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Thermodynamic Analysis of the Use a Chemical Heat Pump to Link a Supercritical Water-Cooled Nuclear Reactor and a Thermochemical Water-Splitting Cycle for Hydrogen Production*

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Abstract

Increases in the power generation efficiency of nuclear power plants (NPPs) are mainly limited by the permissible temperatures in nuclear reactors and the corresponding temperatures and pressures of the coolants in reactors. Coolant parameters are limited by the corrosion rates of materials and nuclear-reactor safety constraints. The advanced construction materials for the next generation of CANDU reactors, which employ supercritical water (SCW) as a coolant and heat carrier, permit improved "steam" parameters (outlet temperatures up to 625ºC and pressures of about 25 MPa). An increase in the temperature of steam allows it to be utilized in thermochemical water splitting cycles to produce hydrogen. These methods are considered by many to be among the most efficient ways to produce hydrogen from water and to have advantages over traditional low-temperature water electrolysis. However, even lower temperature water splitting cycles (Cu-Cl, UT-3, etc.) require an intensive heat supply at temperatures higher than 550–600°C. A sufficient increase in the heat transfer from the nuclear reactor to a thermochemical water splitting cycle, without jeopardizing nuclear reactor safety, might be effectively achieved by application of a heat pump, which increases the temperature of the heat supplied by virtue of a cyclic process driven by mechanical or electrical work. Here, a high-temperature chemical heat pump, which employs the reversible catalytic methane conversion reaction, is proposed. The reaction shift from exothermic to endothermic and back is achieved by a change of the steam concentration in the reaction mixture. This heat pump, coupled with the second steam cycle of a SCW nuclear power generation plant on one side and a thermochemical water splitting cycle on the other, increases the temperature of the "nuclear" heat and, consequently, the intensity of heat transfer into the water splitting cycle. A comparative preliminary thermodynamic analysis is conducted of the combined system comprising a SCW nuclear power generation plant and a chemical heat pump, which provides high-temperature heat to a thermochemical water splitting cycle for hydrogen production. It is concluded that the proposed chemical heat pump permits the utilization efficiency of nuclear energy to be improved by at least 2% without jeopardizing nuclear reactor safety. Based on this analysis, further research appears to be merited on the proposed advanced design of a nuclear power generation plant combined with a chemical heat pump, and implementation in appropriate applications seems worthwhile.

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1. Introduction

One of today's greatest technical challenges is to find a way to replace fossil fuels so as to significantly reduce greenhouse gas and air pollution emissions, resolve concerns about global warming and make our planet cleaner. Perhaps the simplest way to accomplish this is to generate electricity from renewable energy sources like wind, solar, tidal, geothermal, etc., and to produce hydrogen for certain energy applications via electrolysis of water. The hydrogen can be utilized directly as a fuel or as a source for synthetic liquid fuels. The significant quantities of expensive construction materials in renewable power plants lead to higher electricity costs than for natural gas or coal power plants. Life cycle assessments of renewable technologies for producing electricity and hydrogen and reasons for the higher costs associated with renewable-based electricity and hydrogen have been considered $(1)(2)$.

Given the current state of renewable technologies, many argue that the only viable alternative to fossil fuels is nuclear energy. Nonetheless, the use of nuclear energy raises concerns with many. Nuclear energy must be used safely and effectively to produce power.

The efficiency of power generation increases with increasing temperature of the working medium which, in the case of the nuclear energy utilization, leads to increased nuclear-reactor coolant temperatures and pressures. Such increases in working-medium conditions are constrained by the characteristics and corrosion rates of construction materials and concerns about nuclear reactor safety. The advanced construction materials expected to be used in the next generation of CANDU reactors, which employ SCW as a coolant and heat carrier, permit increased "steam" parameters (i.e., an outlet temperature up to 625ºC and a pressure of about 25 MPa). For the first generation of SCW NPPs, this is expected to increase the power generation efficiency from 35 to 45% ⁽³⁾. Increasing the "steam" temperature also makes it possible to utilize it in thermochemical water splitting cycles to produce hydrogen. However, even lower temperature water splitting cycles (Cu-Cl, UT-3, etc.) often require heat at temperatures of 550–600 $^{\circ}$ C or higher ^{(4) (5)}.

