

# Similitude for Thermal Scale-up of a Multiphase Thermolysis Reactor in the Cu-Cl Cycle of a Hydrogen Production

Mohammed W. Abdulrahman

**Abstract**—The thermochemical copper-chlorine (Cu-Cl) cycle is considered as a sustainable and efficient technology for a hydrogen production, when linked with clean-energy systems such as nuclear reactors or solar thermal plants. In the Cu-Cl cycle, water is decomposed thermally into hydrogen and oxygen through a series of intermediate reactions. This paper investigates the thermal scale up analysis of the three phase oxygen production reactor in the Cu-Cl cycle, where the reaction is endothermic and the temperature is about 530 °C. The paper focuses on examining the size and number of oxygen reactors required to provide enough heat input for different rates of hydrogen production. The type of the multiphase reactor used in this paper is the continuous stirred tank reactor (CSTR) that is heated by a half pipe jacket. The thermal resistance of each section in the jacketed reactor system is studied to examine its effect on the heat balance of the reactor. It is found that the dominant contribution to the system thermal resistance is from the reactor wall. In the analysis, the Cu-Cl cycle is assumed to be driven by a nuclear reactor where two types of nuclear reactors are examined as the heat source to the oxygen reactor. These types are the CANDU Super Critical Water Reactor (CANDU-SCWR) and High Temperature Gas Reactor (HTGR). It is concluded that a better heat transfer rate has to be provided for CANDU-SCWR by 3-4 times than HTGR. The effect of the reactor aspect ratio is also examined in this paper and is found that increasing the aspect ratio decreases the number of reactors and the rate of decrease in the number of reactors decreases by increasing the aspect ratio. Finally, a comparison between the results of heat balance and existing results of mass balance is performed and is found that the size of the oxygen reactor is dominated by the heat balance rather than the material balance.

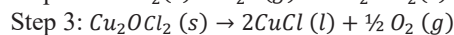
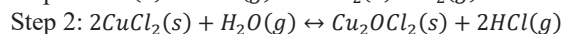
**Keywords**—Clean energy, Cu-Cl cycle, heat transfer, sustainable energy.

## I. INTRODUCTION

THERE is a common belief that in the future, hydrogen will be an effective factor to the supply of sustainable energy, because hydrogen usage would decrease the atmospheric pollution by decreasing the emissions of greenhouse gas [1]. Greenhouse gases can be generated in large quantities from conventional gasoline and diesel vehicles, and in much less quantities from the production of hydrogen gas from various fuel gases.

The alternative methods that can be used for hydrogen production are the thermochemical cycles that are connected to nuclear reactors, where nuclear energy can be used as a clean energy source for large scale production of hydrogen. Cu-Cl

cycle was identified by Argonne National Laboratories (ANL) as one of the promising lower temperature cycles [2], [3]. In this three chemical reactions cycle, there are two thermal and one electrochemical reactions. The three reaction steps of the Cu-Cl cycle are [4];



where *a*, *s*, *l* and *g* denote to aqueous, solid, liquid and gas respectively.

In the oxygen production step of the Cu-Cl cycle (step 3), a solid copper oxychloride ( $Cu_2OCl_2$ ) is decomposed thermally into oxygen gas ( $O_2$ ) and molten cuprous chloride ( $CuCl$ ). The solid  $Cu_2OCl_2$  is fed to the oxygen production reactor from the  $CuCl_2$  hydrolysis reaction (step 2) that operates at a temperature range of 350–450 °C. The materials leaving the oxygen reactor are oxygen gas (which is evolved over a temperature range of 450 to 530 °C) and molten  $CuCl$  [4]. In the oxygen reactor, the decomposition of  $Cu_2OCl_2$  to oxygen and molten  $CuCl$  is an endothermic reaction requiring a reaction heat of 129.2 kJ/mol and a temperature of 530 °C, which is the highest temperature in the Cu-Cl cycle [5]. Thus, heat must be added to increase the temperature of the bulk inside the reactor. The total amount of heat required is the sum of reaction heat and the heat required to raise the reactant temperature from 375 °C (temperature of solid particles from the hydrolysis reaction) to 530 °C, which is the temperature of the decomposition process.

