Hydrogen Production by Nuclear Heat

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A major shift in the way the world obtains energy is on the horizon. For a new energy carrier to enter the market, several objectives must be met. New energy carriers must meet increasing production needs, reduce global pollution emissions, be distributed for availability worldwide, be produced and used safely, and be economically sustainable during all phases of the carrier lifecycle. Many believe that hydrogen will overtake electricity as the preferred energy carrier. Hydrogen can be burned cleanly and may be used to produce electricity via fuel cells. Its use could drastically reduce global $CO₂$ emissions. However, as an energy carrier, hydrogen is produced with input energy from other sources. Conventional hydrogen production methods are costly and most produce carbon dioxide, therefore, negating many of the benefits of using hydrogen. With growing concerns about global pollution, alternatives to fossil-based hydrogen production are being developed around the world. Nuclear energy offers unique benefits for near-term and economically viable production of hydrogen. Three candidate technologies, all nuclear-based, are examined. These include: advanced electrolysis of water, steam reforming of methane, and the sulfur-iodine thermochemical water-splitting cycle. The underlying technology of each process, advantages and disadvantages, current status, and production cost estimates are given.

KEYWORDS: hydrogen, sulfur, iodine, steam reforming, electrolysis, cost, nuclear, HTGR

I. The Energy Carrier of the Future

 Research and development is constantly underway to create the next generation energy technology. It is impossible to know what the future holds, but future energy technology must certainly possess the following characteristics:

- Since energy needs will continue to increase, the new technology must be expandable,
- Future energy sources must be environmentally sound – "green energy" will become an increasingly important motivator,
- The distribution of energy must be effective and allow for expansion to improve the standard of living around the world,
- Energy production, transmission and use must be safe (both secure from outside threats and with a low risk to health and safety),
- The energy technology must be economical.

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 The last major shift in the energy market occurred with the introduction of electricity. Over the last century, electricity has satisfied each of the above criteria. It is expandable. It is an environmental improvement over the direct use of fossil fuels. Its transmission over the grids is robust and expansive. It is easy to use safely. And electricity is affordable. But as energy needs continue to increase, new solutions will need to be added to those offered by electricity.

 With these factors in mind, many believe that hydrogen is poised to be the next big revolution in the energy market. This paper considers several options for the production phase of this energy medium. The collection, transmission, and end-uses of hydrogen are not discussed. Specifically, the potential of three hydrogen production processes under development for the industrial production of hydrogen using nuclear energy are compared and evaluated. These are:

- Advanced Electrolysis,
- Steam reforming, and the
- Sulfur-Iodine water splitting cycle.

 Water electrolysis and steam reforming of methane are proven and are used extensively for the production of hydrogen today. The Sulfur-Iodine cycle, a thermochemical water splitting process, is of particular interest because it produces hydrogen efficiently with no CO₂ byproduct.

 The purpose of this paper is to familiarize the reader with the current status of nuclear-based hydrogen production, and to speculate as to which of these processes is the best candidate technology that will start the age of the "hydrogen economy," which many experts agree is on the horizon.

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II. The Basics of Hydrogen Production and Nuclear Energy

1. Hydrogen

 Hydrogen is the most abundant element in the universe and the third most abundant on Earth [Reference 10]. Hydrogen gas does not exist on the earth or in our atmosphere in significant quantities. Instead, it reacts quickly with other elements to form more stable compounds. Hydrogen compounds are abundant in water and fossil fuels; its supply is effectively limitless.

 Because pure hydrogen is not as readily available as fossil fuels, hydrogen is not considered to be a source of energy but an energy carrier. Like electricity, hydrogen is "manufactured". Energy carriers are a convenient medium to store, transport, and use energy. But the convenience comes at a price, that is, efficiency.

 There are several methods for producing hydrogen. All involve splitting compounds that contain hydrogen and capturing the hydrogen gas that results. To split water directly with heat (thermolysis) requires temperatures in excess of 2500ºC for significant hydrogen generation [Reference 1]. This method is not currently practical for industrial production, as those temperatures cannot be sustained. Instead, thermochemical cycles or electrical drivers allow the splitting to occur at lower temperatures.

