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Hydrogen production using high temperature reactors: an overview

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Abstract. The present work is an attempt to provide an overview, about the status of R&D and current trends in Hydrogen Production using High Temperature Reactors. Bibliographic references from the INIS database, the Science Direct database and the NTIS database were downloaded and analyzed. Ten year data on the subject, published between 2001 and 2010, was selected for the study. Appropriate qued ry formulations on these databases, resulted in the retrieval of 621 unique bibliographic records. Using the content analysis method, all the records were analyzed. Part One of the analysis details Scientometric R&D indicators, Part Two is a subject-based analysis, grouped under: A. International Initiatives and Programmes for Hydrogen Production; B. European R&D initiatives for Hydrogen production; C. National Initiatives and Programmes for Nuclear Hydrogen Production; D. Reactor Technologies for Nuclear Hydrogen production; E. Fuel Developments; F. Hydrogen Production Processes using HTRs and G. Materials Consideration for Nuclear Hydrogen Production. The results of this analysis are summarized in the study.

Keywords: hydrogen production; high temperature reactors; content analysis; VHTR; databases

1. Introduction

High Temperature Reactor (HTR) technology has been developed from the late 1940s in US and Germany. The present Very High Temperature Reactor (VHTR), operating at >750^{0}C, has over the years, evolved into a new reactor concept, designed to be a very efficient and safe system. It is a helium-gas cooled, graphite-moderated, thermal neutron spectrum reactor, which can provide electricity and process heat for wide-ranging applications, including hydrogen production. Hydrogen production through fossil fuels entails CO_2 emissions. Therefore splitting of water to produce hydrogen is a better alternative. There are several methods to extract hydrogen from water and two of the highly used processes are 1.High Temperature Electrolysis (HTE) and 2.Thermo-chemical cycles. Both these processes require very high temperatures, which can be provided by the VHTRs. A typical nuclear hydrogen production Elder and Ray (2009) plant comprises:

1. The VHTR
2. The Hydrogen Production Plant (HPP)
3. Intermediate Heat Exchanger (IHX) and the

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4. Power Conversion System (PCS) (In the PCS, either a Rankine steam cycle or a Brayton gas turbine cycle is used).

2. Materials and method

The present study is a content analysis of publications, in the area of HTRs and Hydrogen production. Bibliographic references from the INIS Database (2001-2010), the Science Direct database (2001-2010) of Elsevier and the NTIS database, National Technical Reports Library (NTRL), (2001-2010), were downloaded and analyzed. Ten year data on the subject, published between 2001 and 2010, was selected for the study. A total of 351 records from the INIS database, 240 bibliographic records from the Science Direct database of Elsevier and 30 records from the NTIS database were retrieved for a ten year period between 2001 and 2010. After removing the duplicate records from all the three databases, a total of 621 records were selected for analysis from the three databases. 2007 was the most productive year with 108 publications, followed by the year 2008 with 106 publications. There were 91 publications in 2006. As far as the type of publications were concerned, 330 of the references, selected from the three databases were journal articles (53%); 83 were technical reports (13%) and the remaining references were reports of seminars, conferences and meetings, conducted by various national and international institutions. The unpublished scientific and technical reports are a unique feature of the INIS database. These technical reports were brought out by the Japan Atomic Energy Agency (JAEA), the Korea Atomic Energy Research Institute (KAERI) and the Idaho National Laboratory. INIS also published some of the reports under its nuclear hydrogen production programmes and activities.

3. International initiatives and programmes for hydrogen production

3.1. The IAEA

Under the programme of Non Electric Applications, Khamis I. (2011) of nuclear power reactors, IAEA has initiated several Coordinated Research Projects (CRPs) in several IAEA member countries. In 2009, a technical document was published, which gave the results of a completed CRP, on advances in nuclear power for process heat applications. A status report on hydrogen production has been completed recently and is under review. IAEA in collaboration with BARC, has released the newly developed Hydrogen Economic Evaluation Programme (HEEP) software, which can be used to perform economic analyses, related to large-scale production of Hydrogen. (A beta version of this software is available at the IAEA website. A CRP on benchmarking and validation of HEEP will be initiated by 2012. IAEA has also developed a toolkit on Hydrogen production, using nuclear energy. The International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) is also working towards meeting the energy needs of its member countries in the 21^{st} century, in a sustainable manner.

3.2. Generation IV International Forum (GIF)

The GIF was chartered in 2001, to develop the Next Generation Nuclear Power (NGNP) Vandel (2007) Systems, to meet the world’s future energy needs. Under this programme, Gen IV nuclear energy systems, deployable from 2030 onwards, will offer true potential for expanded use of nuclear energy, in particular in the field of Hydrogen production. These systems would be sustainable, economically viable, safe and reliable and proliferation-resistant. The VHTR is a Gen IV system and GIF will focus on organizing cooperation in R&D between countries, for harnessing nuclear power. In November 2006, seven GIF members signed a VHTR system arrangement, to set up a system research plan for R&D projects in VHTRs. These include: development and validation of materials to be used in VHTR systems, fuels and fuel cycle issues in VHTR systems and the use of VHTRs to produce Hydrogen.

