MW for distributed generation and industrial co-generation.

In the long-term fuel cells, mainly MCFC and SOFC, will be available for centralised power generation in sizes up to tens of MW.

In the mid-term, fuel cells can constitute a useful bridge from today's energy economy, based on fossil sources, towards a future hydrogen system, in consideration of the fact that:

- over the next 5 years, their development level will reach the realisation of plants of significant size for diversified applications. Such power plants will use hydrogen produced from reforming and will be integrated with systems for CO_2 sequestration and utilisation, as a transitional solution;
- they can be integrated into advanced power generation cycles that combine fuel cells with gas turbines for achieving potentially higher efficiencies and lower emissions than the individual systems separately. Electrical conversion efficiencies of over 70% are calculated for these hybrid configurations.

2.3.1.2. Gas turbines. Over the last few years, many organisations world-wide have been working on the development of advanced gas turbine systems, capable of achieving higher efficiencies and operating with a lower environmental impact than today's units. In the near term, industrial-scale gas turbines with efficiencies of 40–45% will be available, as a result of these R&D efforts. Initiatives are also under way develop gas turbine combined-cycle systems with efficiencies around 55–60%.

Gas turbine technology is mature with natural gas as fuel and hydrogen-powered plants are feasible although hydrogen's different characteristics (i.e. higher flame temperature) require further development activities. There is a need for temperature-resistant materials and better cooling techniques, especially when pure oxygen is used.

The Japanese government, within the World Energy Network (WE-NET) program [29], is currently working with turbine manufacturers (Mitsubishi Heavy Industries, Toshiba) to develop hydrogen-based systems that include combustion turbines. The overall goal of the program is the demonstration of a hydrogen-power system with efficiency exceeding 70% and low NO_x emissions by 2020.

2.3.2. Hydrogen as a fuel in the transportation sector

Hydrogen can also be used in fuel cells and internal combustion engines in transport applications. Many automotive industries are active in programs of development of transport means powered with fuel cell systems and they are allocating significant investments to drive the technology towards the commercialisation [30–32].

Fuel cells are considered the most promising power source for future generation vehicles and the only technology with the potential of competing with internal combustion engines. With pure hydrogen, a fuel cell vehicle is a true ''zero emission'' vehicle, producing only water as by-product. Fuel cell vehicles offer efficiencies two to three times higher than those of conventional vehicles, maintaining similar performances in terms of range, top speed and acceleration. NECAR 4 (Fig. 9), presented by DaimlerChrysler in March 1999, operating with liquid hydrogen, shows an efficiency of 37.7% according to the New European Driving Cycle. This value is high if compared with that typical of ICE vehicles (16–18 and 22–24% for gasoline and diesel vehicles, respectively) [33].

Fig. 9. DaimlerChrysler NECAR 4, powered by a PEMFC and fuelled by pure hydrogen.

Among the various types of fuel cells, the PEMFCs are the most suitable for mobile applications due to high power density, quick start-up and rapid response to load changes. Ballard Power System, Nuvera Fuel Cells and International

Table 4
List of PEMFC-powered vehicles

Organisation	Vehicle type	Year	Fuel
DaimlerChrysler	NECAR 3	1997	CH_3OH
	NECAR4	1999	Hydrogen (liquid)
	NECAR 4A	2000	Hydrogen (compressed)
	NECAR 5	2000	CH_3OH
	Jeep Commander 2	2000	CH_3OH
Ford Motor Co.	P2000	1999	Hydrogen (liquid)
	Focus FC5	2000	CH_3OH
General Motors/Opel	Zafira	1998	CH_3OH
	HydroGen1	2000	Hydrogen (liquid)
Honda Motor Co.	FCX-V1	1999	Hydrogen (compressed)
	FCX-V2	1999	CH_3OH
	FCV-V3	2000	Hydrogen (compressed)
Madza Motor Co.	Demio (2a gen)	1999	Hydrogen (stored in metal hydrides)
	Premacy FC-EV	2000	CH_3OH
Nissan Motor Corp.	R'nessa	1999	CH_3OH
	Xterra	2000	CH_3OH
Toyota	RAV4	1996	Hydrogen (compressed)
	RAV4	1997	CH_3OH
	FCHV-3	2001	Hydrogen (stored in metal hydrides)
	FCHV-4	2001	Hydrogen (compressed)
Hyundai	Santa Fe FCEV	2000	Hydrogen (compressed)
Volkswagen	Capri	1998	CH_3OH
	Bora HyMotion	2000	Hydrogen (liquid)
Renault	Laguna (FEVER)	1997	Hydrogen (liquid)
Fiat	600 Elettra	2001	Hydrogen (compressed)