 Hydrogen has been researched as an energy transport medium since the 1960s. Two recent technological developments have piqued the interest in hydrogen: (1) fuel cell technology to cleanly make electricity on location (or perhaps in vehicles) and (2) the direct use of hydrogen as a fuel, such as in rocket fuel or military aircraft. Currently, hydrogen production is a major area of research throughout the world, especially in the US, Europe, and Japan [Reference 6].

 Burning hydrogen with oxygen, as is done in the space shuttle, creates no pollution. The only byproduct of that combustion is water. Burning hydrogen with air does form some pollutants, such as NOX, but in much smaller quantities than when burning fossil fuels [Reference 10]. Therefore, there are significant potential environmental benefits to the use of hydrogen as an energy carrier.

 As research progresses, the use of hydrogen will become increasingly economical and environmentally sound. But all phases of the life cycle of hydrogen must be effective and efficient for it to thrive in the market. Current production of hydrogen is dominated by electrolysis of water and steam reforming of natural gas (or methane). The majority of end users manufacture

their own hydrogen on site for use as a feedstock in other processes [Reference 13].

2. Nuclear Energy

 Nuclear energy offers an abundant source of energy that will be available well into the future. Nuclear fuel supply is estimated to be readily available even on a oncethrough fuel cycle for fifty to one hundred years. Using breeder reactors and a closed fuel cycle, it is virtually inexhaustible. Because of its relative abundance, nuclear fuel is relatively cheap compared to fossil-based fuels.

 Nuclear power is an extremely clean and environmentally friendly energy source. There are virtually no polluting emissions from its use and only a small amount of waste results.

III. Nuclear Based Hydrogen Production

 While hydrogen has received much attention recently, there are few industrial facilities for large-scale hydrogen production [Reference 8]. Nuclear power offers a unique solution to hydrogen production processing, with several distinct advantages:

- The new hydrogen production technologies powered with nuclear energy offer increases in efficiency and dramatic reductions in pollution, when compared with traditional hydrogen production.
- Nuclear-based hydrogen production potentially satisfies each of the five criteria for a future energy carrier discussed earlier.
- Using nuclear energy offers advantages in each phase of the lifecycle of an energy medium: collection, production, transmission and distribution, and end-use. Production via nuclear is reliable and safe.
- And, it appears to be economically viable.

 Nuclear produced hydrogen has the opportunity to take advantage of clean nuclear energy to create an environmentally friendly and highly versatile energy option.

 This paper examines the three processes that have been proposed for integration with a nuclear reactor for the production of the future hydrogen commodity.

 In any process there are inputs (feedstock and energy) and outputs (desired chemical and losses). Figure 1, illustrates this concept. The thermal efficiency of a production process is defined to be the lower heating value of the hydrogen produced divided by the energy input into the system from all sources (in the form of: heat, separations, work, etc.).

Fig.1 Production Process

 Hydrogen as a consumer commodity is the postulated product of the hydrogen production processes to be compared. Cost of purification and storage costs, in addition to the cost of production itself, ought to be included in the overall cost estimate. However, the major cost for hydrogen production results from the input energy. Therefore, the costs for materials (for the input of water and/or natural gas or other hydrocarbons) and end processes (purification and storage) are not specifically accounted for. These costs exist for all processes compared, and therefore, the effect of any deviation resulting from this assumption will not dramatically affect the relative costs.

IV. Electrolysis

 Electrolysis is the splitting of water molecules by electricity, and is the most well known of the methods for producing hydrogen. It is considered a candidate for hydrogen production with nuclear energy because it may be combined with either existing nuclear electrical generating plants or with new, high efficiency nuclear generating plants.

A simple process schematic is shown in Figure 2.

Fig.2 Electrolysis Process Diagram

 The fundamental reactions that make electrolysis possible are the half reactions of the water ions. These are shown below. Production facilities for hydrogen (or oxygen) via electrolysis use solutions of various compositions (generally salts) to increase the speed of the reaction, since it is not very favorable in pure water (alternatively, membrane-based technologies may also be used, however, these are not examined in this paper).

 $2H_3O^+ + 2e^- \rightarrow H_2 + 2H_2O$ cathode $3H_2O \rightarrow \frac{1}{2}O_2 + 2H_2O^+ + 2e^-$ anode $H_2O \to H_2 + \frac{1}{2}O_2$

 Electrodes, a cathode and an anode, placed in the solution drive the movement of electrons. Hydrogen forms at the cathode, and oxygen at the anode. Electrolysis is a very effective way of producing pure hydrogen in small production facilities.