4. European R&D initiatives for hydrogen production

4.1. The European Commission

The European Commission is funding a programme of development of HTR technologies in the EURATOM 5th Framework Programme initiated by the partners of the European HTR Technology Network (HTR-TN). About twenty organizations from industry, research and higher education of eight countries, including the leading industrial and research organizations for nuclear energy in Europe (e.g., BNFL, CEA, Cogema, EdF, Framatome ANP, Forschungszentrum Jülich (FZJ), NRG, Tractebel, etc,) and the Joint Research Centre (JRC) of the European Commission joined forces and created the (European) HTR Technology Network (HTR-TN) in 2000. The highlights of this programme and its main results, concerning the fuel technology, reactor physics, components, materials, the development of a safety approach adapted to the HTR specific safety features and the waste management issues, were published in a report in 2007 (Hittner *et al*. 2007).

The second phase of the new Integrated Project, V/HTR-IP, proposed by HTR-TN, has been selected for funding in the 6th Framework Programme of the European Commission (FP6) and started in 2005. The main objective of V/HTR-IP is to explore the potential of the HTR technology for higher performances, most particularly higher operating temperatures (in FP5, the reference was an operating temperature of 850°C, in FP6, temperatures above 900°C will be considered) and to search for innovative solutions for overcoming the possible performance limits of existing technologies. This objective is in line with the VHTR programme of the Generation IV International Forum (GIF), to which the FP6 Project, Buckthorpe *et al*. (2007) would contribute, in the frame of the EURATOM participation in GIF. Moreover, the main incentive for increasing the operating temperature is the possible use of the reactor heat generation for hydrogen production from water splitting through high temperature processes (either a thermo-chemical process or high temperature electrolysis), the efficiency of which drastically increases above 900°C. Therefore the reactor cooling system must be adapted to the needs of these processes and new components have to be developed for that purpose in the Project. The following programmes have been initiated by EU in the area of HTR development.

4.2. The Michaelangelo network

The Michaelangelo network, Verfondern *et al*. (2006), was started within the FP5 framework, with the objective to elaborate a general European R&D strategy, for further development of nuclear industry. It was involved in the initiation, preparation, conduction of various EU research projects related to nuclear technologies. One of them was the ReActor for Process Heat, hydrogen

And ELectricity Integrated Project (RAPHAEL-IP). It was launched in 2005 and ended in April 2009. Under RAPHAEL, R&D on advanced gas-cooled reactor technologies and very high temperature applications (>1000^{o}C) was conducted along with other VHT applications such as reactor physics and thermodynamics, fuels, backend, materials and components development and safety. On developments pertaining to Hydrogen production, the HYdrogen NETwork (HYNET) was created in 2002, with the principal aim of developing a European Hydrogen Fuel infrastructure. In 2003, the International Partnership for a Hydrogen Economy (IPHE) (a joint public-private enterprise) representing 15 countries and the EU was launched.

4.3. *HYdrogen Thermo-chemical Cycles* (HYTHEC)

HYdrogen Thermo-chemical Cycles (HYTHEC) Le Duigou *et al.* (2006), was another EC funded initiative for long-term massive Hydrogen production, using solar and nuclear technologies. R&D efforts in Hydrogen production included: Comparison of Sulfur-iodine and Hybrid-Sulfur cycles through flowsheets, industrial scale-up, safety and costs modeling to improve fundamental knowledge and efficiency.

4.4. OECD programmes and projects

One of the earliest experiments on HTR was conducted by OECD under the DRAGON Reactor Experiment (DRE) in 1959 UKAEA and OECD/NEA (2004). OECD encourages R&D cooperation among its member countries through information exchange. The first information exchange meeting on nuclear production of Hydrogen was held in 2000 OECD (2000). The second meeting was held in 2001, on R&D aspects of Materials and engineering developments of HTGRs OECD (2003). The Fourth NEA Information Exchange Meeting on Nuclear Production of Hydrogen was held in 2009. Several aspects of Hydrogen production such as HTE, Thermo-chemical cycles and materials development were discussed. Two recent studies were conducted by OECD. One on materials development (2010) was on the selection of a hydrogen absorbing alloy to assist HTGRs in hydrogen production. Zr(v_1_xFe_x)_2 was selected for the heat pump system, which operates between the Inlet/Outlet temperatures of HTGR and the reaction vessel of the Sulfur-iodine cycle Fukada and Nobutaka (2010). Another 2010 study OECD Nuclear Energy Agency, (2010), focused on the detection methods and system behavior assessment during the IHX tube rupture (IHXTR) scenario. Modeling of VHTR core and optimization of the VHTR core design was performed through the EVEN parity Transport code EVENT and the thermo-hydraulics code THERMIX. The study was conducted in 2005 OECD Nuclear Science Committee (2005).