CSTR which is usually used for multiphase reactions like the oxygen reactor, is a mechanically agitated reactor. In CSTR, there is continuous feeding and withdrawing into the reactor for the streams of reactants and products respectively. Generally, a continuous flow stirred tank reactor (CFSTR) assumes that the fluid is perfectly well mixed. Therefore, the system properties, such as concentration, density and temperature, are uniform throughout the reactor. Like any chemical reactor, the oxygen reactor requires accurate control of heat transfer to obtain optimum productivity. In CSTR, heating is achieved by a number of different mechanisms. The most common one involves the use of a jacket surrounding the vessel.

In the literature, different studies have been performed for the oxygen reactor in the Cu-Cl cycle of hydrogen production. Ikeda and Kaye [6] have investigated the thermochemical properties of  $Cu_2OCl_2$  by developing a method that uses a

M. W. Abdulrahman is with the University of Ontario Institute of Technology, Oshawa, ON, Canada (e-mail: mohammed.abdulrahman@uoit.ca).

stoichiometric amount of CuO and CuCl<sub>2</sub> instead of Cu<sub>2</sub>OCl<sub>2</sub> because of its commercial unavailability. Trevani et al. [7] have also examined the thermochemical properties of Cu<sub>2</sub>OCl<sub>2</sub> by using an easy scalable method to produce pure samples of copper oxychloride with larger amounts.

Serban et al. [3] have studied in details the kinetics of the hydrogen and oxygen generation reactions in the Cu-Cl cycle. They have reported that, depending on the basis of thermodynamics and conversions of the different reactants, the completion of the oxygen generation reactions can be performed at a temperature of 530°C. Zamfirescu et al. [8] have investigated the thermophysical properties of copper compounds in the Cu-Cl cycle such as; Cu<sub>2</sub>OCl<sub>2</sub>, CuO, CuCl<sub>2</sub> and CuCl. Since there is no previous available data related to the thermal conductivity and dynamic viscosity of molten CuCl, they have used estimations to evaluate these properties.

Marin [4] has studied experimentally and theoretically the scale-up of the CuO\*CuCl<sub>2</sub> decomposition reactor. He has determined the endothermic reaction rate of the decomposition process, and solved numerically, the conservation and chemical reaction equations for a suddenly immersed particle in a viscous medium. Abdulrahman et al. [1] have studied the scale up of the oxygen reactor in the Cu-Cl cycle from the perspective of material balance for different hydrogen production rates and different residence times. They have investigated in details the factors that influence the size of the oxygen reactor, such as; solid particles characteristics (size, shape and concentration), oxygen bubbles, reactor heating rate and fouling.

From the review, it can be concluded that there is a research blank in the thermal analysis of the oxygen reactor. The design and scale-up of the oxygen reactor require, among others, precise knowledge of the kinetics, material balances as well as heat balances. This paper investigates the scale-up of the oxygen reactor in the Cu-Cl cycle that is heated by using a half pipe jacket. Half pipe coils can be used for high temperature applications and have many advantages, such as; the high rate of heat transfer due to high turbulence and velocity, the strength of the structure that can reduce the thickness of the inner reactor wall, and the ability to withstand higher pressure. In this paper, the heat balance is investigated for different hydrogen production rates to specify the required size of the oxygen reactor. The thermal resistance of each section of the jacketed oxygen reactor system is investigated to examine the effect of each section on heat balance and identify which one has the dominant contribution on heat transfer. Also, two types of working fluids in the service side of the reactor are examined which are the high pressure helium gas and the molten salt (CuCl). Furthermore, two types of nuclear reactors are investigated as heat sources for the oxygen reactor, which are CANDU SCWR and HTGR. Moreover, the effects of reactor diameter and aspect ratio on the size of the oxygen reactor are studied from the perspectives of heat balance.

## II. MODELING OF HEAT BALANCE IN THE OXYGEN REACTOR

In the oxygen reactor, the total amount of the required heat,  $\dot{Q}$ , is the sum of the reaction heat and the heat required to raise

the reactant temperature from 375°C to 530°C. This amount of heat can be calculated as;

$$\dot{Q} = \Delta H_r \xi + \dot{n}_s \int_{375}^{530} C p_s dT, \quad (1)$$

where  $\Delta H_r$  is the reaction heat in (J/kg.K),  $\xi$  is the extent of reaction in (mol/s),  $\dot{n}_s$  is the mole flow rate of copper oxychloride solid particles in (mol/s) and  $C p_s$  is the specific heat of copper oxychloride solid particles in (J/mol.K) [8]. In the oxygen reactor, the heat flow can be expressed as;

