ASPECTS OF HYDROGEN CO-GENERATION USING A THERMOCHEMICAL CYCLE LINKED TO A SUPERCRITICAL WATER-COOLED NUCLEAR REACTOR

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Abstract

Various methods for large-scale hydrogen production are currently being researched worldwide. Thermochemical cycles such as the copper-chlorine (Cu-Cl) cycle operate at high temperatures to decompose water into hydrogen and oxygen through recycled intermediate compounds. Generation IV nuclear reactor concepts, specifically SuperCritical Water-cooled nuclear Reactors (SCWRs), can be used to supply process-heat requirements for the Cu-Cl cycle. The coolant used in SCWRs is light water at supercritical conditions with pressures of approximately 25 MPa and a reactor outlet temperature of 625°C. A fraction of coolant can be diverted to heat exchangers located at select locations in the reactor loop to provide the required high-temperature heat to the thermochemical cycle for hydrogen co-generation. This paper discusses some aspects of the Cu-Cl cycle along with the development of a link to a SCWR.

1. Introduction

Research into the production of hydrogen gas for commercial applications has increased significantly over the past decade due to attempts to reduce global reliance on fossil fuels. Carbon based fuels emit greenhouse gases when burned and contribute to global warming. Hydrogen has been identified as one of the world's next generation energy carriers to be used extensively in the automobile industry. Widespread use of hydrogen has already been adopted within the petroleum, chemical, plastics and food industries. Approximately 45−50 Mt of hydrogen is currently produced annually worldwide. However, this is significantly less than the United States of America Department of Energy (DOE) predictions stating that the annual US demand will reach 65 Mt for hydrogen fuel-cell powered automobiles by the year 2040 [1]. Both non-fossil based hydrogen production and reliable storage technologies must be actively pursued in order for a hydrogen economy to be achieved.

2. Description of the copper-chlorine cycle

The copper-chlorine (Cu-Cl) cycle is one of several thermochemical cycles that have been considered for the production of hydrogen. It is particularly desirable for its relatively low reaction temperature requirements (~530°C), which can be supplied by a SuperCritical Water-cooled nuclear Reactor (SCWR). The cycle involves several reaction steps, which ultimately result in the decomposition of water into its constituent substances: hydrogen and oxygen gases.

Additional advantages of this cycle include high hydrogen yields and favourable kinetics for the oxygen and hydrogen generation reactions [2]. Within this cycle intermediate copper and chloride compounds are recycled such that process inputs are water, thermal energy and electricity only. The two variations of the Cu-Cl cycle are the 5 and 4 step cycles. Reaction steps are shown in Table 1.

Table 1. Reaction steps for the Cu-Cl cycle. [2]

Step	Reaction	Temp. Range (°C)	Feed/Output	
1	$2Cu\ (s) + 2HCl\ (g) \rightarrow 2CuCl\ (l) + H_2\ (g)$	430-475	Feed:	Electrolytic Cu+ dry HCl+ *Q*
			Output:	H_2 + CuCl(l) salt
2	$2CuCl\ (s) \rightarrow 2CuCl\ (aq) \rightarrow CuCl_2\ (aq) + Cu\ (s)$	Ambient (electrolysis)	Feed:	Powder/granular CuCl and HCl + *V*
			Output:	Electrolytic Cu and slurry containing HCl and $CuCl_2$
3	$CuCl_2(aq) \rightarrow CuCl_2(s)$	<100	Feed:	Slurry containing HCl and $CuCl_2$ + *Q*
			Output:	Powder/granular $CuCl_2$ + H_2O/HCl vapours
4	$2CuCl_2(s) + H_2O(g) \rightarrow CuO{*}CuCl_2\ (s) + 2HCl\ (g)$	400	Feed:	Powder/granular $CuCl_2$ +$H_2O(g)$ + *Q*
			Output:	Powder/granular $CuO{*}CuCl_2$ + 2HCl(g)
5	$CuO{*}CuCl_2\ (s) \rightarrow 2CuCl\ (l) + 1/2O_2\ (g)$	500	Feed:	Powder/granular $CuO{*}CuCl_2$ (s) + *Q*
			Output:	Molten CuCl salt + oxygen
Q, thermal energy; *V,* electrical energy Alternative 4-step primary reaction: $2CuCl(aq) + 2HCl(aq) \rightarrow H_2(g) + 2CuCl_2(aq)$				