 Because electrolysis uses electricity, the overall thermal efficiency of the process includes the efficiency of the electrical power generation, as well as that of the electrolysis itself (see Figure 2). The electrolysis process efficiency is generally about 75%, but the efficiency of electrical power generation is only about 30% in most cases. This means that the overall thermal efficiency for hydrogen generation from standard electrolysis technology is only about 25%. The range of possible thermal efficiencies for electrolysis is 25- 45%.

1. Advantages and Disadvantages

 The main disadvantage to electrolysis is the limited efficiency of electrical generation.

 Electrolysis in conjunction with new nuclear reactor technology could increase the efficiency of the process simply by providing the cheaper electricity to the process. This is because newer nuclear plants operate at higher temperatures and have higher thermal efficiencies.

 Another possibility for improving the economic conditions for electrolysis is through cooperation with electricity generators. If agreements could be made between the electricity generators and the hydrogen production facilities, costs may be reduced by using off peak electricity for the electrolysis production. Electricity generators would benefit by stabilizing production and eliminating transients [Reference 8].

 Because electrolysis uses electricity as an input, it does not have the geographical restrictions that a thermal process implemented with nuclear heat does. The plant may be located some distance from the nuclear station and therefore reduce or eliminate many safety concerns. It also allows flexible and remote siting of hydrogen generators, providing distributed generation of this energy carrier, without requiring physical transport and large scale storage of the gas itself.

2. Current Status

 While water-splitting by electrolysis has been used extensively for decades, its use has been limited to small production units. Electrolysis is not generally considered viable for larger plants because of the low efficiency as compared with steam reforming. (There have been several large electrolytic hydrogen plants, consuming over 100MW, as well as thousands of smaller ones [Reference 9].

 However, recent research has advanced the technology and increased the efficiencies that are possible in hydrogen production by electrolysis.

 High-pressure high-efficiency cells have been successfully tested [Reference 9]. Theoretically, the cell efficiency in these units is about 90% and when coupled with high efficiency electric power plants these constitute the high end of the previously mentioned efficiency range for electrolysis.

 In electrolysis there is a trade off between the efficiency of the process and the speed at which it takes place. Therefore, multiple variables go into the design of an electrolytic cell. For the current comparison, the process described in Reference 8 has been assumed. This process is more efficient than those most common in plants today; however, it represents the future of electrolysis technology that would likely be used in the hydrogen economy.

V. Steam Reforming

 Steam reforming is a thermochemical process that is commonly used in industry for the production of hydrogen. The process consists of reacting methane (or, more commonly, natural gas) and steam at high temperature.

Fig.3 Steam Reforming Process

 There are two reactions which occur. The first, a reforming reaction, is highly endothermic and occurs with a catalyst at high temperature. The second reaction, the shift reaction, is exothermic. A follow-on separation process removes the carbon dioxide and purifies the hydrogen. Enthalpies of these reactions are listed in Table 1, below.

Table 1 Steam Reforming Process Reactions

| Step | Reaction Equation | Name | ΔH |
|------|-------------------------------------|-------------------|----------|
| | $CH_4 + H_2O \rightarrow CO + 3H_2$ | Reforming | 205.82 |
| | $CO + H2O \rightarrow CO2 + H2$ | Shift Reaction | -41.17 |
| | Separation Step | | |

 The conventional process occurs in a chemical reactor at temperatures of about 800-900ºC. When fueled with fossil fuels it is the most economical method of producing hydrogen today [Reference 7]. The heat is generally supplied by burning an excess of the methane. This results in a loss of both the reactant, and some of the product hydrogen. Typical thermal efficiencies for steam reforming processes are about 70% [Reference 7].

 Steam reforming reaction temperatures are achievable in a helium cooled nuclear reactor. By using a nuclear reactor for heat, the reactant loss may be avoided. In a reforming process with heat supplied by a nuclear reactor, the reformer cannot be the traditional furnace type. Instead, the heat must be supplied by a secondary loop from the nuclear side and be transferred to the methane/steam mixture via a heat exchanger type reactor. The diagram in Figure 4 shows a version of the Japanese concept for how this process might be connected with a High Temperature Gas Reactor [Reference 2].