A sufficient increase in the temperature of heat transfer from the nuclear reactor to a thermochemical water splitting cycle, without jeopardizing nuclear reactor safety, might be achieved by application of a heat pump, which increases the temperature of the heat supplied via a cyclic process driven by mechanical or electrical work. The development of high-temperature heat pumps and their applications in high-temperature nuclear reactors is a substantially new area in engineering. Safonov et al. ⁽⁶⁾ proposed a high-temperature continuous chemical heat pump connected to a gas-cooled nuclear reactor, where a shift from heat absorption to heat release was achieved by a pressure drop of the gaseous reaction mixture. Kato et al. $(7)(8)$ investigated a chemical heat pump that uses the reaction system of calcium oxide/lead oxide/carbon dioxide, which was developed for use on high-temperature heat (above 800°C). The heat transfer from lower (about 800°C) to higher (about 900^oC) temperature proceeds periodically through the heat storage and heat output modes. The same periodic character is a feature of other designs of such chemical heat pumps (9) .

In this paper, a chemical heat pump is proposed and investigated that works continuously, absorbing heat at temperatures lower than 873 K (600ºC) and releasing it at higher than 873 K (600°C). These parameters facilitate its implementation in proposed designs for SCW-CANDU nuclear power generation plants. The transferred heat is appropriate for use in a chemical water splitting cycle to produce hydrogen. The purpose

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of this preliminary thermodynamic study is to evaluate the influence of introducing the proposed heat pump on the efficiency of the nuclear power generation plant.

Nomenclature

Greek symbols

Subscripts

Superscripts

2. Integration of a Chemical Heat Pump into SCW-CANDU Power Generation Cycle

It is proposed to integrate a chemical heat pump into a SCW-CANDU power cycle. The operating principles of the proposed heat pump and its performance are described here. At higher temperatures, the exothermic conversion reaction of a synthesis gas (a mixture of carbon monoxide and hydrogen) into methane is carried out:

$$
CO + 3H_2 \rightarrow CH_4 + H_2O + Q \tag{1}
$$

At lower temperatures, the inverse related endothermic reaction for methane conversion occurs:

$$
CH_4 + H_2O \rightarrow CO + 3H_2 - Q \tag{2}
$$

Reactions (1) and (2) proceed simultaneously with the rapid water-shift reaction:

$$
H_2 + CO_2 \leftrightarrow H_2O + CO \tag{3}
$$

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The chemical equilibrium is shifted by changing the steam content in the reactor inputs. Reaction (1) in the methanator (device 1; Fig. 1) proceeds at a deficiency and reaction (2) in the reformers (devices 2 and 4; Fig. 1) at a significant excess of steam.

Fig. 1 Scheme of a high-temperature heat pump implemented into the second power generation cycle of a SCW-CANDU nuclear plant. Numbers indicate devices according to the following legend:

1: methanator; 2,4: autothermal methane converters; 3, 5, 9, 14: reheaters; 6: low-pressure steam turbine; 7: condenser; 8: compressor; 10: pump; 11: boiler; 12: superheater; 13: high-pressure steam turbine.