5. National initiatives and programmes for nuclear hydrogen production

5.1. USA

The US Department of Energy (DOE)'s Nuclear Hydrogen Initiative (NHI) is investigating candidate technologies for large scale hydrogen production, under the New Generation Nuclear Plant (NGNP) programme. The NGNP project will demonstrate emissions-free nuclear-assisted electricity and Hydrogen production by 2017. A technical report on the status of the NGNP programme was published in 2007 Vilim (2007), which described studies on both Low and High

Temperature Electrolysis, thermo-chemical cycles and reactor technologies for Hydrogen production. Another study published in 2007 Sherman (2007), explored the heat transfer characteristics and pressure drop of the Phase Change Heat Exchanger (PCHE), with Helium as the primary coolant and Sodium as the secondary coolant. Westinghouse, General Atomics GA), AREVA NP, Idaho National Laboratory (INL), Sandia National Laboratory (SNL), Argonne National Laboratory (ANL) and Savannah Research Laboratory are working on the R&D and operational aspects of the NGNP. In 2007, ANL conducted a study on power requirements at the VHTR/THE interface, VIlim (2009) for Hydrogen production. A part of the NHI programme, the interface would connect the VHTR to the HPP. The intermediate heat transport loop comprising the interface will transfer the high temperature thermal energy from the reactor to the HPP. The INEL, Oh *et al*. (2007), is also conducting a series of R&D programmes on the design and development of heat exchangers, heat transport systems, materials safety etc. which are the main components of the heat transfer loop. The Sandia National Laboratory, Martin *et al*. (2007) developed a new tool called the MELCOR-H2, which would enhance visualization, to maximize Hydrogen and electricity generation. Through a GUI, this single tool could simulate Hydrogen production through the VHTR/HPP system. Component sizing studies and control system strategies were described in an INL report, Oh (2007), for achieving plant production and operability goals for Hydrogen production. In 2007, INEL reported a steam generator component model, Oh *et al*. (2007), which combined both Brayton and Rankine cycles. This combined cycle would be used as one of the Power Conversion Units to be coupled to a VHTR.

5.2. France

Under the AREVA group, new plant design is managed by FRAMATOME NP, Copsey, B. *et al*. (2004), which together with Electricite de France (EDF) and Centre for Atomic Energy (CEA) and other research and industry partners, is leading the European HTR development. Under the ANTARES project, an industrial HTGR prototype is being developed. R&D programmes include: the development of computer codes: CRONOS for neutronics and STAR CD for thermal hydraulics, materials development, fuel fabrication and development of key components of the nuclear hydrogen production plant.

5.3. China

The Institute of Nuclear Energy Technology (INET) developed the HTGR, HTR-10, Zongxin *et al*. (2002), which reached its first criticality in Dec. 2000. HTR-PM, Zuoyi *et al*. (2006) is another HTGR demonstration plant with pebble bed modular technology, being developed by INET. Hydrogen production through Sulfur-Iodine and High Temperature Electrolysis would be accomplished. Under this programme, several fundamental studies are being carried out. These could be categorized under the following:

1. Separation, concentration and purification of Hydrogen
2. Demonstration of the feasibility of using planar solid oxide cell (SOC) technology.
3. Analysis of the degradation mechanism of the SOC cells used in HTE
4. Development of new materials for corrosion resistance and high performance and
5. Cost of performance assessment of the HTE plant.

Three studies on reformers were reported from China; one in 2006 and two in 2007. Tsinghua University and the Inst. of Nuclear and New Energy Technology, Yin *et al.* (2006), conducted a study on the steam reformer using a one-dimension quasi-homogeneous phase dynamic model. A finned central tube improved the reformer's performance as compared to a smooth central tube. One of the studies reported in 2007, Yin *et al.* (2006), was an initial study on reformers, wherein the methane conversion rate and hydrogen output of the reformer were measured through numerical analysis, using a one-dimensional steady-state model. The second study in 2007, Yin *et al.* (2007), was based on steady-state, pseudo-homogeneous, one-dimensional model of the reformer, where the influence of the process parameters on the performance was discussed.

5.4. Japan

At present, Japan is the only country in the world, which has a working HTGR and Hydrogen production programme, Kasaharae *et al.* (2005). The HTR programme started very early in Japan around 1969, with the development of the High Temperature Engineering Test Reactor (HTTR), by the Japan Atomic Energy Agency (JAEA). The system attained criticality in 1998. In 2004, the HTTR achieved a coolant outlet temperature of 950^0C. A pilot plant test project is currently under way, to produce Hydrogen using the Sulfur-Iodine cycle. By 2020, it is planned to integrate this pilot plant with the HTTR. JAEA is also planning a 600 MW Gas-Turbine HTR, the GTHTR300C unit, for cogeneration of Hydrogen and electricity. Design and development of the GTHTR300C was initiated in 2001, Nishihara and Takeda (2005).