Table 5
List of fuel cell-powered buses

Organisation	Vehicle type	Year	Fuel	Fuel cell type
Ballard Power Systems/New Flyer	Transit bus, 12 m	1997	Hydrogen (compressed)	205 kW PEFC Ballard
DaimlerChrysler	NEBUS	1997	Hydrogen (compressed)	250 kW PEFC Ballard
	ZEBUS	1999		190 kW PEFC XCELLSIS
	Citaro bus, 12 m	2002		250 kW PEFC XCELLSIS
Neoplan	Midi-bus, 8 m	2000	Hydrogen (compressed)	55 kW PEFC Nuvera
MAN Nutzfahrzeuge AG	MAN bus, 12 m	2000	Hydrogen (compressed)	120 kW PEFC Siemens
		–	Hydrogen (liquid)	150 kW PEFC Nuvera
Proton Motor	Neoplan bus (Bayer-Bus II)	2000	Hydrogen (compressed)	80 kW PEFC
Georgetown University	Novabus, 12 m	1995	Methanol	100 kW PAFC IFC
	X1 bus	2000		100 kW PEFC XCELLSIS
FCBus Project	SCANIA city-bus	2000	Hydrogen (compressed)	60 kW PEFC Nuvera
FIAT/Irisbus Project	City-bus, 12 m	2001	Hydrogen (compressed)	60 kW PEFC IFC
Toyota Motor Corp.	FC-BUS 1 City-bus, 10.5 m	2001	Hydrogen (compressed)	90 kW PEFC Toyota

Fuel Cells, industry leaders in PEMFC technology development, are supplying products to major automakers, including DaimlerChrysler, Ford, Volkswagen, Honda, Nissan, Hyundai and FIAT. The SOFC technology could be successful as an alternative to PEMFCs in niche applications such as auxiliary power units for large vehicles.

As shown in Tables 4 and 5, most of the car manufactures are involved in PEMFC-powered vehicle development and are carrying out field tests to analyse technical and economical feasibility in passenger cars and buses. Strategic alliances among fuel cell developers, automotive industries and oil companies have been created to accelerate the development and marketing of commercial FCFVs. The goal is to introduce them on the market place starting from 2004 to 2005.

However, there are a number of technical and economic barriers to overcome before these vehicles will achieve significant market shares and can be fully accepted by consumers. The high hydrogen production costs and lack of appropriate infrastructures, both hydrogen supply and delivery, remain critical.

A key issue is how to provide hydrogen to the fuel cell. There are two options: either to store it on board of the vehicles or to produce the hydrogen on the vehicle by means of a fuel processor. Transition to a widespread use of hydrogen is, therefore, being made gradually. In an early phase, this fuel will only be the solution for fleet vehicles, especially in urban areas where centralised refuelling stations might be used. In the near term, for light-duty vehicles, the hydrogen will be still produced on-board from conventional fuels; many experts consider methanol as an ideal hydrogen carrier.

Research is also focused on the development of internal combustion engines that operate on pure hydrogen or hydrogen-blended fuels. Demonstrative activities on hydrogen-powered internal combustion engines are currently underway by DaimlerChrysler and BMW. For over 20 years, the BMW group has been working in this field. After four vehicle generations of improved technology and design, BMW in May 2000 introduced the 750hL, a liquid-hydrogen car built for the first time under conditions similar to those of series production [34]. A fleet of 15 vehicles is already on the road performing shuttle services in Munich and Hanover.

3. International programs

Most of the major R&D programs in the energy field are paying attention to hydrogen as a fuel and energy carrier and to the development of related technologies.

3.1. United States

In the United States, the Department of Energy (DOE) Hydrogen R&D Program, started in 1979, promotes applied R&D activities in the areas of hydrogen production, storage, and utilisation, for the purpose of making hydrogen a cost-effective energy carrier for utility, buildings, and transportation applications [35,36].

Goals of the DOE Hydrogen R&D Program are to facilitate the transition of hydrogen energy production from fossil fuel-based sources (mainly natural gas) to renewable sources (including solar, wind and biomass) and to cost-share with industry the validation of production, storage and utilisation technologies leading to commercialisation.