In the five-step cycle, shown in Figure 1, solid copper reacts with high-temperature hydrogen chloride gas resulting in the production of hydrogen gas and liquid cuprous chloride. Although the reaction is exothermic, reactants must be initially heated to the threshold temperature of approximately 475°C. The combination of exothermic and endothermic reactions in the cycle allows for heat recycling within the system. In step 3 of the cycle, solid cupric chloride may be obtained from the drying of a slurry or solution of $HCl/CuCl_2$ in preparation of the hydrolysis reaction. Naterer et al. [2] determined that drying a solution rather than a slurry precipitate would be the most heat-intensive step in the cycle increasing the overall heat requirement of the facility by a factor of 2.5. With a slurry drying process the overall heat requirements of the cycle (endothermic reactions, heating, and drying) are 277.4 kJ per gram of H_2 produced while the heat released (exothermic reaction, cooling, and solidification) is 116 kJ per gram of H_2 [2]. It is desirable to maximize the amount of heat recycled within the system to minimize external-heat requirements. A fraction of the heat produced within the cycle is considered to be low grade, such as low-temperature water or solid powders. This factor, along with the associated costs may limit full-scale development of Cu-Cl cycle facilities [3].

A modified 4-step process combines steps 1 and 2, thereby eliminating the need for solid copper and other solids handling within the system, thus simplifying the process. The resulting electrochemical reaction, however, is not exothermic. Thus, while this configuration removes solids handling it also removes an internal heat energy source, requiring increased external heat demand. This cycle has been investigated extensively by Argonne National Laboratory (ANL) and termed a 3-step cycle as some sources do not recognize the drying of $CuCl_2$ as a reaction step [4]. From an external-heat source perspective, both cycles require high-temperature heat at the same locations within the system: the hydrolysis and decomposition reactors. The oxygen-production reactor must be very large to allow copper oxychloride ($CuO*CuCl_2$) to decompose into molten cuprous chloride and oxygen gas. Heat energy must be distributed evenly to ensure this reaction proceeds to completion.

Hydrogen production by SCWR-based electrolysis has been measured at efficiencies of approximately 30%; simulated efficiencies for thermochemical hydrogen production via a SCWR linked Cu-Cl cycle could reach as high as 54% [5].