$$\dot{Q} = U A \frac{T_{H_{in}} - T_{H_{out}}}{\ln\left(\frac{T_{H_{in}} - T_c}{T_{H_{out}} - T_c}\right)}, \quad (2)$$

where  $U$  is the overall heat transfer coefficient,  $A$  is the heat transfer area,  $T_{H_{in}}$  and  $T_{H_{out}}$  are the inlet and outlet service side temperatures respectively, and  $T_c$  is the inside reactor temperature which is assumed constant and equal to 530°C. In the oxygen reactor, the total thermal resistance ( $R_t$ ) is the sum of the thermal resistances in each section of the reactor as indicated in Fig. 1, and the overall heat transfer coefficient can be determined from;

$$\frac{1}{UA} = R_t = R_p + R_{FP} + R_W + R_S + R_{FS}, \quad (3)$$

where  $R_p$ ,  $R_{FP}$ ,  $R_W$ ,  $R_S$ , and  $R_{FS}$  are the thermal resistances of the process, fouling process, wall, service and fouling service sections respectively. In (3), the thermal resistance ( $R$ ) of each section can be defined as;

$$R_p = \frac{1}{h_p \pi D_R H_R}, R_{FP} = \frac{f_p}{\pi D_R H_R}, R_W = \frac{\ln\left(\frac{D_R + 2t}{D_R}\right)}{2 \pi k_W H_R}$$

$$R_S = \frac{1}{h_s \pi (D_R + 2t) H_R}, R_{FS} = \frac{f_s}{\pi (D_R + 2t) H_R}$$

where  $h$  is the heat transfer coefficient,  $f$  is the fouling factor,  $D$  is the diameter,  $k$  is the thermal conductivity, and  $H_R$  is the reactor height. The subscripts  $P$ ,  $S$ , and  $W$  denote to the process, service, and wall sections respectively.

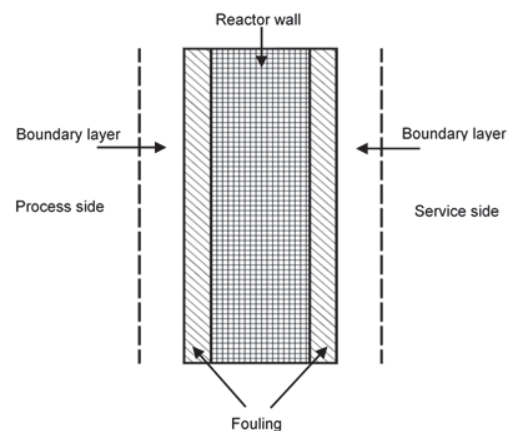


Fig. 1 Thermal resistances through the reactor wall sections

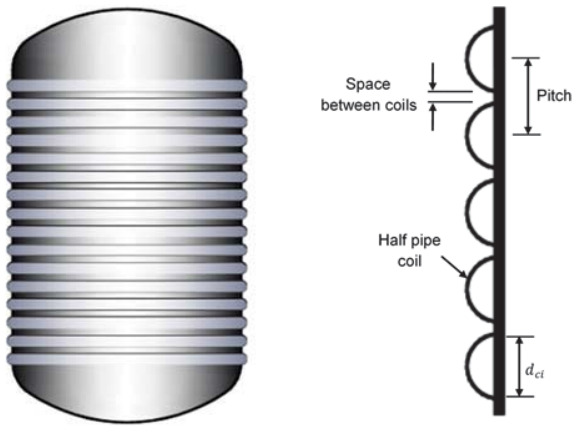


Fig. 2 Schematic diagram of a half pipe jacket

According to the ASME Code, the wall thickness ( $t$ ) (in inches) for cylindrical shell under internal pressure is [9];

$$t = \frac{PR_o}{SE - 0.6P} + t_c, t \leq 0.25D_R, P \leq 0.385 SE, \quad (4)$$

where  $t_c$  is the corrosion allowance in inches,  $R_o$  is the outside radius of the cylindrical shell in inches and  $S$  is the maximum allowable working stress in psi. The value of the joint efficiency,  $E$ , is between 0.6 and 1. The pressure,  $P$  in (4) is the design pressure of the oxygen reactor which is taken here as 1.7 bar greater than the normal total pressure, where it is usual to increase the operating pressure by 10% or 0.69-1.7 bar, whichever is greater. The normal total pressure is the sum of the operating pressure ( $P_o$ ) and the static pressure ( $P_{st}$ ). Thus the design pressure is;

$$P = P_{st} + P_o + 1.7 \text{ (bar)}, \quad (5)$$

where  $\rho$  is the density of the slurry.

