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International Conference on Nuclear  
Engineering (ICONE-14)

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July 2006

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U.S. Department of Energy  
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## A PROCESS MODEL FOR THE PRODUCTION OF HYDROGEN USING HIGH TEMPERATURE ELECTROLYSIS

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### ABSTRACT

High temperature electrolysis (HTE) involves the splitting of steam into hydrogen and oxygen at high temperatures. The primary advantage of HTE over conventional low temperature electrolysis is that considerably higher hydrogen production efficiencies can be achieved. Performing the electrolysis process at high temperatures results in more favorable thermodynamics for electrolysis, more efficient production of electricity, and allows direct use of process heat to generate steam. This paper presents the results of process analyses performed to evaluate the hydrogen production efficiencies of an HTE plant coupled to a 600 MWt Modular Helium Reactor (MHR) that supplies both the electricity and process heat needed to drive the process. The MHR operates with a coolant outlet temperature of 950 C. Approximately 87% of the high-temperature heat is used to generate electricity at high efficiency using a direct, Brayton-cycle power conversion system. The remaining high-temperature heat is used to generate a superheated steam / hydrogen mixture that is supplied to the electrolyzers. The analyses were performed using the HYSYS process modeling software. The model used to perform the analyses consisted of three loops; a primary high temperature helium loop, a secondary helium loop and the HTE process loop. The detailed model included realistic representations of all major components in the system, including pumps, compressors, heat exchange equipment, and the electrolysis stack. The design of the hydrogen production process loop also included a steam-sweep gas system to remove oxygen from the electrolysis stack so that it can be recovered and used for other applications. Results of the process analyses showed that hydrogen production efficiencies in the range of 45% to 50% are achievable with this system.

### INTRODUCTION

Because of its ability to produce high-temperature helium, the MHR is well suited for a number of nuclear process-heat applications, including hydrogen production. Two hydrogen-production technologies have emerged as leading candidates for coupling to the MHR: thermochemical water splitting using the sulfur-iodine (SI) process and high-temperature

electrolysis (HTE). In this paper, we discuss a conceptual design developed for coupling the MHR to the HTE process. [1, 2, 3, 4].

### MODEL DESCRIPTION

A process model of the high temperature electrolysis (HTE) plant combined with a high temperature helium reactor was completed using Hyprotech's HYSYS process modeling software. The model, shown in Figure 1, consists of three loops: a primary high temperature helium loop, a secondary helium loop and the HTE process loop.

Helium in the primary loop passes through the reactor, providing cooling for the high temperature reactor fuel by removing 600 MW of thermal power from the reactor core. The high temperature helium leaving the reactor is then split into two streams, with a small fraction of the flow (13%) delivered to an intermediate heat exchanger to provide heat to the HTE process streams, and the majority of the flow passing through a Brayton-cycle gas turbine producing electrical power. After passing through the gas turbine, the major flow stream is further cooled through a recuperator and an ambient precooler heat exchanger. The cool gas is compressed to an intermediate pressure via the low-pressure compressor. The heat of compression is removed using an intercooler heat exchanger. The gas is further compressed to full pressure as it passes through the high-pressure compressor. The gas is then heated by passing through the recuperator. The resulting stream is mixed with the exit stream of the intermediate heat exchanger before entering the reactor. The second stream (13% of the total primary system flow) is passed through an intermediate heat exchanger where the heat is transferred to the secondary helium loop, after which the stream is then slightly compressed using the primary side circulator. Tables 1 & 2 list the properties found in the primary loop.

The secondary helium loop consists of the other side of the intermediate heat exchanger and three additional process heat exchangers. Two of the heat exchangers provide heat to the hydrogen/water side of the HTE process and the remaining heat exchanger heats the steam used to sweep the

oxygen from the electrolysis process. Tables 3 & 4 show the conditions calculated for this loop.