One of the earliest reports published by JAERI on the subject, described R&D activities aimed at operational level by 2030s. A number of R&D programmes and activities are being conducted by Japan Atomic Energy Research Institute (JAERI) now JAEA, to achieve the above targets. In 2001, Takeda and Iwatsuki (2001), JAERI published a report on counter permeation of Deuterium and Hydrogen through Inconel 600. Permeation of Hydrogen isotopes through high temperature alloys used in the IHX and the catalyst pipes is an important issue in the Hydrogen production system. In 2002, another Hydrogen permeation study, Takeda *et al.* (2002) was conducted by JAERI, wherein the permeability of the isotopes through Hastelloy XR and other high temperature alloys was quantitatively evaluated. The effect of the oxidized film on reducing Hydrogen permeability through the pipe was also evaluated. A 2003 JAERI report Nishihara *et al.* (2003), described the system layout and component design of the HTTR. A technical review was published in 2003, Ishiyama (2003), describing a conceptual multi-power conversion system, (a trigeneration process: thermal-electrical and chemical energy). Development of a High Temperature Isolation Valve (HTIV) for the HTTR Hydrogen production system was reported by JAERI in 2004, Nishihara (2004). The HTIV is installed on the hot Helium gas piping, penetrating the reactor containment vessel. The heat transfer characteristics of the steam reformer, Takeda and Ichimiya (2007) in the HTTR were examined in another 2004 JAERI study. Numerical analyses were carried out by JAERI as reported in 2005, to evaluate Hydrogen dispersion, Nishihara (2005), around the Hydrogen production facility, in case of a Hydrogen pipe rupture accident. Four reports were published by JAERI in 2006: Another Hydrogen permeation study detailed permeation through heat transfer pipes of the IHX, during the 950^oC operation of the HTTR, Nariaki, Hirofumi and Tetsuaki (2006). Heat transfer and flow characteristics of a heat exchanger tube filled with porous material were the subjects of the second report, Ichimiya *et al.* (2006). A dynamic analysis code N-HYPAC was used, to evaluate the system transient behavior of the Hydrogen production plant, which would be coupled with the HTTR, Maeda *et al.* (2005). The fourth study in 2006 reported an evaluation method to analyze the characteristics of Hydrogen gas dispersion, in case of blast overpressure, Murakami *et al.* (2006) in a VHTR. One of the first studies in 2007, reported purification methods for the Hydrogen produced through the Sulfur-Iodine process. Three purification methods were evaluated: Pressure Swing Adsorption (PSA) method, Cryogenic distillation method and Membrane separation method, Kasahara *et al.* (2007). To improve the design of the coupled plants, a Probabilistic Safety Assessment (PSA) was carried out by JAEA. Safety concerns of the Hydrogen production plant were addressed by JAERI in 2008. A spark ignition model, Kang *et al.* (2008), with selected values of pressure and volume was used, to determine the safety distance between the HTGR and the HPP. Another safety assessment of VHTR and the HPP was carried out against fire, explosion and acute toxicity, Murakami *et al.* (2008). A new concept of a decomposer was proposed, which involved the integration of the Sulfuric acid decomposer, Sulfur trioxide decomposer and the process heat exchanger, Kanagawa *et al.* (2007).

5.5. Korea

Korea has a programme for large-scale hydrogen production using VHTRs in 2020s as part of its long term energy policies. Towards this end, an operating license for a Nuclear Hydrogen Development and Demonstration (NHDD) reactor has been planned. It is a 200 MWth VHTR reactor concept, operating at 950^0C and using the Sulfur-Iodine thermo-chemical cycle. Current R&D efforts are in the areas of: development of design tools, assessment of high temperature anti-corrosion materials and components, development of a small-scale gas loop, a bench-scale TRISO fuel fabrication facility, a bench-scale Sulfur-Iodine thermo-chemical cycle closed loop and high pressure sections of the Sulfur-Iodine thermo-chemical process. In 2004, KAERI launched a nuclear hydrogen programme in collaboration with Korea Institute of Energy Research (KIER) and the Korea Institute for Science and Technology (KIST), Lee (2009). KAERI has R&D cooperation with JAEA in Japan, GA in US and INET in China for collaborative HTR research. In 2002, KAERI published a report on enthalpy analysis of Hydrogen production processes, Jeong *et al.* (2002). Under the high temperature and high pressure gas loop, a project was carried out by KAERI in 2005: a ceramic heat exchanger was developed, Hong *et al.* (2005), and corrosion resistance of the heat exchanger was increased, by ceramic coating on a super alloy plate. In 2006, KAERI developed a plate-fin type, Kim *et al.* (2006), sulfur trioxide decomposer for the nuclear hydrogen production system. The plate of Ni-based material was coated with SiC to enhance its corrosion resistance. It also carried out a CFD assessment of a compact high temperature heat exchanger, Kim *et al.* (2006). Four studies were reported by KAERI in 2007. The first one was on the NHDD plant designs, Chang *et al.* (2007), the second one was on the conceptual design of the main components of the NHDD such as the IHX and the Hot Gas Duct (HGD), Song *et al.* (2007).

Design activities focused on identification of design code criteria, material selection and selection of the type of IHX. The third study focused on the effect of catalyst on the thermal design of a lab-scale sulfur trioxide decomposer, Kim *et al.* (2007), and the fourth study of 2007 was a report of the VHTR R&D Steering Committee, Lee *et al.* (2007). In 2009, two studies were reported by KAERI on the decomposer. One was a simulation study of the decomposer using a chemical process simulator, Seo *et al.* (2009), and the other presented the Hydrogen production rates using the decomposer, Kim *et al.* (2009).