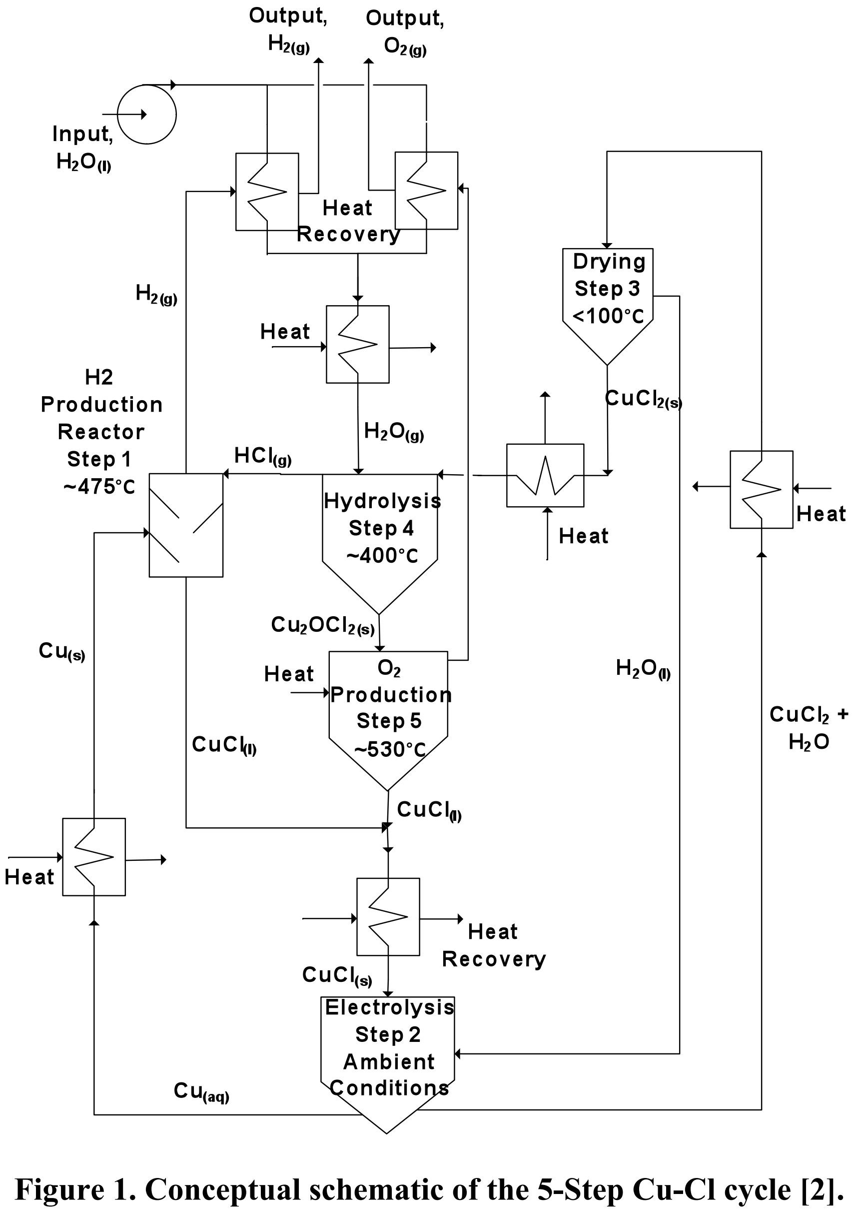

Figure 1. Conceptual schematic of the 5-Step Cu-Cl cycle [2].

3. Select alternative hydrogen-production methods

Hydrogen is the most abundant element in the universe. However, it is not readily accessible on Earth and must be produced for commercial and industrial applications. As the demand for hydrogen increases, reliable and efficient production processes need to be developed in order to achieve widespread adoption of the fuel. Currently, the most popular and cheapest method of hydrogen production is steam reforming of fossil fuels (e.g., Steam Methane Reforming (SMR)) accounting for approximately 50% of world hydrogen production [6]. Jones and Thomas [7] quote the fraction as high as 90% of the world's supply. If hydrogen is to become a sustainable energy source the reliance on fossil fuels for production must be significantly reduced.

Gasification and SMR are the most common fossil-fuel-based hydrogen production methods in use today. Gasification involves the net-exothermic reaction of carbon-based materials such as coal, methane or other petrochemical by-products with steam and oxygen under reducing conditions. Required reaction-chamber operating conditions of gasifiers are on the order of 1,250-1,575°C and 2 MPa [7]. The resulting products, carbon monoxide and hydrogen gases, generically known as synthetic (syn) gas are separated for various applications. The H_2/CO monoxide ratio of the product varies depending on the gasifier type, the oxygen concentration, reactant feed rate, and the carbon feedstock composition; for example, natural gas has a H_2/CO ratio of 1.75 whereas coal has a ratio of 0.80 [7]. The CO gas component of syngas can be further reacted with steam at high temperatures under the water gas shift reaction to generate more hydrogen gas and carbon dioxide.

In SMR, fossil fuels such as methane gas are reacted with steam over a nickel-based catalyst at high temperatures producing syngas. The reaction involving methane is endothermic requiring 252 kJ per mole of methane at standard temperature-pressure (STP) conditions. Addition of oxygen into this reaction creates an autothermal reformer, where the exothermic methane/oxygen reaction, known as a partial oxidation reaction, assists in providing heat for the primary reaction [1].

Nuclear-based hydrogen production may also be achieved through water electrolysis or steam electrolysis, which requires a combination of high-temperature and electrical-energy input. The latter process involves the passing of steam from a Nuclear Power Plant (NPP) over a solid-oxide electrolyte. Efficiencies for High-Temperature Electrolysis (HTE) can reach as high as 50−60% [7] due to the lower electrical overpotentials required, improved gas diffusivity and the thermal energy by-product [8]. Ryland et al. [8] investigated linkage of the Advanced CANDU (CANada Deuterium Uranium) Reactor (ACR-1000) developed by Atomic Energy of Canada Limited (AECL) to an HTE facility, which generated efficiencies of approximately 35%.