Fig.4 Flow diagram of a hydrogen production system [adapted from Reference 2]

1. Advantages and Disadvantages

 The purity of the gas resulting from steam reforming must be improved for many of today's high purity hydrogen applications. The CO2 is removed by alkaline scrubbing, either with an amine solution or a regenerative caustic solution, and finally the hydrogen-rich gas is refrigerated to low temperatures and purified. This purification process incurs costs and is not, for example, needed in the electrolysis process.

 Although the steam reforming process and the nuclear reactors are both fairly well understood, there are some foreseeable problems involved with linking the nuclear reactor and the hydrogen production system. Even though there will be an intermediate heat exchanger to keep the primary and secondary sides separate, hydrogen can diffuse through the metallic wall, resulting in the mixing of hydrogen in some of the helium coolant. Circulating this hydrogen through the core can produce tritium that can diffuse the other way and can contaminate the hydrogen product. Other material concerns include the core graphite. If hydrogen is introduced into the core, then there is a possibility of corroding the graphite material. Studies are underway on how to deal with these issues and to establish countermeasures [Reference 2].

2. Current Status

 The current research on steam reforming in combination with a nuclear reactor is being led by the Japan Atomic Energy Research Institute (JAERI). They will use the HTTR (High Temperature Test Reactor), which is a

high temperature helium cooled reactor to provide the heat to the steam reforming system.

 They have developed the heat exchanger type reactor for the reforming process, and have done out-of-pile testing under simulated conditions to verify that the system operates properly and as desired. HTTR recently became fully operational, and they are on track to implement hydrogen production via steam reforming in 2008 [Reference 4].

VI. Sulfur-Iodine

 The Sulfur-Iodine (S-I) process is a thermochemical water splitting cycle. It consists of three chemical reactions, which sum to the dissociation of water. These reactions are given below.

 Theoretically, the only reactant that will need to be added to the cycle is water, as shown in Figure 5.

Fig. 5 Sulfur-Iodine Process

 Heat energy enters a thermochemical cycle through one or more endothermic high-temperature chemical reactions. Heat is rejected via exothermic low temperature reactions. All of the reactants, other than water, are regenerated and recycled. In the S-I cycle, most of the input heat goes into the dissociation of sulfuric acid. Sulfuric acid and hydrogen iodide are formed in the exothermic reaction of H2O, SO2 and I2, and the hydrogen is generated in the mildly exothermic decomposition of hydrogen iodide. [Reference 1)

 Figure 6 modified from Reference 11 shows a flow process diagram for the cycle.

Fig.6 Cycle schematic for the sulfur-iodine process [adapted from Reference 11]

 The efficiency of the sulfur-iodine process is not simple to estimate because the process is complex. For a reaction to be favorable the Gibbs free energy must be negative, or at least not too positive. From the recent General Atomics [Reference 1] study, the enthalpies of reaction (∆H) and the Gibbs free energies (∆G) for the three reactions in this process are summarized in Table 2.

Table 2 Enthalpy and Gibbs Free Energy for S-I Cycle Reactions

 The net loss in enthalpy of the process if we assume that the 3 reactions are the only contributors is about 12.5 kJ/mol. This might suggest that there is actually energy being released by the process, and that the energy input is insubstantial; however, this is not actually the case. A great deal of the necessary input energy is used in the separation steps. In addition, the process steps occur in various phase states, mostly liquid, and even a two-phase liquid. Complex modeling is necessary to determine the predicted cycle efficiency. The data provided in the references is therefore used to evaluate the process. These suggest that a thermal efficiency of hydrogen production of 50% is realistic [Reference 1].

 The key to successful implementation of the cycle, as found by GA, is using an excess of molten iodine in reaction 1 to give a two-phase solution, a light phase containing sulfuric acid and a heavy phase containing hydrogen iodide and iodine. The block diagram in Figure 7 shows the flow diagram of the cycle based on this separation [Reference 1].

Fig.7 Two-phase sulfur-iodine process [adapted from Reference 1]

1. Advantages and Disadvantages

 A major advantage of the S-I cycle is that there are virtually no harmful byproducts or emissions from the process. If heated with a nuclear source, it could prove to be an ideal environmental solution to hydrogen production.