This program co-ordinates with a number of other DOE programs that are closely correlated with hydrogen technology including: coal gasification and fuel cell programs under the Office of Fuel Energy, the vehicle fuel cell program of the Office of Transportation Technologies and the biomass gasification program within the Office of Utility Technologies. Another program linked to hydrogen is the Vision 21 Program (Clean Energy Plants for the 21st Century) [37]. The primary goal of this program is to develop, by 2015, power plants operating on fossil fuels with practically zero environmental impact, in which CO_2 emissions can be reduced to zero through sequestration [38].

3.2. Japan

In Japan, the WE-NET Project, started in 1993 by New Energy and Industrial Technology Development Organisation (NEDO), aims at developing, by 2020, basic technologies for realising a hydrogen-based energy economy. The WE-NET program proposes to convert hydropower and other renewable energy, that are available in abundant quantities, into hydrogen through water electrolysis and other appropriate processes. Hydrogen would be stored close to the energy consumption areas and used to generate electricity by means of hydrogen combustion turbines and in other applications, such as hydrogen vehicles and fuel cells etc.

3.3. European Union

Since the early 1990s, various demonstration projects on hydrogen have started in Europe [39]. In the framework of the EQHHPP (Euro-Quebec Hydro-Hydrogen Pilot Project) Project, many European industries and research organisations took part and worked on different hydrogen applications. Nowadays, activities are carried out mainly in Norway and Germany. In Norway, a group of national industries and institutes has carried out a feasibility study on hydrogen as a future environmentally friendly energy carrier. This study concludes that Norway is well-disposed for the industrial development of the hydrogen economy, partly because it is a natural gas producer and has large amounts of hydropower, but also because of the existing expertise of industries, universities and research institutes in the production of hydrogen by electrolysis.

Moreover, Norsk Hydro, a Norwegian's hydroelectric company, is designing a demonstration plant for generating electricity from hydrogen, produced by natural gas, in combined-cycle gas turbines. The CO_2 produced in the process will be separated and injected into a deep saline aquifer. Activities for CO_2 sequestration are already taking place at Sleipner field, off the coast of Norway, where around 1 million of tonnes per year of CO_2 are removed from extracted natural gas. The separated CO_2 is injected into an aquifer 1000 m under the North Sea [40].

Intensive R&D work has also been done in Germany, where activities for the energetic use of hydrogen have been running since the 1970s with the involvement of several private companies and research institutions [41]. These activities included not only the development of prototypes and demonstration vehicles equipped with fuel cells or internal combustion engines, using compressed or liquid hydrogen, or the demonstration of fuel cell plants for cogeneration, but also basic research in the field of hydrogen production and storage.

Recently, a new company, the Icelandic New Energy Ltd., has been set up as a joint-venture company owned by VistOrku hf (EcoEnergy),[1] DaimlerChrysler AG, Norsk Hydro ASA and Shell Hydrogen BV, with the aim of investigating the potential for eventually replacing fossil fuel use in Iceland with hydrogen and creating the world's first hydrogen economy [42]. Iceland was chosen because in this country they can operate a "hydrogen-based fuel project" in a CO_2 neutral environment and they have similar standards and transportation systems to most other developed countries and, therefore, the results could easily be adapted elsewhere. The transition to hydrogen would occur over a 30–40 years time scale. By the end of 2002, three DaimlerChrysler buses with hydrogen-powered fuel cells will be demonstrated on the streets of the capital, Reykjavik, with refuelling available from a Shell service station. The joint venture ultimately aims to convert both the public and private transportation sectors, including the fishing fleet. The principal problem to be overcome is the limitation of current on-board hydrogen stðrage options for vehicles.

In the framework of European Commission Programs, the 5th Framework Program [43] included hydrogen activities, even if no specific reference was made to this fuel. In the program "Energy, environment and sustainable development", the activities concerned with the generation of electricity and/or heat with reduced CO_2 emission from biomass or other fuels. Moreover, development and demonstration of new and renewable energy sources into energy systems was foreseen.

Fuel cells received considerable attention because of their attractiveness as a key technology for meeting the European Union's policy goal of sustainable development. Fuel cells provide an essential link between today's fossil-based energy economy and future renewable energy systems.

Future hydrogen activities will certainly be promoted under the 6th Environment Action Program of the European Union which outlines the priorities for action on the environment for the next 5–10 years. The new program focuses on four major actions areas: climate change, health and the environment, nature and bio-diversity and natural resource management [44].

The achievement of the Community's 8% emission reduction target for 2008–2012 under the Kyoto Protocol is a focus of the proposed new program. However, the Commission also calls for more far-reaching global emission cuts in the order of 20–40% by 2020 and cites the scientific estimate that in the longer term a 70% global GHG reduction as compared to 1990 will be needed.