The performance principles of a nuclear power generation plant with a chemical heat pump producing high-temperature heat are presented in Fig. 1. Water, after compression in a pump (device 10), is heated, evaporated and superheated in a boiler (device 11) and a superheater (device 12). The heat required is delivered with the steam from the first cycle in the SCW-CANDU nuclear reactor. Then, the superheated steam enters a high-pressure turbine (device 13) where it generates mechanical work as its pressure and temperature decrease. After reheating the steam in device 14, it is mixed with the methane-containing reaction mixture. The resulting gaseous flow consequently enters two autothermal (adiabatic) methane reformers (devices 2 and 4) and two intermediate reheaters (devices 3 and 5). In the methane reformers the endothermic methane-conversion reaction of the mixture of hydrogen and carbon monoxide (equation 2) is carried out. Due to the endothermic nature of this reaction, the temperature of the gaseous flow decreases and, in order to increase the mechanical work from the low-pressure turbine (device 6), it is heated in devices 3 and 5. Downstream of the low-pressure turbine (device 6), the gaseous flow is cooled in a condenser (device 7) where the steam is separated into water and a reaction mixture as it condenses. The gaseous reaction mixture enters a compressor (device 8), is heated in a reheater (device 9) and is directed into the methanator (device 1). Reaction (1) is carried out in device 1 with a steam deficiency. The heat (Q_1) at temperature $T_1 \ge 600^{\circ}$ C is produced with the purpose of utilizing it in a combined chemical water-splitting cycle to

produce hydrogen. In such cycles, high-temperature heat is employed to drive some endothermic conversions in order to avoid or reduce the electricity consumption during the stage of electrolysis. Theoretically, the electricity consumption for electrolysis can be reduced by the value of the high temperature heat employed (Equation (6)).

The minimum pressure (P_{min}) in the power generation cycle should be increased because of the presence of uncondensed gases in the gaseous flow which enters the condenser (device 6); here, this pressure is chosen to be 1 atm (0.1013 MPa) . In typical steam-water power generation cycles, the minimum pressure is about 0.003–0.005 MPa (10). At these low pressures, the uncondensed gases occupy an extremely great volume, leading to a significant increase in the size of the condenser and pipes. At $P_{min} = 1$ atm steam condensation starts at a temperature of around 100° C and some of the released heat (Q_{rec}) is used for preliminary water heating in the boiler (device 11). The excess released heat (O_7) Q_{rec}) in the condenser of the power generation cycle with a heat pump (Fig. 1) is transmitted to a standard power generation cycle with a minimum pressure $P_{min} = 0.0314$ atm (\approx 0.0032 MPa) (Fig. 2). There, it is used for preliminary water heating in the boiler.

Fig. 2 Scheme of simplified standard second power generation cycle in a SCW-CANDU nuclear plant.

Numbers indicate devices according to the following legend: 1: reheater; 2: low-pressure steam turbine; 3: condenser; 4: pump; 5: boiler; 6: superheater; 7: high-pressure steam turbine.

The thermodynamic analysis in the next section assesses the effect of introducing the proposed heat pump on the thermal efficiency of the SCW-CANDU power generation plant.

3. Thermodynamic Analysis

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The general assumptions applied in the thermodynamic analysis of the proposed design follow: (i) energy losses due to mechanical friction are negligible; (ii) thermodynamic and chemical equilibria are achieved at the outlet of the methanator (device 1) and methane reformers (devices 2 and 4); and (iii) the performance of the turbines and compressors is considered ideal.

Property data are evaluated using the following simplifications: (i) all gases except steam are modeled as ideal; (ii) thermodynamic data for liquid water and steam are taken from the NIST standard reference database (version 7.0), (iii) thermodynamic properties of others gases are taken from (11) ; and (iv) thermodynamic properties of gaseous mixtures are calculated assuming an additive input of the components.

The general parameters used in the analysis of nuclear power generation cycles with a heat pump (Fig. 1) and without one (Fig. 2) are listed in Table 1.

Table 1 Parameters of the two cycles of the nuclear power generation plant.

The temperatures of the gaseous mixtures and steam at the output of the reheaters (devices 3,5,9,12 and 14 in Fig. 1; device 1 in Fig. 2) and superheaters (device 12 in Fig. 1 and device 6 in Fig. 2), where heat exchange with a nuclear reactor occurs, are taken equal to $T_{\text{max}} = 873$ K. The high-potential heat is generated in the methanator (device 1, Fig. 1). Assuming that the inlet and outlet temperatures are even, i.e., $T_1^{out} = T_1^{in} = T_{max} = 873 \text{ K}$, the heat can be supplied at a temperature not lower than 873 K.