 However, the constituents of the cycle are very corrosive, both the sulfuric acid and the hydroiodic acid solutions are very harmful and must be contained properly.

 Also, the control of the reaction conditions is a significant hurdle. It has been demonstrated in a laboratory, but control of an industrial size facility will be challenging.

 The hydrogen iodide solution has a quasi-azeotropic composition, making that separation step impractical by distillation. Electrodialysis with a new ceramic membrane has been employed to improve the hydrogen separation and has the potential to significantly improve upon the best efficiencies to date [Reference 5].

 Finally, the S-I cycle is at a disadvantage with the other processes considered because it remains in the research phase.

2. Current Status

 The Sulfur-Iodine cycle was developed at General Atomics, Inc. (GA) and first described in the mid 1970's. General Atomics conducted bench scale experiments of the total process, and the process was matched with the design characteristics of a high-temperature nuclear reactor in 1978 and 1980, with predicted efficiencies of 47% and 52% respectively [Reference 1].

 Researchers at the University of Aachen further demonstrated that the hydrogen iodide does not need to be separated from iodine before the decomposition step. Significant increases in efficiency and a 40% decrease in the cost of hydrogen are predicted compared with the standard flowsheet. Current research is working on incorporating these enhancements into the process [Reference 1].

 GA and the Japan Atomic Energy Research Institute (JAERI) are the leaders in current research and development on the use of the sulfur-iodine process in conjunction with nuclear power. JAERI has been doing extensive research and testing on the cycle for the past 20+ years. They have made considerable progress in improving the understanding of the cycle and have succeeded in continuous experimental operation for 48 hours of in a glass laboratory (see Figure 8) in 2001. Control of the reaction conditions is a significant hurdle, but they have proven that it is possible by lab testing.

Fig.8 Continuous hydrogen production test apparatus (Reference 3)

 JAERI is developing the process as a program for implementation with the High Temperature Test Reactor (HTTR). This reactor is a High Temperature Gas-Cooled Reactor (HTGR) which uses helium as the coolant [Reference 2]. In their independent research, General Atomics also concluded that a High Temperature Gas Reactor is the best option for nuclear implementation of this process and has done preliminary cost estimates for the cycle which are given in Table 3 [Reference 1].

 Continuing development will be carried out both by GA and JAERI. JAERI plans to extend the experimental results with the construction of a scaled-up test facility, and continues research on materials to be used in construction of the facility and for improvements to the process. GA is continuing work to incorporate new developments into an integrated flowsheet and will continue development.

VII. Comparative Analysis

 Table 3 summarizes the comparison of the three candidate processes for hydrogen production via nuclear energy. It includes cost estimates for hydrogen production for new plants of each type, including the cost of the nuclear reactor or electrical power plant as appropriate.

 As shown in the table, steam reforming remains the cheapest hydrogen production method based on the latest estimates, even when implemented with a nuclear reactor. The S-I cycle offers a close second, and electrolysis remains the most expensive of the options. In risk assessment, electrolysis has the lowest risk associated with both implementation and safety. Steam reforming and the S-I cycle each have considerable development risks and have some inherent technical risk in locating a hydrogen plant so close to a nuclear facility.

 If the reactants in the S-I cycle may be recycled efficiently as suggested, then the S-I cycle may prove to be an environmentally sound production method. In normal operation, there are no emissions other than hydrogen and oxygen. Similarly, electrolysis has no byproducts other than hydrogen and oxygen. Implemented with electricity from a nuclear power plant, the environmental impact of each of these options is minimal. The steam reforming process, however, does require fossil fuels, and produces greenhouse gas as a byproduct.

 Schedule to implementation of the proposed processes is also an important factor for consideration. Electrolysis development can currently support implementation of electrolysis plants, and if new nuclear plants were built, these could power such electrolysis operations right away. Steam reforming with nuclear power is a bit further off into the future. The first operation expected with a nuclear facility is in Japan in 2008 as part of the HTTR program. The S-I cycle implementation is still over the horizon. Research efforts to date have indicated that this is a feasible process for implementation. However, at the current rate it will likely be more than 10 years until we will see the S-I cycle in full scale operation with a nuclear reactor.