5.6. South Africa

The South African Dept. of Science & Technology has created competence centres including one on Hydrogen Infrastructure called the Hydrogen Infrastructure Competence Centre (HICC). It is hosted by the NW University and CSIR. The HICC will look into developing Hydrogen production, storage and distribution as well as developing standards and codes. The Pebble Bed Modular Reactor (PBMR) was developed in 1999, to be used for a number of process heat applications including Hydrogen production. Two PBMR process heat applications have been configured. The first one in the intermediate temperature range (750^{0}C) produces helium for high pressure steam production and cogeneration. The second one in the high temperature range of 950^{0}C) will be used for HTSE and thermo-chemical hydrogen production. In 2007, a status paper on thermo-chemical water splitting using nuclear heat was published, Ravenswaay *et al*. (2009).

The ENEA in Italy has recently launched a project on nuclear hydrogen production, using the Sulfur-Iodine thermo-chemical cycle. The Instituto National de Investigaciones Nucleares in Mexico has also planned for a nuclear hydrogen production plant at the Laguna Verde site. Evaluation of capital and operating costs as well as preliminary risk analysis of a hydrogen production plant (using the methane reforming process) coupled to a high temperature nuclear reactor was reported in a 2003 publication. In 2005, a HAZOP study was carried out, Flores *et al.* (2005), with the purpose of obtaining initiator events for the probabilistic safety analysis of a Hydrogen production plant. The fault-tree event analysis was done using SAPHIRE programme.

6. Reactor technologies for nuclear hydrogen production

6.1. Japan

Japan and JAERI/JAEA have been at the forefront of R&D in reactor technology. Some of the early reports published in 2002, Ogawa and Shiozawa (2002), and 2001, Shusaku *et al*. (2001), by JAERI, detail the status of R&D in the Hydrogen production and evaluation of the HTR system. A demonstration test of Hydrogen production using HTGE as well as a simulation test on normal startup and shutdown of HTTR was reported in 2004, Ohashi *et al*. (2004). The deployment of GTHTR300C cogeneration system was also a highlight of 2004, Kazuhiko and Xing (2004). In 2005, JAERI reported the achievement of a coolant temperature of 950^{0}C, for the first time in the world, Kawasaki *et al*. (2005). The HTTR, which is the first HTGR in Japan, started operation in 2004. The high temperature Helium gas with a temperature of 950^{0}C was extracted from the reactor. JAEA also experimented with small-sized cogeneration HTGRs, for developing countries, where sufficient infrastructural facilities for power grids were not available. In this context, the High Temperature Reactor 50 Cogeneration (HTR 50C) was analyzed for its economic viability in 2008, Sakaba *et al*. (2008). Another 2008 study described the nuclear design of the reactor core for the GTHTR300C system. The microindentation method was used, to detect residual stresses during the degradation of graphite components, Taiju (2008). JAEA reported a series of preliminary tests and studies conducted to develop commercial HTGRs such as safety demonstration tests, nuclear heat supply characteristics tests, burned core tests, reactivity insertion tests, fuel failure tests, annular core tests and tritium measurement tests.

6.2. In Korea

In Korea the first phase of safety demonstration tests for HTTR was reported by KAERI in 2003, Shin *et al*. (2003). KAERI conducted a preliminary study on coupling options for the HTGR and the HPP. Six design options were considered and overall plant efficiencies were evaluated in 2004, Kim (2004). Experimental data for the core design of the VHTR was published by KAERI in 2004, Park *et al*. (2004). In 2005, in collaboration with INET, Monte Carlo benchmark calculations HTR-10, Kim *et al*. (2005). initial core were performed. The significance of graphite in VHTR systems was substantiated with two studies reported by KAERI. The first one in 2005, Kim *et al*. (2005), gave benchmark calculations for graphite-moderated reactors using different evaluated nuclear data. The second in 2006, Chi and Kim (2006), was on evaluation of differences in the oxidation behavior of two nuclear graphite grades of different fillers. The NHDD project will also be using graphite as moderator and reflector. To evaluate the technical feasibility of the NHDD system, a methodology was devised, to determine the optimum VHTR design and also analyze the various Hydrogen production methods. Tritium diffusion was estimated both in the pebble-bed type and the prismatic type gas cooled reactor core in a study published in 2006, Lee *et al*. (2006) by KAIST. A series of Monte Carlo benchmark calculations for Hydrogen production using VHTR and criticality calculations for a 400MWth PBMR core (using the MCNP5 code) were performed by KAERI in 2006, Kim H.C. et al. (2007). KAERI also experimented with a cooled reactor design as an option for the Reactor Pressure Vessel (RPV) design of the NHDD plant. Several HTGRs were surveyed and their design characteristics were evaluated. CFD analysis was used for the evaluation. Various design codes for VHTR meant for high temperature analysis were studied: the ASME code, French RCC-MR, UKR5 and Japanese codes were investigated, Kim *et al*. (2006). Safety characteristics of the circular and annular pebble cores for a prototype VHTR, Lim *et al.* (2007), for Hydrogen production were determined in a 2007 study. Another 2007 study by KAERI, detailed the thermo-hydraulic optimization of a VHTR block type core. Design modifications to reduce the reactor vessel operating temperature without compromising on the passive safety features of the system were described, Matts *et al.* (2007). Two more reported studies by KAERI in 2007. One was on the preliminary core design analysis of a 200MWth pebble bed type VHTR, Jo *et al*. (2007), and the other was on analysis of source term evaluation method for tritium behavior mechanisms, Lee (2005). In collaboration with Seoul National University, KAERI published a report in 2008, on thermo-fluid investigation on a double-sided cooled annular fuel for the prismatic VHTR. In this study, a double-sided cooled annular fuel concept was proposed, Nam IlT, (2008), to overcome the issue of high fuel temperature in a VHTR. A detailed thermo-fluid analysis using a CFD code was carried out to investigate the thermo-fluid performance of the proposed fuel design. It also reported the development of a new computer code system, for the analysis of the VHTR cores (both prismatic and pebble-bed types), based on the existing HELIOS/MASTER code system. KAIST in collaboration with INL conducted a study on predicting the graphite density changes and mechanical strength variations of VHTR during an air-ingress accident. The GAMMA thermal-hydraulic analysis system code was used, No *et al*. (2009).