In the service side of the oxygen reactor, the central angle of the half-pipe coil jackets used in this paper is  $180^\circ$  (see Fig. 2). The heat transfer coefficient in the service side ( $h_s$ ) is calculated from [10];

For  $Re < 2100$ ;

$$Nu = 1.86 Re^{0.33} Pr^{0.33} \left(\frac{\mu_b}{\mu_w}\right)^{0.14} \left(\frac{D_{ej}}{L_j}\right)^{0.33}, \quad (6)$$

For  $Re > 10000$ ;

$$Nu = 0.027 Re^{0.8} Pr^{0.33} \left(\frac{\mu_b}{\mu_w}\right)^{0.14} \left(1 + 3.5 \frac{D_{ej}}{D_{cj}}\right), \quad (7)$$

where;

$$D_{ej} \text{ (equivalent heat transfer diameter)} = \pi \frac{d_{ci}}{2}, \quad (8)$$

$$D_{cj} \text{ (jacket centerline diameter)} = D_{ji} + \frac{D_{jo} - D_{ji}}{2}, \quad (9)$$

$$L_j \text{ (half pipe coil length)} = n \sqrt{(\pi D_R)^2 + p_j^2}, \quad (10)$$

In (6)-(10),  $Re$  is the Reynolds number,  $Pr$  is the Prandtl number,  $\mu_b$  and  $\mu_w$  are the dynamic viscosities at the bulk and wall temperatures respectively,  $D_{ji}$  and  $D_{jo}$  are the inside and outside jacket diameter respectively,  $d_{ci}$  is the half pipe inner diameter, and  $p_j$  is the pitch of the half pipe coil.

The process side of the oxygen reactor contains different phases of solid particles ( $\text{Cu}_2\text{OCl}_2$ ), molten salt ( $\text{CuCl}$ ) and oxygen gas ( $\text{O}_2$ ). In this study, it is assumed that the oxygen gas will leave the reactor immediately after it is formed, therefore the presence of oxygen gas is neglected. Since the density of the  $\text{Cu}_2\text{OCl}_2$  solid ( $4080 \text{ kg/m}^3$ ) has a value close to that of the  $\text{CuCl}$  molten salt ( $3692 \text{ kg/m}^3$ ), it is assumed that solid particles and molten salt are well mixed and form a homogeneous slurry. This well mixed homogeneous slurry will lead to a more uniform temperature profile inside the oxygen reactor. That is why the temperature profile is assumed to be constant and equal to  $530^\circ\text{C}$ . Thermo physical properties for the slurry mixture are calculated at  $530^\circ\text{C}$ . For agitated reactor, the process side heat transfer coefficient ( $h_p$ ) can be determined from;

$$Nu_D = C Re^a Pr^b \left(\frac{\mu}{\mu_w}\right)^c, \quad (11)$$

where,  $C$  is constant,  $Re$  (Reynolds number) =  $\left(\frac{N_A D_A^2 \rho}{\mu}\right)$ ,  $N_A$  is the agitator speed and  $D_A$  (agitator diameter) =  $D_R/3$ . The values of the constant  $C$  and the indices  $a$ ,  $b$  and  $c$  depend on different factors, such as; agitator type, baffles existence, and whether the heat transfer is to the coils or to the reactor wall. According to the Penny's graph [11] that depends on liquid viscosity and vessel volume, the type of the impeller that can be used for the oxygen reactor is a propeller with 420 rpm. For a baffled reactor, a three blades propeller and heat transfer to reactor wall, the Nusselt number equation is;

$$Nu_D = 0.64 Re^{0.67} Pr^{0.33} \left(\frac{\mu}{\mu_w}\right)^{0.14} (Re > 5000) \quad (12)$$