The sulphur-iodine thermochemical cycle is a 3-step process, which has been widely investigated with a laboratory-scale test loop in place in Japan and operated by the Japan Atomic Energy Research Institute (JAERI). Hydrogen output for this facility has been recorded up to 30 L/h [7]. The process involves the decomposition of sulfuric acid at temperatures above 800°C, processing of intermediate liquid and gas materials and further decomposition of hydrogen iodide to produce hydrogen. Efficiencies as high as 50% have been predicted for this cycle [7]. The heat requirements of this cycle can only be met by a select number of technologies including the modular helium reactor which is characterized by reactor outlet temperatures up to 850°C [10].

As part of the Nuclear Hydrogen Initiative (NHI) Program from the U.S. DOEs Office of Nuclear Energy several thermochemical cycles have been evaluated considering factors such as chemical viability (no significant competing reactions/high yields), engineering feasibility (simulated operation), and efficiency: cerium-chlorine (Ce-Cl), copper chlorine (Cu-Cl), iron-chlorine (Fe-Cl), vanadium-chlorine (V-Cl), copper sulphate (Cu-SO_4), magnesium-iodine (Mg-I), hybrid chlorine, and a metal alloy cycle potassium-bismuth (K-Bi) [9]. The majority of these cycles are characterized by low efficiencies, undesirable by-products, poor chemical kinetics or high-temperature requirements. Results of the NHI program determined that the Cu-Cl cycle to be the prime cycle to pursue further development.

4. Use of an SCWR to supply process heat for a hydrogen-production facility

The thermal requirements of the Cu-Cl cycle can be satisfied by the heat output from a SCWR. Current Generation IV designs are capable of reaching core outlet temperatures in excess of the requirements of the Cu-Cl cycle. The coolant outlet temperature of the SuperCritical Water-cooled CANDU reactor is 625°C. This is significantly greater than the oxygen production reaction in the Cu-Cl cycle which requires temperatures of ~530°C.

The connection between a SCWR plant and hydrogen-production facility is established by a Heat Exchanger (HE). A HE transfers energy from high-pressure, high-temperature SCWR coolant to the low-pressure, high-temperature steam on the hydrogen production loop. Naidin et al. [11] investigated several reactor-loop layouts, which could be integrated into a SCWR co-generation NPP. Extraction points on the reactor loop would be selected based on the reactor-design layout.

Several process heating layouts have been investigated including direct and indirect cycles involving no reheat and single reheat loops. The direct-steam single-reheat cycle has been identified as the optimal layout for a SCWR NPP given its high-gross efficiency and similarities to current fossil-fuelled power plants [12]. The HE(s) will be designed for supercritical water/superheated steam conditions or superheated/superheated streams, depending upon the location within the reactor side loop. Double-tube, recuperator-type HEs are the primary consideration for this application due to large differences in operating pressure between the SCWR NPP and the hydrogen-production facility. The supercritical-water coolant would flow through the inner tube while the low-pressure hydrogen-side steam would pass through the annulus. Thus, material strength limitations are an important consideration in the selection of location for a HE on the reactor-side loop. The HE(s) in question would need to be located within the NPP's containment structure.

The high-quality heat source at the reactor outlet is the first candidate extraction point and is common to all SCWR NPP layouts. Shown in Figures 1 and 2 the reactor-outlet conditions approximate the inlet conditions for the HE, which has coolant at 25 MPa and 625°C. This HE would operate at supercritical water conditions on the reactor side and superheated steam conditions on the hydrogen-production facility loop. Mokry et al. [13] have investigated the heat transfer coefficient of supercritical-water flowing upward in a vertical bare tube at conditions similar to those of the reactor outlet HE.

The no-reheat cycle presents a second candidate location for a HE immediately following the High Pressure (HP) turbine outlet. Coolant transition from supercritical conditions to superheated steam occurs as it passes through the HP turbine. The lower coolant pressure is favourable as this reduces material stress concerns for a HE. For the no-reheat option, a third potential HE location would be along the coolant extraction line from the HP turbine. For this position, the mass flow rate would be relatively low and potentially inadequate to supply both needs of the NPP and the hydrogen-production facility.