Decay heat analysis for a 200MWth prismatic VHTR core was done, using the Monte Carlo Depletion calculation in a 2009, Lee *et al*. (2009) KAERI study. A VHTR simulated experimental helium loop is being constructed at KAERI. A 2009 KAERI report, gave results of a CFD analysis of a very high temperature helium heater, which is an important component of the helium loop. Six potential materials (B, Gd, Er,Eu,Sm and Dy) were evaluated as burnable poisons for reactivity management in a VHTR, Yoon *et al*. (2009). A Physics study was performed by KAERI in 2009, to find the optimal application of a burnable poison for an excess reactivity management in a 600MWth block-type VHTR. A Probabilistic Safety Assessment (PSA) study was conducted by the Seoul National University, for a VHTR, using statistical modeling, to test the passive safety reliability of the system, Tae and Un (2009). Another study by KAERI in 2007 highlighted the importance of safety in a VHTR. Multi-states criteria were used, to determine the safety of the Reactor Cavity Cooling System (RCCS), Lee *et al*. (2006). Several R&D papers were published by KAERI in 2010. Decay heat analysis of VHTR cores using the Monte Carlo depletion calculation; Use of the GAMMA+ system analysis code to identify and quantify key parameters affecting the predictions of fission product transport and plate-out behavior in the coolant circuits of a VHTR, Jun *et al.* (2010); Use of a thermo-fluid code and a commercial CFD code during normal and accident operations, to assess the performance of the vessel cooling options. Three types of vessel cooling options were considered for the design of the RPV, Min *et al*. (2010), which is an important issue of VHTR systems, due to their high operating temperatures.

6.3. In USA

In USA the Modular Helium Reactor (which is an HTGR) is in operation at a power level of 600MW(e). Hydrogen production using MHR is referred to as the H2-MHR concept. Two programmes are under way to produce Hydrogen. The first programme uses the Sulfur-Iodine thermo-chemical process and is known as SI-based H2-MHR and the second programme employs High Temperature Electrolysis for Hydrogen production and is known as HTE-based H2-MHR. A status report was published by General Atomics in 2006, Richards (2006). The Idaho National Laboratory and the Idaho National Engineering Laboratory published a series of reports between 2006 and 2008 for nuclear hydrogen production. Three advanced reactor power cycle combinations were studied:

1. Helium cooled reactor coupled to a direct Brayton power cycle
2. Supercritical CO_2 cooled reactor coupled to a direct recompression cycle
3. Sodium cooled fast reactor coupled to a Rankine cycle.

In 2007, Vilim (2007), ANL described the results of preliminary plant control and stability studies for assessing the operability of a VHTR coupled to an HTSE plant. A Tritium Permeation Analysis Code (TPAC) was developed by INL using the MATLAB SIMULINK package, for analysis of tritium behaviors in the VHTR/HPP system, Kim *et al.* (2010). The USNUREG and ORNL published a report, Morris *et al*. (2008) on Fission Product Transport (FPT) in the HTGR. Three models of operation were examined; Normal operation, anticipated transients and postulated accidents.

7. Fuel developments

TRISO (Triisotropic) coated particle fuels are the fuels of choice for nuclear hydrogen production. These particles can withstand the high temperature reactor environments, due to their three isotropic ceramic layer coatings: the middle SiC layer enclosed by two pyrocarbon layers. One of the first papers on TRISO particles was published by INEEL, describing key differences in the fabrication, irradiation and high-temperature accident testing of US and German TRISO coated particle fuel and their implications on fuel performance.