The program points to the need for structural changes, especially in the transport and energy sectors, and calls for stronger efforts in energy efficiency and energy saving,

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[1] VistOrku (EcoEnergy) is owned by Icelandic New Venture Fund, University of Iceland, IceTech, Fertilizer Plant, Suðumes Regional Heating Corporation, Iceland National Power Company and Reykjavík Energy.

further research and technological development and awareness-raising with citizens, so that they can all contribute to reducing emissions. In this specific area, hydrogen activities could be foreseen.

In Italy, ENEA, in co-operation with Enitecnologie, has been working to define a national program to develop the technologies necessary for the introduction of hydrogen as an energy carrier. During the initial phase of the project, hydrogen will also be derived from fossil sources (natural gas), and then it will be generated from renewable resources. The program is aimed at promoting, in an organic framework, a series of actions regarding the whole hydrogen cycle (production, storage, transport and utilisation). In parallel, research activities will be addressed to the development of technologies for separation and sequestration of CO_2. The long-term goal of the program is to make environmentally compatible the remaining fossil energy sources, extending their utilisation, and to promote new technologies and renewable energy sources, giving them industrial validity and maximising the social and economical impact in various sectors.

In the meantime, various activities related to hydrogen use are underway or will be started before long. AEM S.p.A., the municipal energy company of Milan and the German automaker BMW announced a plan for building jointly the first Italian hydrogen filling station. The filling station is to be erected in a technology area, named '*Bicocca*', near Milan on the 1.3 MW PAFC demonstration plant site. The hydrogen will be used for demonstrations of public transport vehicles and for supplying a stationary fuel cell power plant (the plant will use a 500 kW MCFC unit by Ansaldo Ricerche).

In Italy, Nuvera Fuel Cells Europe and Ansaldo Ricerche are working on the development of PEMFC units for stationary and transport applications. Currently, Nuvera is testing and demonstrating the commercial viability of small-scale polymer electrolyte fuel cell power modules (1–50 kW) [45]. In Fig. 10, the last generation of a 1 kW Nuvera PEMFC generation unit is shown.

In regard to MCFCs, Ansaldo Ricerche is developing a "*Series 500*" unit (Fig. 11), designed for both direct use and as "building block" for larger plants, up to 20 MW (Fig. 12) [46,47]. This unit should lead to the transition of MCFC technology from laboratory to commercial-scale demonstrations.

The development of fuel cell vehicles in Italy is just starting, even if various industries have been involved in the past in international projects. In February 2001, FIAT introduced its first prototype of fuel cell car, the "Seicento Elettra H_2 Fuel Cell" — developed at Fiat's Research Centre with the support of the Italian Ministry of the Environment [48].

The first hydrogen-powered urban fuel cell bus, developed in Italy was presented in June and will be in public service in the city of Turin within 2001 [49]. In this project, the following are involved: ATM (municipal transport company of Turin), Iribus Italia, Sapio Produzione Idrogeno Ossigeno, Compagnia Valdostana Acque (CVA), Ansaldo Ricerche, the Ministry of the Environment and ENEA. The bus, in hybrid configuration, is fuelled with electrolytically produced hydrogen and equipped with a battery system. The FC (supplied by International Fuel Cells) has a power of 60 kW (Figs. 13 and 14).

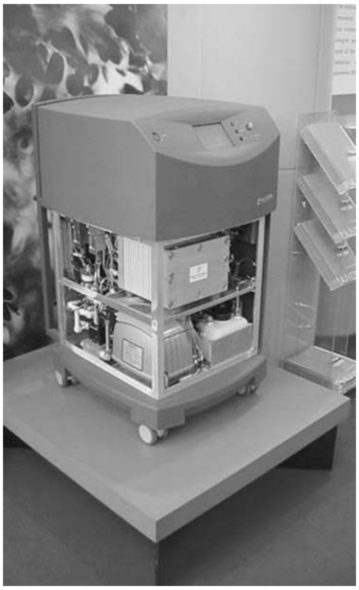

Fig. 10. A 1-kW Nuvera PEMFC generation unit.