Stream	Temperature (°C)	Pressure (MPa)	Flow (kg/s)
11 - Reactor Inlet	590	7.07	321
1 - Reactor Outlet	950	7.00	321
2 - Turbine Inlet	950	7.00	279
5 - Turbine Outlet	600	2.80	279
3 - Precooler Inlet	129	2.77	279
4 - Precooler Outlet	26	2.74	279
6 - Low Pressure Compressor Outlet	93	4.31	279
9 - High Pressure Compressor Inlet	26	4.27	279
10 - High Pressure Compressor Outlet	104	7.14	279
8 - High Pressure Recuperator Outlet	575	7.07	279
7 - Intermediate Heat Exchanger Inlet	950	7.00	42
13 - Intermediate Heat Exchanger Outlet	679	6.93	42
12 - Primary Side Circulator Outlet	689	7.07	42

Table 1. Temperatures, pressures, and flows of primary loop

Component	Power or Heat Flow (kW)
Reactor Heat	600,000
Turbine Power	510,215
High Pressure Compressor Power	114,959
Low Pressure Compressor Power	98,676
Primary Side Circulator Power	2,224
Recuperator Duty	683,261
Precooler Cooling	149,424
Intercooler Cooling	97,489
Intermediate Heat Exchanger Duty	58,730

Table 2. Power and heat flows of primary loop

Stream	Temperature (°C)	Pressure (MPa)	Flow (kg/s)
14 - Intermediate Heat Exchanger Inlet	292	6	18.1
15 - Intermediate Heat Exchanger Outlet	917	5.94	18.1
22 - HX4 Outlet	854	5.88	18.1
27 - HX3 Outlet	530	5.82	18.1
23 - HX2 Outlet	280	5.76	18.1

Table 3. Temperatures, pressures, and flows of secondary helium loop

Component	Power or Heat Flow (kW)
Secondary Side Circulator Power	1,154
Intermediate Heat Exchanger Duty	58,730
HX2 Duty	23,473
HX3 Duty	30,440
HX4 Duty	5,971

Table 4. Power and heat flow of secondary helium loop

The final loop is the HTE process loop which consists of the hydrogen-steam side and the steam sweep side. Make-up water is pumped to the electrolysis pressure in the liquid phase and is combined with the recycled water. The water is heated to a saturated state through heat exchanger HX1. The heat from heat exchanger HX2 boils off the remaining water after which recycled hydrogen is added to the stream. In this case study, the composition of the stream after the hydrogen addition is 10% hydrogen and 90% steam by mole basis. The hydrogen is added to the inlet stream in order to maintain reducing conditions at the steam/hydrogen electrodes of the electrolysis cells. Heat exchanger HX3 raises the temperature of the steam/hydrogen mixture to as close to the desired electrolysis temperature as possible using heat from the helium secondary side. A high temperature electrical heater (High Temperature Heater) is used to provide the additional heat needed for the electrolysis process. The hydrogen/steam stream leaving the electrolysis process is 90% hydrogen and 10% steam on a mole basis. The water from the hydrogen/steam leaving the electrolysis process is condensed using the heat exchanger HX1. The condensed water is recycled and the hydrogen stream is split into two streams. The first stream is the hydrogen product stream and the second stream is the recycled hydrogen added before the electrolysis process.

The sweep gas is created by pumping water to the electrolysis pressure. The water is boiled and superheated in the heat exchanger HX5 by the sweep steam and oxygen leaving the electrolysis process. The steam is further heated in heat exchanger HX4 to the electrolysis temperature using heat from the helium secondary loop. After the electrolysis process, the steam/oxygen stream composition is 50% steam and 50% oxygen on a mole basis. The steam/oxygen stream is cooled in heat exchanger HX5 to a saturated state. The water is removed at the High Pressure H<sub>2</sub>O/O<sub>2</sub> Knockout Tank and then expanded through a turbine to atmospheric pressure. The stream is further cooled in the Low Pressure H<sub>2</sub>O/O<sub>2</sub> Knockout Tank to the ambient temperature and the water is removed from the stream. See Tables 5 and 6 for the resulting conditions in this loop.