An excellent review of TRISO-coated particle nuclear fuel performance models was published from China in 2006, Bing *et al*. (2006). Evaluation of the ceramic layer thickness and of the degree of sphericity of these fuel particles is necessary during the fabrication process. In 2007, the IN2P3/CNRS reported the immobilization of inert TRISO coated fuel in glass prior to geologic disposal, Abdesselam *et al*. (2006). Another study on waste management published in 2007, reported the behavior of HTR fuel elements in aquatic phases of repository host rock formations, Fachinger *et al*. (2006). CEA/AREVA NP conducted a HTR fuel development and qualification programme for nuclear fuel fabrication technology. A report was published in 2006, Greneche and Szymczak (2006), which detailed the fuel concept selected for the ANTARES project. ORNL described a new method to fabricate fuel compacts for the NGNP programme which was published in 2008, Pappano *et al*. (2008). JAERI experimented with the use of Zirconium Carbide instead of SiC for the coatings, Park *et al*. (2006). Korea published a status report on HTGR fuel technology. The IN2P3/CNRS published a report on HTR fuel waste management, Fabrice *et al*. (2010). In this study, two methods were described to separate the fuel from irradiated graphite.

8. Hydrogen production processes using HTRs

8.1. Thermo-chemical Cycles

A series of chemical reactions are used, to split water into Hydrogen and Oxygen in various thermo-chemical cycles.

8.1.1. The Sulfur-Iodine (SI) Cycle

The Sulfur-Iodine (SI) Cycle was developed by General Atomic during the 1970s. It consists of three sections

- Bunsen section
- Sulphuric acid section
- Hydroidic Acid (HI) Section.

Japan, USA and the EU under Framework 6 (under the HYTHEC programme) are all involved in the R&D of the SI cycle. R&D work on refractory and corrosion-resistant pressure sensor, Ishiyama (2003), was reported by JAERI/JAEA in 2003. This sensor with Tantalum/SUS welding type diaphragm was developed for the HI cycling test in the HPP and was tested for its fatigue and corrosion properties. Another report from JAERI in 2003, related to the Bunsen Section of the SI cycle. The excess Iodine present in this section, needs to

be separated from the acids which are the intermediate products. This separation of Iodine and the acids was achieved through a membrane reactor, Kasahara S. et al. (2003) system. A pilot test plan was detailed for the IS process in 2004, Kubo S. et al. (2004). A sintering technique was used to develop a full scale ceramic block unit with heat channels for the Sulphuric acid vaporizer, Ishiyama (2004). These ceramic blocks (Rs-SiC) with very high tensile strength showed high leak-tight behavior against the pressurized Helium gas. Six reports were published by JAERI/JAEA in 2007. The first was an evaluation study on tritium activity concentration, Ohashi H.

et al. (2007). This was done to check the permeability of tritium through heat transfer tubes and to check deuterium and tritium concentrations in the secondary helium coolant. The second report was on the measurement of vapor-liquid equilibrium of the HI acid, Hodotsuka *et al.* (2007). It was measured with an equilibrium still, made of corrosion-resistant metal. The third report dealt with the development of hydraulic analysis code for optimization of the IS process, especially in the Bunsen Section, Terada *et al*. (2007). The fourth report was on the SO_3 decomposer. In the Sulphuric acid Section, H2SO4 decomposes first into SO_3 and then into SO_2. The report described the proposed structure, results of thermal-hydraulic and mechanical strength analysis, the mockup model test fabrication and the catalyst tests for the SO_3 decomposer, Kanagawa (2007). (Catalysts are used during the decomposition of SO_3 to SO_2). The fifth report was on the construction of H_2SO_4 loop. For the H_2SO_4 decomposer, the H_2SO_4 Flow Test Loop (SFTL) was constructed to obtain flow boiling characteristics and to confirm the applicability of the components such as pipelines, pumps and instrumentation, Noguchi *et al.* (2007). The sixth report highlighted a new concept of the decomposer, Kanagawa (2007), which featured a counter-flow-type heat exchanger. This exchanger made of SiC ceramics showed good resistance to severe corrosion conditions in the decomposer. A 2008 study focused on the thermo-mechanical simulation of the SiC blocks. This was done using a general purpose FEM code, Ishikura *et al*. (2008). A new mixer-settler type of Bunsen reactor and combined H_2SO_4 decomposer was developed, with enhanced performance and reduced costs, Sakaba N. et al. (2008). The structural design concept was described. A preliminary safety analysis was carried out for the SI cycle in a 2008 JAEA report, Somolova *et al*. (2008). A small break in the Hydrogen pipeline was analyzed. Another 2009 study dealt with numerical evaluation of the flow rate of the undecomposed HI and the Iodine product at the outlet of the decomposer, Ohashi *et al*. (2009).

The KAERI in 2007 published a report on the stress fields for a coated surface of an SO_3 decomposer, Kim *et al*. (2007). Another report of 2007 by KAERI presented a visualization programme for a distillation column for VHTR-assisted hydrogen production. A computer programme was developed to visualize dynamic simulation results from a multi-stage distillation column to concentrate the Sulphuric acid as it is one of the Sulphuric acid concentrators. This column is common to both SI and the Hybrid-Sulfur cycles and computer simulation would assist in the design requirements and optimum operation of the column, Jang *et al*. (2007). In 2008, a small-scale test loop was developed by KAERI, to test the integrity of the SO3 decomposer under high temperature and pressure operating conditions, Kim *et al*. (2008). The KAERI Dynamic Simulation Code (KAERI DySCo) reported in 2009 an integrated application software that simulated the dynamic behavior of the VHTR-SI process, Chang *et al*. (2009). In 2009, the Aspen Plus chemical process simulator was used by KAERI, to model a high temperature Sulphuric acid loop, Seo *et al*. (2009).