Since the performance of turbines and compressors is regarded as ideal and adiabatic, the unknown temperature (input or output) is determined by equating the entropies of the input and output flows and the mechanical work (consumed or released) by evaluating the difference in enthalpies of the same flows. The heat (released or consumed) in the heat exchangers and condenser is equal to the difference between the input and output enthalpies of the input and output flows considered. The composition of the gaseous mixture at the output of the methanator (device 1, Fig. 1) is defined by the chemical equilibrium assumed at its outlet; the composition and temperature of the gaseous mixture at the output of methane converters (devices 2 and 4, Fig. 1) are simultaneously defined by the chemical equilibrium assumed at their outlets and the adiabatic condition (no external heat supplied) applied to their performance. As is generally accepted in thermodynamic calculations for power plants, the energy consumed by the water pump is assumed negligible. An iterative procedure is applied to achieve convergence between the gaseous composition at the methane converter (device 4, Fig. 1) outlet and at the methanator (device 1, Fig. 1) inlet.

An absence of inert gases in the mixture with steam in the standard cycle (Fig. 2) permits lowering the minimum pressure P_{min} and carrying out condensation in device 3 at a constant and minimum temperature T_{min} . In the case where a heat pump is introduced (Fig. 1) this condensation (device 7) starts at a temperature of about 373 K and finishes at T_{min} = 298 K. The released heat is used to heat water downstream of the pump (device 10) and can be used to heat water in a standard nuclear power generation cycle (Fig. 2) if the two technologies are combined. Clearly, an increase in P_{min} for the scheme with a heat pump leads to a reduction in the power generated.

4. Results and Discussion

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The results of the thermodynamic analysis are listed in Tables 2–6. Tables 2 and 3 present heat balances. Data are given per 20 moles of steam circulated in the considered schemes (Figs. 1 and 2). The work obtained in the cycle must be equal to the difference between the consumed and released heat.

Table 2 Work and heat flows and energy balance* for the nuclear power generation cycle with a high-temperature heat pump (Fig. 1).

* Data are given per 20 moles of water circulated in the system.

** Due to a steam enthalpy drop when decreasing its partial pressure as it is mixed with the reaction mixture, a negligible amount of heat is required to maintain a constant temperature at the inlet of the methane converter 2.

Table 3 Energy balance* of the standard nuclear power generation cycle (Fig. 2).

* Data are given per 20 moles of water circulated in the system.

As seen in these Tables 2 and 3, the mechanical work (power) generated in the standard cycle (Fig. 2) is higher than that generated in the scheme with a heat pump (Fig. 1). The high-potential heat with a temperature equal to or higher than 873 K has a magnitude Q_1 = 94.2 kJ.

The composition of the gaseous flow in the heat pump cycle is presented in Table 4.

In the methanator (device 1), H_2 , CO and CO₂ are converted to CH₄, increasing its quantity from 0.39 to 0.89 moles, and heat is released. In the methane converters (devices 2 and 4), the opposite process occurs: in an excess of steam, methane is converted to hydrogen, carbon monoxide and carbon dioxide. The heat content and temperature of the gaseous flow in the reactors both decrease (see Table 5) but this reduction is compensated for in the reheaters (devices 3 and 5).

As seen in Table 5, water condenses out of the mixture of gases in the condenser (device 7, Fig. 1) at a variable temperature, i.e., T_7^{in} = 368 K to T_7^{out} = 298 K. Some of the heat *Qrec* released in the condenser can be recovered for preliminary water heating in the boiler (device 11) from 298 to 368 K.

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Table 4 Composition of the gaseous flows circulated in the high-temperature heat pump.

Table 5. Input and output working-fluid temperatures and pressures for devices in the nuclear power generation cycle with a high-temperature heat pump (Fig. 1).