Since the electrolyzer is not a standard HYSYS component, a custom high-temperature electrolysis model was developed for inclusion in the overall system model. The electrolysis model, shown on the right side of Figure 1, is based on the first law of thermodynamics and the Nernst equation. Details of the model can be found in the papers by O'Brien, et. al. [5, 6, 7] For this simulation, the electrolyzer cells have cell cross sectional areas of 225 cm<sup>2</sup> and a current

density of 0.25 A/cm<sup>2</sup> resulting in a total current of 56.25 A. The electrolysis inlet temperature was set to 1100 K. Four millions cells were used, requiring 292 MW of electrolysis power to produce 2345 g/s of hydrogen. The area specific resistance (ASR) of each cell is temperature averaged but based on a value specified at 1100 K. For this model, the ASR at 1100 K was set at 1.25 ohms\*cm<sup>2</sup>, resulting in a temperature averaged ASR of 1.132 ohm\*cm<sup>2</sup>. The model predicted the operating voltage as 1.3 volts per cell. The electrolysis process was operating slightly above thermal neutral which resulted in an electrolysis outlet temperature that was above the inlet temperature. At the thermal neutral voltage, the electrolysis exit temperature is the same as the inlet temperature. The thermal neutral voltage for this case is 1.286 volts per cell at a current level of 47.5 amperes; therefore the extra voltage and current provided heating to the products of the electrolysis process. This extra heat can be recovered by preheating the process or sweep streams.

## EFFICIENCY

As discussed in the following section, overall hydrogen production efficiencies will be affected by the assumed performance of the heat exchangers used in the process. In this analysis each heat exchanger has an effectiveness of 0.95 or less. The effectiveness of a heat exchanger is defined as the ratio of the actual heat transfer to the maximum possible heat transfer. Pressure losses within each heat exchanger were set at 1% of the maximum pressure within each loop. Table 7 shows the effectiveness and minimum approach temperatures of each heat exchanger in the system.

The efficiency of the system is calculated in the following manner. The control volume for the efficiency calculation surrounds all three loops and the sweep gas. First the net power of the turbomachinery is found.

$$\text{Net Power}_{\text{Turbomachinery}} = \sum \text{Power}_{\text{Turbines}} - \sum \text{Power}_{\text{Compressors}} - \sum \text{Power}_{\text{Pumps}} \quad (1)$$

The net total of the electric power into the total system is also found. This total is the sum of the electrical power into the electrolysis process and the sweep gas heater.

$$\text{Net Power}_{\text{Electrical}} = \text{Power}_{\text{Electrolysis}} + \text{Power}_{\text{Sweep Heater}} \quad (2)$$

Also the heating value power of the hydrogen product is calculated.

$$\text{Power}_{\text{H}_2} = \dot{m}_{\text{H}_2 \text{ Produced}} \times \text{Lower Heating Value}_{\text{H}_2} \quad (3)$$

The efficiency is calculated as follows:

$$\eta = \frac{\text{Net Power}_{\text{Turbomachinery}} + \text{Power}_{\text{H}_2} - \text{Net Power}_{\text{Electrical}}}{\text{Power}_{\text{Reactor}}} \quad (4)$$

The numerator is the net power produced and the denominator is the heat power into the system. The efficiency as shown in equation (4) can be also defined as the net power produced over the net power into the system.