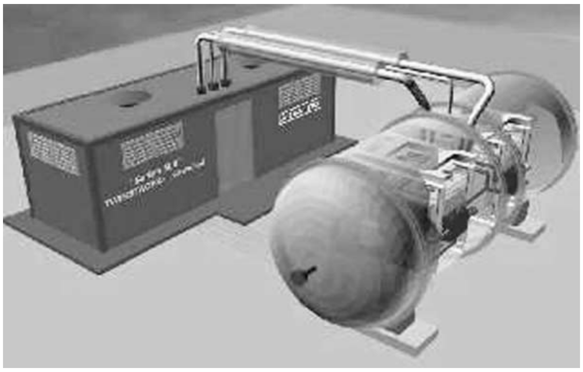

Fig. 11. The *Series 500* unit.

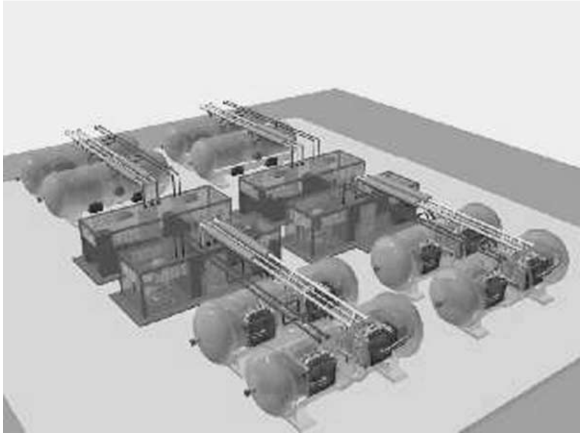

Fig. 12. Study for a 4-MW power plant based on the *Series 500*.

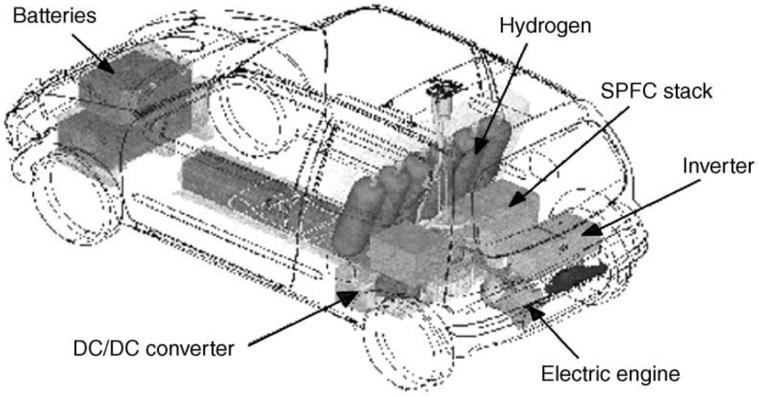

Fig. 13. FIAT 600 Elettra H_2.

Fig. 14. Prototype of a PEMFC Irisbus City Class public bus.

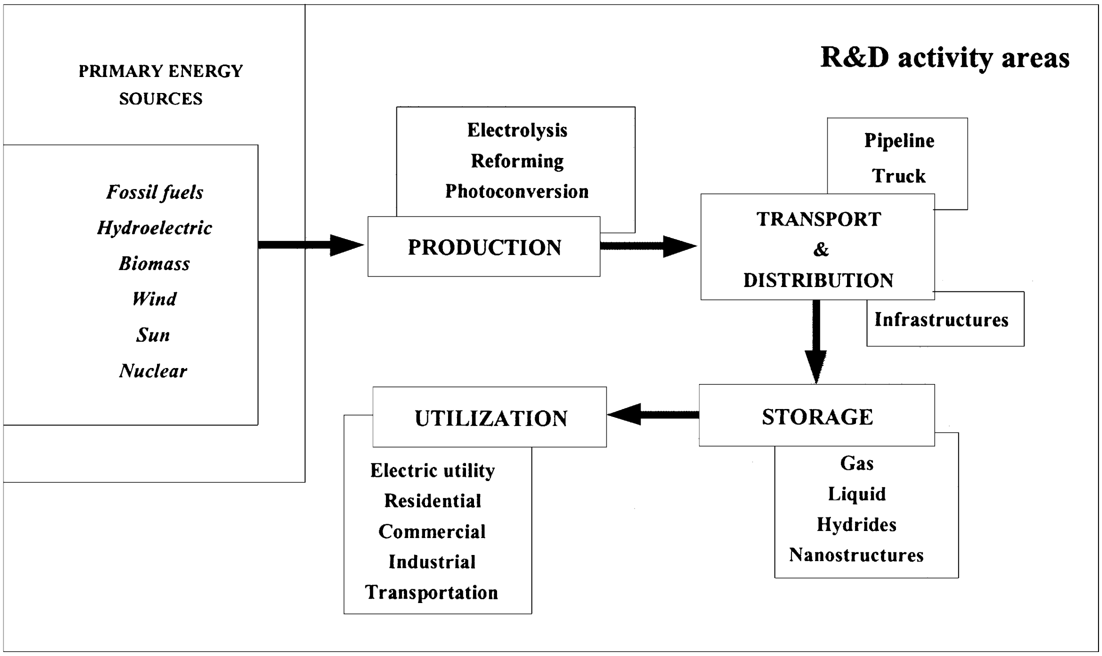

Fig. 15. Major R&D activity areas for the pathway to hydrogen economy.