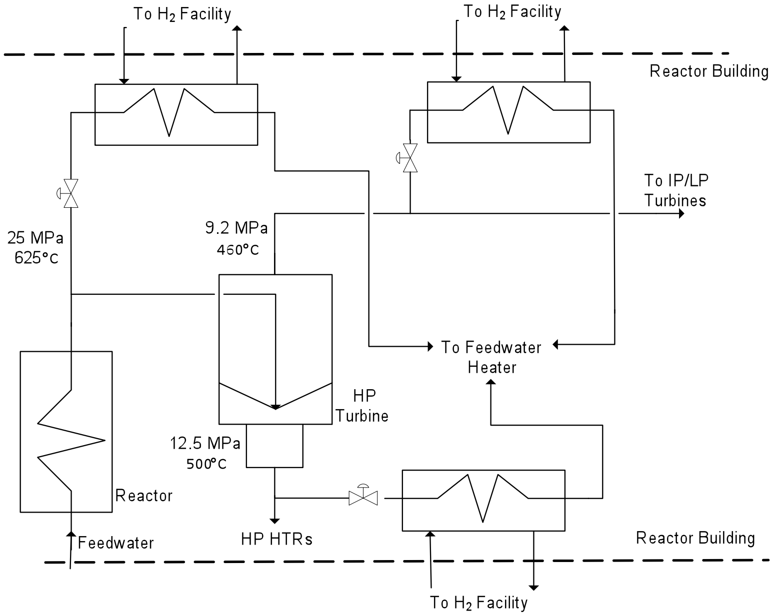

Figure 2. Heat-exchanger locations linked to H_2 production facility with a proposed no-reheat SCWR layout [14].

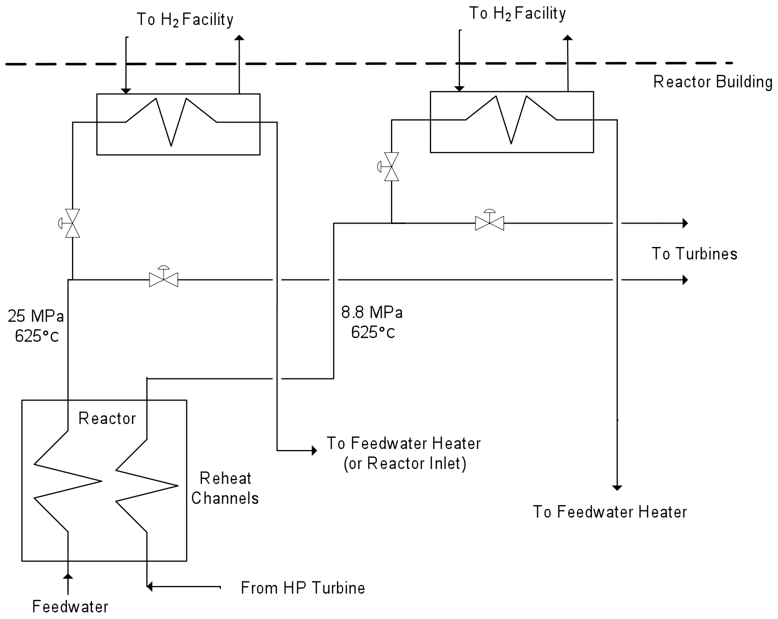

Figure 3. Heat exchanger locations linked to H_2 production facility with proposed single-reheat SCWR layout [14].

A SCWR designed with reheat channels introduces an additional extraction point for a HE as depicted in Figure 3. Higher-coolant enthalpy at the reheat outlet compared to other scenarios improves the amount of heat that can be transferred from the SCWR with a reduced mass-flow rate. Identical temperature conditions to the single-reheat-cycle reactor outlet coupled with subcritical coolant pressure makes this location desirable for a HE.