## RESULTS

Figures 2 through 7 are temperature vs. heat flow plots of the heat exchangers in the secondary loop and the electrolysis loop. Figure 2 show the profile in the heat exchanger HX1. The cold stream is the recycled water combined with the make-up water. HX1 heats this stream to a saturation state. The hot stream is the 90% hydrogen-10% water stream. This stream is superheated as it enters the heat exchanger. As can be seen on the plot, the dew point of the stream is 147.7 °C at which point the steam begins to condense. Most of the steam is condensed, but is temperature-limited by the temperature of the incoming water stream on the cold side. Figure 3 is the profile plot of heat exchanger HX2. The cold stream is the water stream from HX1. This stream is heated until the stream is slightly superheated to allow the addition of recycled cold hydrogen from the hydrogen product gas. The hot stream is the helium in the secondary loop. Figure 4 shows the temperature vs. heat flow plot of heat exchanger HX3. Although it would appear more heat could be transferred from the secondary helium loop to the steam/hydrogen stream, the heat duty of HX3 is limited due to the heat requirements of HX4 and HX5 to superheat the sweep water to electrolysis temperatures. If the duty of HX3 were to increase, the required duty of HX5 could not be achieved and an external heater would need to be added to heat the sweep gas. An additional external heater is needed to heat up the stream/hydrogen stream to the inlet electrolysis temperatures. Figure 5 shows the temperature/heat flow profile of HX4. The primary purpose of HX4 is to provide the remaining duty required to heat the sweep gas to the electrolysis inlet temperature. As can be seen in Figure 6, HX5 is limited by two phase regions on both sides of the heat exchanger. The cold stream is incoming water that is to be used as the sweep gas. HX5 provides the heat to superheat the water. The hot stream is the 50% oxygen/50% steam stream coming from the electrolysis process. The steam in the hot stream begins to condense at 218.8 °C. The final plot is of the intermediate heat exchanger, see Figure 7. Both streams are helium, the hot stream is from the primary loop and the cold stream is from the secondary loop.

The high temperature reactor supplies both high temperature heat and electric power to the HTE process. The high temperature heat was used to heat the sweep gas to the electrolysis temperature. However, there was not quite enough high temperature heat to heat the process stream to

the desired electrolysis temperature, due primarily to the 95% effectiveness constraint on the heat exchangers. However, the resistance heater (High Temperature Heater) provided the additional heat needed from electric power generated by the Brayton power cycle. When compared to a low temperature reactor, the high temperature reactor provides electricity at a higher thermal efficiency thereby increasing the overall hydrogen production efficiency.

For this system the calculated overall efficiency is 47.9%. In a previous model, the heat exchangers were maximized for heat transfer but limited by a 5°C minimum approach temperature and the pressure drops were set at 20 kPa. In the previous model the overall efficiency was 51.3%. Also in the previous model, there was sufficient heat to raise both the process and sweep streams to the desired electrolysis temperatures, thereby eliminating the High Temperature Heater. As can be seen, using more realistic heat exchanger design parameters decreases the overall efficiency of the system by about 3.4%.

## CONCLUSIONS

A process model of the Modular Helium Reactor combined with a high temperature electrolysis process has shown that efficiencies of nearly 50% can be achieved. In developing these models, realistic heat exchange effectiveness and pressure drops need to be considered to prevent over estimating the expected efficiencies.