Table 6 lists efficiency indicators for the compared schemes. The energy (or thermal) efficiency of the schemes are evaluated as

$$
\eta_T^{(i)} = \frac{W_i}{Q_{nuc}^{(i)}}
$$
\n⁽⁴⁾

where index *i* takes on a value of 1 or 2 for the schemes in Figs. 1 and 2, respectively, W_i is the work produced, and $Q_{nuc}^{(i)}$ is the heat released by the SCW-CANDU nuclear reactor and consumed by the second cycle of the overall power generation plant. Values for *W* and Q_{nuc} are given in Tables 2 and 3. Here, Q_{nuc} is equal to the sum of the consumed heats (taken with opposite signs) in the schemes. Taking into account that nuclear heat consumption Q_{nuc} in the scheme with a heat pump can be reduced by Q_{rec} (Table 6, column 3), its thermal efficiency can be written as follows:

$$
\eta_{\rm T}^{(1)} = \frac{W_{1}}{Q_{\rm nuc}^{(1)} - Q_{\rm rec}}
$$
 (5)

where the indexes refer to scheme 1. Thermal efficiencies for Schemes 1 and 2 (Figs. 1) and 2) are found to be 0.37 and 0.487, respectively. These data favor the standard scheme (scheme 2).

* Data are given per 20 moles of water circulated in the system.

We now assume that electricity is employed in the system where low-temperature electrolysis and chemical decomposition of water are applied simultaneously, like in the Cu-Cl and UT-3 processes. Applying a general approach to determine the efficiency of chemical water decomposition, the following expression for hydrogen energy production can be inferred:

$$
LHV_{H_2} = \eta_e W + Q_1 (T \ge 873 K)
$$
\n(6)

where LHV_{H_2} is the lower heating value of hydrogen; η_e is the low-temperature electrolysis efficiency; *W* is electrical work, and Q_1 is an external heat supply at a temperature higher than or equal to 600ºC. Then the efficiency for nuclear heat utilization for hydrogen production is expressible as follows:

$$
\frac{LHV_{H_2}}{Q_{\text{nuc}}} = \eta_e(\frac{W + Q_1}{Q_{\text{nuc}}}) = \eta_e\eta_T^{H_2}
$$
\n
$$
W + Q_1
$$
\n
$$
(7)
$$

$$
\eta_{\scriptscriptstyle T}^{\scriptscriptstyle H_2} = \frac{\eta_{\scriptscriptstyle e}}{\mathcal{Q}_{\scriptscriptstyle nuc}} \tag{8}
$$

where $\eta_{\tau}^{H_2}$ is thermal efficiency of nuclear energy use, calculated assuming high-temperature heat generated in the heat pump is employed for hydrogen production in low-temperature water decomposition processes. The efficiency $\eta_{\tau}^{H_2}$ for the nuclear power generation cycle with a heat pump is calculated for a typical electrolysis efficiency (per hydrogen LHV) $\eta_e = 0.72^{(4)}$ and listed in Table 6, column 6. In line with Equation (8) for the standard nuclear power generation cycle (Fig. 2), $\eta_T^{H_2} = \eta_T$. It is seen in this table that $\eta_T^{H_2}$ for the cycle with a heat pump is lower than that for the standard cycle.

Analyzing values for the recovered heat $Q_{rec} = 105.1$ kJ (Table 6) and the low-temperature heat released in the condenser (device 7, Table 2) $Q_7 = 918.2$ kJ, it is seen

that only a small part of this heat is utilized. Most of the low-potential heat can be employed for preliminary water heating in the boiler of the standard nuclear power generation scheme (Fig. 2). This observation means that cogeneration is possible from the standard nuclear power generation cycle and a heat pump. The maximum degree of such cogeneration *N* is estimated as follows:

$$
n \le N = \frac{Q_7 - Q_{rec}}{Q_{rec}} \tag{9}
$$

where Q_7 is the heat released in the condenser of the scheme with a heat pump and Q_{rec} is the heat which could be utilized for heating a certain amount of water (20 mol in our case) in a boiler. The value $n = 0$ means no cogeneration occurs, while $n = 1$ means that 20 mol of water in the standard nuclear power generation cycle is heated by the heat from the condenser of the cycle with a heat pump, and $n = N$ means that N^*20 moles of water are heated in this way. Increasing *v*alues of *n* increases the relationship between the power capacities of the two schemes. The formulas for η_T and $\eta_T^{H_2}$ for a combined system reflect this dependence as follows:

$$
\eta_{\scriptscriptstyle T} = \frac{nW_{\scriptscriptstyle 2} + W_{\scriptscriptstyle 1}}{n(Q_{\scriptscriptstyle nuc}^{(2)} - Q_{\scriptscriptstyle rec}) + (Q_{\scriptscriptstyle nuc}^{(1)} - Q_{\scriptscriptstyle rec})}
$$
(10)

$$
\eta_{T}^{H_{2}} = \frac{nW_{2} + W_{1} + Q_{1}}{n(Q_{\text{nuc}}^{(2)} - Q_{\text{rec}}) + (Q_{\text{nuc}}^{(1)} - Q_{\text{rec}})}
$$
(11)

where the indexes refer to scheme 1 or 2.

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The efficiencies η_{T} and $\eta_{T}^{H_2}$ as a function of a cogeneration degree *n* are presented in Fig. 3. It follows from this graph for *n* of about 3 the thermal efficiency η ^T of a combined system is equal and $\eta_T^{H_2}$ is greater than the standard one. This is achieved as a result of reducing the power generation capacity of the scheme with a heat pump (this reduction is proportional to *n*) with respect to the standard one.

Fig. 3 Variation of thermal and hydrogen production efficiencies η_{T} and $\eta_{T}^{H_2}$ with cogeneration factor *n*.

Fig. 4. Advanced scheme involving a heat pump implemented into the second power generation cycle of a SCW-CANDU nuclear plant. Numbers indicate devices according to the following legend: 1: methanator; 2, 4: autothermal methane converters; 3, 5, 10, 15: reheaters; 6: low-pressure steam turbine; 7: low-pressure condenser; 8, 11: pumps; 9: high-pressure condenser; 12: boiler; 13: superheater; 14: high-pressure steam turbine.

Figure 4 represents an alternative approach regarding the utilization of released heat, which avoids compromising the power generation capacity when a chemical heat pump is applied.

The application of exergy analysis, a type of thermodynamic analysis based on the second law, to this scheme is the subject of ongoing research by the authors. An exergy analysis will allow additional efficiencies to be determined, that assess performance relative to ideality, and losses to be clearly identified in terms of type, location and cause. Thus the results of an exergy analysis are expected to assist efforts to improve designs.

5. Conclusions

A heat pump that permits an increase in the temperature of the heat transmitted from the first cycle in a SCW-CANDU nuclear reactor is proposed. This heat pump employs a catalytic methane conversion reaction where the reaction mixture of methane, steam, hydrogen, carbon monoxide and carbon dioxide is the working medium. Heat transfer to an external consumer occurs during methane synthesis and its absorption during the adiabatic (autothermal) methane-conversion to hydrogen and carbon monoxide. The reaction shift from exothermic to endothermic and back is achieved by changing the steam concentration in the reaction mixture. This heat pump is implemented in the second power

generation cycle of a SCW-CANDU nuclear plant.

A preliminary comparative thermodynamic analysis is conducted of the combined system comprising a SCW nuclear power generation plant and a chemical heat pump, which provides high-temperature heat to a thermochemical water splitting cycle for hydrogen production. Applying the proposed chemical heat pump improves utilization of nuclear energy at least by 2% without jeopardizing nuclear reactor safety. Based on the analysis an advanced design of a combined nuclear power generation plant with a chemical heat pump is proposed.

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