Based on thermodynamic analysis performed by Naidin et al. [14] on basic arrangements of the no-reheat, single and double re-heat cycles, the single re-heat with heat regeneration cycle is suggested to be the basis for the development of a SCWR NPP linked to a hydrogen production facility due to the high primary side efficiency. The single reheat loop is characterized by increased complexity due to the introduction of reheat channels. Thus, integration of a hydrogen-production facility with a SCWR NPP is also dependent on the successful development of the SCWR technology.

5. Energy requirements

The external-heat requirement for a Cu-Cl cycle-based hydrogen-production facility will depend upon the level of heat recycling incorporated into the system design. For an ideal 5-step cycle described above using the slurry-drying method the heat requirements equal to 277.4 kJ per gram of H_2 gas produced. Delivering all of the heat generated from the exothermic reactions within the cycle to those areas, where heat is required would reduce the heat requirement to 161.4 kJ per gram of H_2 produced. The proceeding analysis assumes that no heat is recycled within the hydrogen facility maximizing the external heat required from a SCWR. Fluid heat losses occurring between the NPP and hydrogen facility would further increase the amount of heat to be diverted.

For a layout incorporating a HE immediately after the reactor outlet the mass-flow rate diverted away from the main reactor loop would depend upon the reactor cycle: no-reheat or single reheat. Table 2 outlines heat-requirement data given variable heat losses between an NPP and hydrogen-production facility for an NPP rated at 1200 MW_{el} output. Listed mass-flow rates are fractions of the total mass-flow rate at that point in the reactor loop. This analysis assumes that hydrogen is produced at a rate of 1 kg/s or 86,400 kg/day, categorized as a medium-size hydrogen-production operation. Current SMR plants range in size from producing approximately 5,000 kg/day to 500,000 kg/day [2].

Table 2. Fraction of Reactor Power and Mass-Flow Rates Required For a Heat Exchanger Located at the Reactor Outlet for a Hydrogen-Production Facility Operating at H_2 Production at 1 kg/s.

	System Heat Losses, %	**0**	**10**	**20**
Reactor Outlet Heat Exchanger	No Reheat - Total P_{th} Required, %	11.9	13.2	14.8
	No Reheat - Mass Flow, kg/s	142.6	158.5	178.3
	Single Reheat - Total P_{th} Required, %	12.0	13.3	15.0
	Single Reheat - Mass Flow, kg/s	142.5	158.4	178.2
HP Turbine Outlet - No Reheat Cycle	Total P_{th} Required, %	11.9	13.2	14.8
	Mass Flow, kg/s	151.4	168.3	189.3
Reheater Outlet Heat Exchanger – Single Reheat Cycle	Total P_{th} Required, %	12.0	13.3	15.0
	Mass Flow, kg/s	114.9	127.6	143.6

The overall fraction of energy diverted from the SCWR NPP to the hydrogen facility is minimized when the facility linked HE is located at the reactor outlet channels or HP turbine outlet in a no-reheat layout. This would draw 11.9% of the total reactor power compared to 12.0% for a single-reheat layout. From this the overall energy fraction is cycle dependent and not based on the location within the cycle. The layout consisting of an HE at the outlet of the HP turbine extraction line was found not to be thermodynamically sufficient to supply the entire hydrogen facility's heat requirements. An additional HE would be required to supply the full heat load if this scenario were selected.

6. Operational and safety concerns

Both SCWRs and thermochemical hydrogen-production facilities have unique operational concerns. Operating conditions differ significantly between the two facilities. The Cu-Cl cycle's operating pressure is on the order of several atmospheres compared to SCWR coolant pressure at 25 MPa. The maximum temperature demand of the Cu-Cl cycle is ~530°C at the oxygen-production reactor compared to the SCWR outlet temperature of 625°C. Regardless of the process layout that is selected, the linking HE must be located within the NPP's containment structure. Therefore, superheated steam on the hydrogen-facility loop must be returned via pipeline to the facility for distribution. The minimum distance between these two facilities has been estimated at 100−150 m. To ensure feasibility of the facility linkage, adequate insulation and routine inspection of piping would be mandatory. Development of control mechanisms to regulate coolant diversion from the reactor loop will need to be developed such that NPP operation does not undergo undesired transients during operation of the hydrogen production facility.