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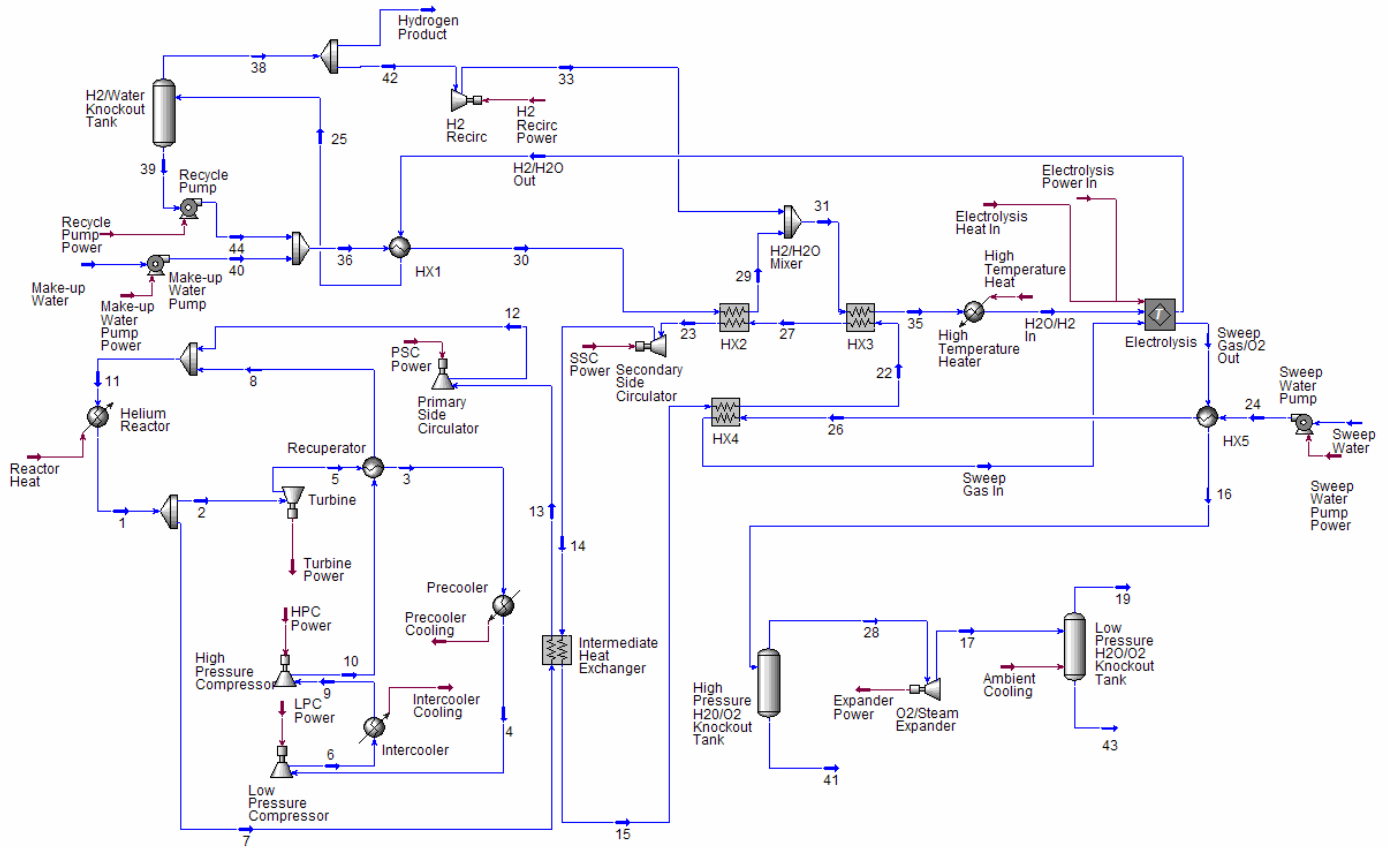


Figure 1. Process flow diagram of high temperature electrolysis process

Stream	Temperature (°C)	Pressure (MPa)	Flow (kg/s)
Make-up Water	21.1	0.101325	21.0
40 - Make-up Water Pump Exit	21.5	5.20	21.0
44 - Water at Recycle Pump Exit	27.2	5.20	2.60
36 - Water Into HX1	22.2	5.20	23.6
30 - Steam/Water Out of HX1	267	5.15	23.6
29 - Steam Out of HX2	281	5.10	23.6
31 - Steam/Hydrogen Into HX3	258	5.10	23.9
35 - Steam/Hydrogen Out of HX3	772	5.05	23.9
H2O/H2 In – Steam/Hydrogen Into Electrolysis	827	5.00	23.9
H2/H2O Out – Steam/Hydrogen Out of Electrolysis	862	5.00	5.26
25 - Hydrogen/Water Out of HX1	27.2	4.95	5.26
39 - Water at Recycle Pump Inlet	27.2	4.95	2.60
38 - Hydrogen at Vapor Outlet of H2/Water Knockout Tank	27.2	4.95	2.66
Hydrogen Product	27.2	4.95	2.36
42 - Hydrogen into H2 Recirc	27.2	4.95	0.295
33 - Hydrogen Out of H2 Recirc	30.7	5.10	0.295
Sweep Water - Water Into Sweep Water Pump	21.1	0.101325	10.5
24 - Water Out of Sweep Water Pump	21.5	5.10	10.5
26 - Steam Out of HX5	586	5.05	10.5
Sweep Gas In – Sweep Gas Into Electrolysis	827	5.00	10.5
Sweep Gas/O2 Out - Steam/Oxygen Out of Electrolysis	862	5.00	29.1
16 - Steam/Water/Oxygen Out of HX5	202	4.95	29.1
41 - Water Out of High Pressure H2O/O2 Knockout Tank	202	4.95	4.45
28 - Steam/Oxygen Into O2/Steam Expander	202	4.95	24.6
17 - Steam/Water/Oxygen After O2/Steam Expander	67.0	0.101325	24.6
19 - Oxygen Out of Low Pressure H2O/O2 Knockout Tank	21.1	0.101325	18.9
43 - Water Out of Low Pressure H2O/O2 Knockout Tank	21.1	0.101325	5.78