SNL developed a recuperative bayonet Sulphuric acid decomposition reactor, wherein the heated parts were made of SiC, Gorensek and Edwards (2010). CEA published a report on the SI cycle, COE-INES (2006). It also proposed a new SI process flowsheet for large scale Hydrogen production using the ProSim code. The data and results of this simulation would be crucial for the VHTR/HPP coupling, Cerri *et al*. (2010).

8.1.2. The Hybrid Sulphur (HyS) cycle or Westinghouse cycle

The Hybrid Sulphur (HyS) cycle or Westinghouse cycle, uses both electrolysis and thermo-chemical processes for Hydrogen production. It consists of two steps

- Electrolysis of water and sulphur dioxide
- Decomposition of sulphuric acid.
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SNL, ORNL, KAERI, CEA, EU under Framework 7, Canada and the Savannah River National Laboratory are investigating various aspects of the HyS cycle.

8.1.3. The Hybrid Copper Chloride cycle multi-stage thermo-chemical cycle

The hybrid copper chloride cycle is being developed at ANL. One of the first studies of the hybrid copper chloride cycle was its comparison with the SI cycle, Wang *et al*. (2010). Heat quantity, heat grade, thermal efficiency and Hydrogen production costs were compared for both the cycles. It was found, that the hybrid copper chloride cycle is more advantageous because of its lower maximum temperature. Several studies were reported for the hydrolysis of Copper Chloride; design and reliability assessment of the control system for the hybrid copper chloride cycle; heat recovery from molten CuCl at various points within the cycle; the Oxygen production step; Cost-benefit analysis of CuCl Hydrogen production.

8.2. High Temperature Electrolysis (HTE) or High Temperature Steam Electrolysis (HTSE)

High temperature electrolysis (HTE) (800^0C to 1000^0C) is based on the technology of solid oxide fuel cells (SOFCs). The INL, Oh *et al*. (2007), Oh and Kim (2009) and JAERI are conducting extensive studies on HTSE. The INL conducted sensitivity studies of advanced reactors coupled to an HTSE HPP; Economic advantages of using the HTSE process; thermodynamic analysis of the efficiency of the HTSE; technological and scale up assessment for large scale HTSE Hydrogen (through the Integrated Lab Scale (ILS) facility) and Syngas production.

9. Materials consideration for nuclear hydrogen production

R & D on the thermo-chemical iodine-sulfur process for large-scale hydrogen production is crucial, since the IS process uses strong acids such as sulfuric acid. Equipment such as pipes, pumps and instrumentation are exposed to corrosive environments, and therefore, flow tests of concentrated sulfuric acid, Noguchi *et al*. (2010), have to be carried out using a test apparatus made of candidate materials. JAERI demonstrated that Glass lining pipes and PTFE gaskets used for high-temperature service were sound during the test term, whereas high-silicon-containing austenitic stainless-steel pipes suffered general corrosion. The corrosion properties of 18 commercial materials were evaluated in boiling Sulfuric acid by JAERI in 2004, Hong *et al.* (2004). Several alloys and high temperature metallic materials were tested for their mechanical properties. Superalloys Alloy 617 and Haynes 230, Kim *et al.* (2010), Cabet and Rouillard (2009), were tested for embrittlement in an impure Helium environment and for high temperature aging. Alloy 617 has been selected for the IHX of the HPP. High temperature oxidation and quench behavior of Zircaloy 4 and E110 cladding alloys were examined by a joint study conducted by KIT, PSI, KFKI and IBRAE, Steinbruck *et al.* (2008). Laser brazing was used for the production of high temperature resistant joints in ceramic materials (Al_2O_3 and ZrO_2). In Europe, materials studies were conducted within the Strategic Research Agenda of the Sustainable Nuclear Energy Technology Platform, Fazio *et al*. (2009). Under this platform, R&D on structural materials for Gen IV reactors is being carried out. KAERI has also developed the high temperature Sulfuric acid loop, for testing of components under its NHDD programme. Corrosion and mechanical properties of the Process Heat Exchanger, one of the important components were evaluated, Chang *et al*. (2006). A multiscale modeling methodology was developed for modeling of advanced structural materials for Gen IV reactors. The Institute of Energy at Petten is also investigating construction materials for Hydrogen production, Kosmidou and Haehner (2009). Materials aspects are also important for Hydrogen storage. Mg-Ni based alloys, Mg-Al alloys and surface-modified advanced alloys were investigated for separation, purification and storage of Hydrogen.

10. Conclusions

R&D on Nuclear Hydrogen production is well under way. Several countries particularly Japan, Korea, US and China are leading in every aspect of R&D. Japan has already demonstrated the feasibility of large scale nuclear hydrogen production. R&D on the interface between the VHTR and the HPP is also a major problem area. International programmes and collaborations are further strengthening R&D linkages. R&D on material aspects needs to be taken up more vigorously. These developments would soon lead the way to large scale nuclear hydrogen production, which would soon become the green global alternative as the fuel of the future.

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