Table 6
Roadmap for the development of a hydrogen economy

Areas	Activity lines		Tasks
Hydrogen production	From fossil fuels	Short- to medium-term	Optimisation of current industrial processes starting from natural gas, coal and oil
			Small-scale demonstrations to prove technical feasibility of CO_2-free hydrogen production plants
		Long-term	Membrane-based reformer for joint production and purification of hydrogen
			Methane pyrolysis, which directly produces carbon and hydrogen without CO_2
	CO_2 logistics and sequestration	Short- to medium-term	Study and research on various separation and sequestration methods
			Study of technology and economics of CO_2 transport
			Demonstration of CO_2 sequestration in geological caverns or aquifers
		Long-term	Biological and chemical processes for the sequestration and recycling of CO_2
	From renewable energy sources	Short- to medium-term	Hydrogen from biomass by gasification and pyrolysis
			Electrolysis with wind and sun power
Transport, distribution and infrastructure		Long-term	Water dissociation with thermal solar energy
			Processes of biological and photoelectrochemical conversion
		Short- to medium-term	Study and research of various solutions
			Technical and economical study of infrastructure
		Long-term	Demonstration of small networks
Storage		Short- to medium-term long-term	Optimisation and demonstration in vehicles of conventional methods: liquid, compressed, metal hydrides
		Long-term	More performing and cheap metal hydrides (e.g. Mg-based)
			Complete development of fabrication and purification techniques of carbon nanostructures
End use	Stationary applications	Short- to medium-term	Feasibility study for a centralised electricity production with combined cycles
			Distributed electricity generation with fuel cells, microturbines, hybrid systems
		Long-term	Development of solid oxide and molten carbonate fuel cells
			Demonstration of pilot plants with new generation of fuel cells
	Transport	Short- to medium-term	Development of fuel cell stacks, storage systems, ancillaries, electronics
			Fuel cell generators integrated in prototypes
			Demonstration of pre-series fuel cell vehicles
		Long-term	Improvement of production processes
			Widespread applications of fuel cell vehicles
			Establishment of filling stations
Safety standards, codes and regulations			Creation of safety standards and regulations for hydrogen use in vehicles
			Creation of standards and regulations for refuelling infrastructure

4. A vision for a 21st century hydrogen economy

The selection and use of clean fuels and energy sources can be envisaged as the most intriguing challenge for the future of the environment and society. The flexibility in production from a variety of feedstocks such as fossil fuels up to renewable energy sources, the cleanliness in various phases of the energy chain and the large availability make hydrogen an ideal candidate for becoming the best solution as a fuel and also energy carrier. Various experts agree upon the vision that the future energy economy will be mainly based on hydrogen (probably in combination with electricity) [31,35]. The pathway for the transition from current energy economies to a hydrogen economy is paved with many scientific, technological and economical drawbacks. Only in a not-close future, the hydrogen vision will be fully accomplished; several transitional, gradual steps will be required. The most significant milestones for the hydrogen pathway must be mainly based on the intensification of R&D programs, even of an international nature, on critical technologies. Fig. 15 depicts and recommends R&D priority areas.

The acceleration of the pace towards a hydrogen economy will require a concentration of efforts on critical topics in R&D areas, sketched in Fig. 15. Table 6 outlines some of the major activities, which are mostly shared among major R&D programs.

The global climate change demands a new era of clean, safe and sustainable energy supply. The creation of a hydrogen energy economy will require the implementation of the initiatives and programs briefly described above, which gradually move from a new way to use conventional fuels and energy sources (mainly fossil fuels) to a completely new energy economy based upon renewable energy sources and hydrogen/electricity fuels and carriers. The joint effort of major stakeholders (research organisations, industry, international bodies, association, standard setting bodies) at national and international level will be essential for an early development and diffusion of hydrogen as a fuel and energy carrier by means of a world-wide network of expertise, knowledge and products.

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