Table 5. Temperatures, pressures, and flow of electrolysis steam loop

Component	Power or Heat Flow In (kW)
Make-up Pump Power	141
Recycle Pump Power	0.860
HX1 Duty	43,425
HX2 Duty	23,473
HX3 Duty	30,440
High Temperature Heater Heat	3,350
Electrolysis Power In	291,989
H2 Recirc Power	14.8
HX4 Duty	5,971
HX5 Duty	37,258
Sweep Water Pump Power	69.1
O2/Steam Expander Power	8,342
Ambient Cooling For Low Pressure Knock Out Tank	10,503

Table 6. Power and heat flow of electrolysis steam loop

Heat Exchanger	Effectiveness	Minimum Approach Temperature Difference (°C)
Recuperator	0.950	24.8
Intermediate Heat Exchanger	0.950	32.9
HX1	0.793	5.0
HX2	0.950	13.2
HX3	0.863	81.6
HX4	0.727	90.3
HX5	0.841	5.0

Table 7. Effectiveness of the heat exchangers in the process model

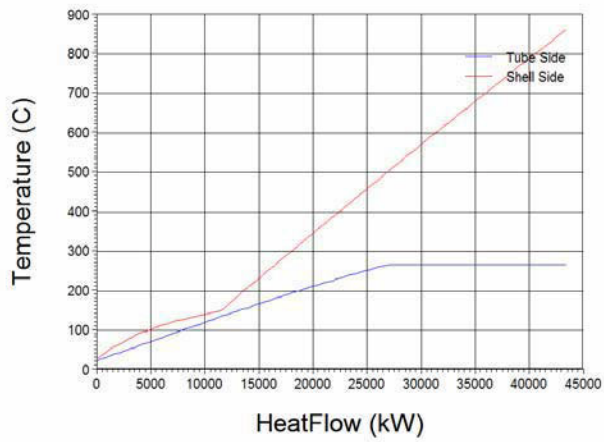


Figure 2. Temperature vs. Heat Flow Plot of HX1

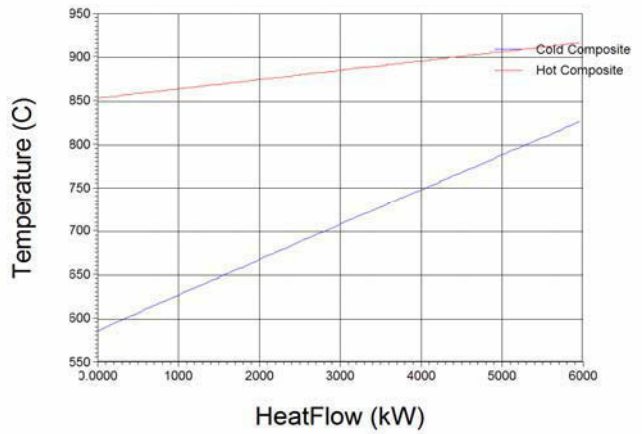


Figure 5. Temperature vs. Heat Flow Plot of HX4

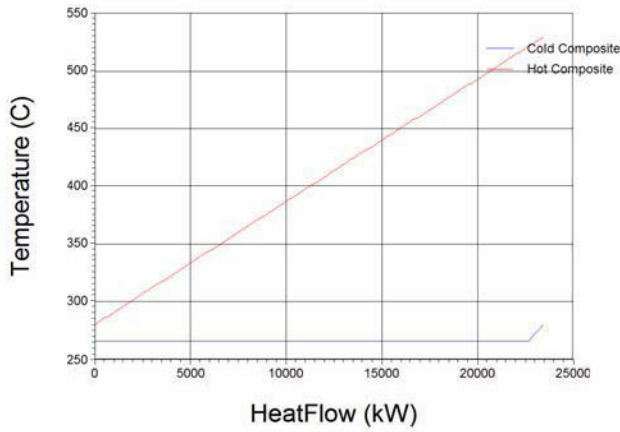


Figure 3. Temperature vs. Heat Flow Plot of HX2

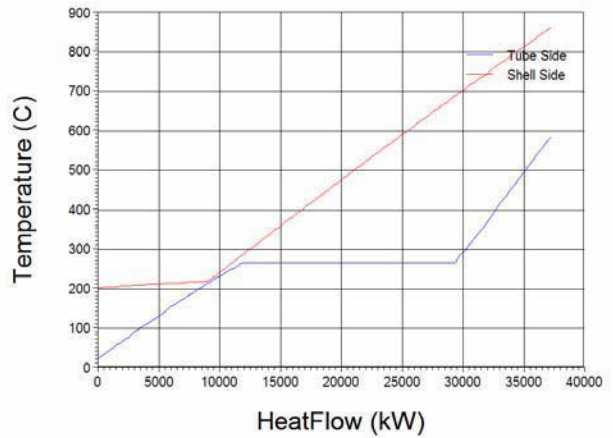


Figure 6. Temperature vs. Heat Flow Plot of HX5

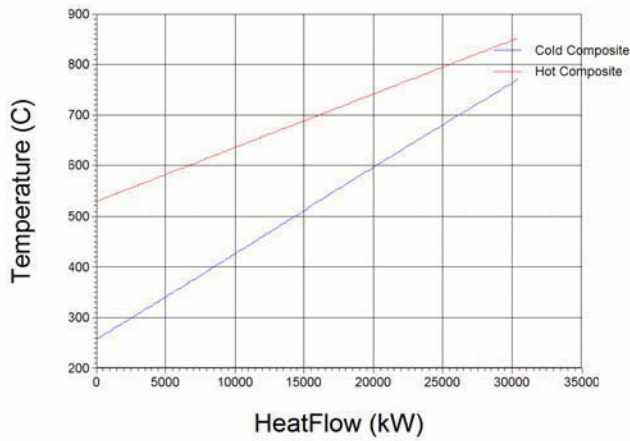


Figure 4. Temperature vs. Heat Flow Plot of HX3

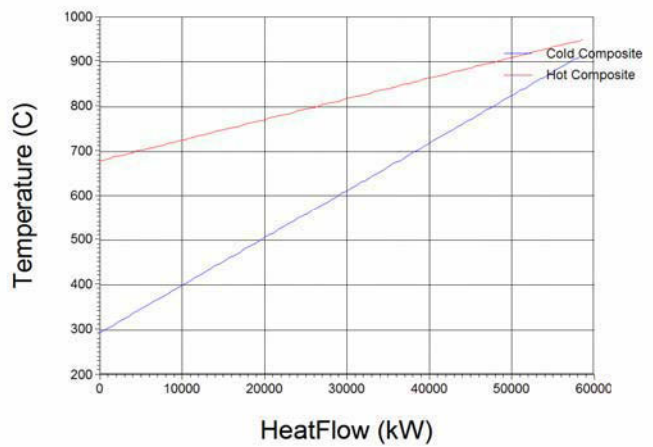


Figure 7. Temperature vs. Heat Flow Plot of Intermediate Heat Exchanger