Table 3: Comparison Summary

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 1 The capital cost for the construction of the Modular Helium Reactor (as used in Reference 1) has been used for the nuclear capital cost in all three options (including the cost of the turbine-generator in the electrolysis case). The cost is based on a capital recovery factor for a 10% interest rate and a 40 year lifetime with zero recovery value at the end of plant life. Operating costs are assumed to be a similar percentage of capital cost to that estimated for the S-I plant in Reference 1. The production cost estimate for the S-I plant is given in Reference 1.
² The production cost for electrolysis is taken at the current minimum rates described in Reference 7.

³ The production cost for steam reforming is taken at the rate described in Reference 7.

⁴ Efficiency is defined to be the energy available from hydrogen produced (based on lower heating value) divided by the total of all energy input into the process (including energy used to produce electricity in the case of electrolysis).

⁵ Assumes 100% efficiency in conversion of hydrogen to energy.

VIII. Conclusion

 In the push to develop hydrogen as a long-term energy solution, many different production concepts have been developed and studied. Nuclear-based technologies are on the forefront of new hydrogen production research.

 A large amount of energy is required to produce hydrogen. For many hydrogen production processes, input energy is needed in the form of either heat or electricity. Nuclear energy has the ability to provide either form with several advantages:

- Nuclear energy is abundant and does not depend on fossil fuels.
- It is environmentally friendly, with virtually no pollution emissions.
- Nuclear reactor technology has developed to the point where very high temperatures and efficiencies are possible with advanced reactor designs.

 Three nuclear powered hydrogen production processes have been compared and evaluated.

 The basic cost estimates for future advanced electrolysis production indicate that this process, while very simple and clean, will have difficulty competing with thermochemical cycles. Electrolysis is not expected to disappear from the market, but it will likely only continue in niche markets. Electrolysis is a viable option for small production facilities where freedom of geographic location is vital. Furthermore, if electricity can be purchased at off-peak prices, then larger peak-shaving electrolysis stations may also be economically viable. Electrolysis capacity factors are flexible and can quickly change demand under power variations. This makes it uniquely positioned to provide a benefit to large electrical suppliers by balancing out transients on the grid.

 While electrolysis is much less efficient than other hydrogen production technologies, it could fill market niches and be used in areas where the transportation and storage of hydrogen is difficult.

 Steam reforming of methane is a promising thermochemical process that offers high efficiency production. Nuclear production facilities will provide even greater efficiencies, and will reduce pollution levels from those of fossil fired reformers. However, it requires a source of light hydro-carbons from fossil fuels. The

process produces CO2, a greenhouse gas as a byproduct, therefore negating some of the benefits of converting to hydrogen use as a fuel.

 The knowledge and lessons that will be learned from a successful use of the steam reforming process powered by nuclear heat will be invaluable to the success of other thermochemical processes to be combined with nuclear heat sources. The steam reforming process is making steady progress toward implementation with nuclear power. The Japan Atomic Energy Research Institute (JAERI) is currently conducting testing and research with the goal of demonstrating the hydrogen production process using nuclear heat in 2008. There is still significant research and development that must be conducted, but the outlook for steam reforming is bright.

 The Sulfur-Iodine process appears to be the best long term candidate for hydrogen production with nuclear heat. The S-I process offers higher efficiency than electrolysis without the production of carbon dioxide or the use of any fossil fuels that is required by steam reforming. While this process is still in the fundamental research stage, the S-I process could become the most attractive method for producing hydrogen in the future.

 Although successful experimentation has been conducted, there are significant technical obstacles to the design of an industrial facility capable of being linked with a nuclear reactor. For example, the highly corrosion resistant materials to build the vessels still need to be developed. Meanwhile, valuable knowledge of a nuclear/hydrogen facility will be gained by system operation using the steam reforming method. That data will be extremely useful when applied to a hydrogen production using the thermochemical S-I process.

 If this cycle can be safely coupled to a nuclear reactor, and operated at its maximum efficiency over the long term, it will surely be a welcome addition to industrial hydrogen production in the coming century.

 Overall, large-scale nuclear-based hydrogen production technologies are still in their infancy and the market for large hydrogen production facilities remains small. While it remains to be seen whether hydrogen will replace fossil fuels and electricity as our primary energy suppliers, the research and development of efficient and clean means of producing hydrogen will feed the desire to use hydrogen as the preferred energy carrier of the future.

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