As hydrogen is colourless, odourless and flammable, a safety risk is introduced to the neighbouring NPP as abnormal operating scenarios at the hydrogen-production facility could have adverse impact on reactor operation. Risk levels for different conditions at a Cu-Cl facility have been assessed through fault-tree analysis with solutions proposed for select cases [15]. Heat input and recovery systems such as the inter-facility HE have not been considered and will need to be assessed in the future.

Due to the large difference in operating pressure between the two facilities, material limitations may restrict the design of an HE located at the reactor outlet channels. At this particular location the pressure differential would be the largest. For this scenario, an intermediate coolant loop would be required between the two facilities. A supercritical water/superheated steam intermediate HE located within NPP containment would interface with a secondary superheated steam/superheated steam HE on the hydrogen production facility side. This would provide a step-down in pressure differential between the operating loops to reduce material impacts on HE equipment thereby lessening the severity of potential accidents.

7. Impact on the electricity production

Nuclear power plants are effective in providing base-load supply to the electrical grid. Cycling reactor thermal power output to meet electrical demand is undesirable both in regards to reactor kinetics and the added stress on system components due to transient operation. For a SCWR linked to a hydrogen-production facility reactor output would be controlled independently of process-heat requirements. Analysis conducted by Naidin et al. [11] for the no-reheat and single-reheat SCWR cycles identified efficiencies of approximately 50% and 51%, respectively for a 1200 MW_{el}-rated plant. With the linkage of a Cu-Cl cycle facility to an SCWR, reactor coolant would be passed through the high-temperature HE in quantities appropriate to ensure adequate process heating for hydrogen production and electrical grid stability.

Preliminary analysis for a SCWR supplying 100% of the required process heat to a hydrogen facility indicates that electrical power output of the NPP would drop from 1200 MW_{el} to approximately 1035 MW_{el} for a no-reheat reactor loop assuming an ideal Cu-Cl cycle process with no associated heat losses. This assumes that the percentage of power lost in the HP turbine and LP turbines is equal. Further accounting for the 31.3 kJ per gram of H_2 required by the electrolysis reaction step, the net electrical output of the NPP would fall to approximately 1004 MW_{el}. With scheduled operation of the hydrogen-production facility alternate generating resources may be arranged to reconcile the difference in supply to ensure a controlled transition at the NPP and minimal impact on electrical-grid stability.

8. Conclusions

Hydrogen production using the thermochemical Cu-Cl cycle may be supported through process heat supplied by a SCWR. Select SCWR design concepts currently under development have coolant pressures of 25 MPa and reactor outlet temperatures of 625°C, which exceed the highest temperature requirement for reaction steps in the Cu-Cl cycle. Development of an interface between a NPP and hydrogen-production facility can be achieved through the use of a HE. This linkage would support non-carbon based hydrogen production while increasing the overall utilization factor for the NPP. The following conclusions can be made from the above discussion:

1. The Cu-Cl cycle is attractive for hydrogen production due to its low-temperature requirements and equipment costs compared to other cycles and laboratory-tested/validated reaction steps.
2. A SCWR-Cu-Cl cycle hydrogen-production facility linking HE may be designed for the no-reheat and single-reheat reactor loops;
   i) the no-reheat-cycle layout may involve an HE at the reactor outlet, the HP turbine outlet or the HP turbine extraction point outlet;
   ii) the single-reheat-cycle layout may involve an HE at the reactor outlet or the reheater outlet.
3. An HE located at the reactor outlet or HP turbine outlet would supply the necessary heat to the hydrogen-production facility while minimizing the thermal energy diverted from the NPP: for the ideal case, as little as 11.9% of the SCWR's thermal power for a 1 kg/s production rate of H_2 gas; an HE part of a no-reheat cycle located downstream of the HP turbine extraction outlet would not be capable of supplying the entire energy requirement for the hydrogen production facility and would require an additional HE on the reactor loop to supply the difference.
4. An assessment of conditions and potential accident scenarios must be conducted for SCWR-Cu-Cl facility HEs at each potential location within the loop.
5. A SCWR linked to a hydrogen-production facility would not have adverse impacts on an electrical grid provided that electricity demand would govern the availability of hydrogen production.

9. Acknowledgements

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10. Nomenclature

Q	Thermal Energy
V	Electrical Energy
P_{th}	Thermal Power

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