The slurry dynamic viscosity ( $\mu_{sl}$ ) can be related to the viscosity of the liquid phase as;

$$\mu_{sl} = \mu_r \mu_l, \quad (13)$$

where  $\mu_r$  is the relative dynamic viscosity (dimensionless), and  $\mu_l$  is the dynamic viscosity of the liquid ( $\text{CuCl}$  molten salt). There are different equations to calculate the relative viscosity as a function of the volumetric solid concentration ( $C_s$ ). For very low solid concentrations, Einstein's equation can be used [12], where  $\mu_r = 1$ . For higher solid concentrations, the equation of Guth and Simba [13] can be used;

$$\mu_r = 1 + 2.5 C_s + 14.1 C_s^2 \quad (14)$$

Other thermo physical properties of the slurry mixture can be calculated from the volume percent of the solid and molten salt. For example, the average density of the slurry  $\rho_{sl}$  can be calculated as;

$$\rho_{sl} = \rho_s C_s + \rho_l (1 - C_s), \quad (15)$$

where  $\rho_s$  and  $\rho_l$  are the densities of solid and liquid respectively. The average specific heat of the slurry is;

$$C_{p,sl} = \frac{\rho_s C_{p,s} C_s + \rho_l C_{p,l} (1 - C_s)}{\rho_{sl}}, \quad (16)$$

where  $C_{p,s}$  and  $C_{p,l}$  are the specific heats of the solid and liquid respectively. The effective thermal conductivity of the slurry  $k_{sl}$  is calculated from [14];

$$k_{sl} = k_s C_s + k_l (1 - C_s), \quad (17)$$

where  $k_s$  and  $k_l$  are the thermal conductivities of the solid and liquid respectively.

The recommended method that can be used to determine the effect of fouling, is to calculate the values of individual fouling resistances for the process side  $R_{FP}$  and for the service side  $R_{FS}$ . For helium gas heating fluid,  $R_{FS}$  is expected to be negligible since there should be no build-up associated with clean dry helium.

### III. TYPE OF WORKING FLUID IN THE SERVICE SIDE OF OXYGEN REACTOR

Two types of fluids are highly recommended as a working fluid in the oxygen reactor; CuCl molten salt and high-pressure helium gas (He). Helium gas is recommended amongst the other noble gases because it is inert and relatively has good transport properties [15]. There are substantial differences between molten salts and high-pressure helium that must be considered in selecting the working fluid as a heating medium in the oxygen reactor.

TABLE I  
 COMPARISON OF THERMO-PHYSICAL PROPERTIES OF HELIUM AND CUCL  
 MOLTEN SALT AT AN AVERAGE TEMPERATURE OF 750°C

Material	Molten Salt (CuCl) [8]	Helium (7.5 MPa) [16]
$T_{melting}$ (°C)	430	-
$T_{boiling}$ (°C)	1490	-
$\rho$ (kg/m <sup>3</sup> )	3692	3.5
$C_p$ (kJ/kg.°C)	0.66	5.2
$\rho C_p$ (kJ/m <sup>3</sup> .°C)	2450	18.2
$k$ (W/m.°C)	0.23	0.37
$\mu \times 10^5$ (Pa.s)	260	4.7
$\nu \times 10^6$ (m <sup>2</sup> /s)	0.7	13.4
$Pr$	4.29	0.66

The thermophysical properties for high-pressure helium and CuCl molten salt are summarized in Table I. In this table, it can be seen that the volumetric heat capacity,  $\rho C_p$ , of the CuCl molten salt is greater than that of high-pressure helium gas by more than 100 times. This indicates that the molten salt has higher heat transfer capability than high pressure helium gas. Another advantage of molten salt on helium gas, is that, molten salt requires smaller diameters pipes and less pumping power for transportation than those required for high-pressure helium gas. This advantage will decrease the capital cost and will allow

the process heat to be delivered for large distances. Clean helium clearly does not have the potential to corrode loop materials, whereas, molten salts exhibit higher corrosion rates. Both the molten salt and helium gas provide stored energy sources that can be released rapidly. The stored energy for helium gas, comes from the high pressure of the gas, and the large volume of gas due to the large duct sizes required for transferring helium with reasonable pressure losses. For molten salts, the stored energy comes from the high temperature and high heat capacity of the liquid. This energy can be released if the molten salt mixes with a volatile liquid (e.g. water).

### IV. RESULTS AND DISCUSSION

The total amount of heat required in the oxygen reactor can be calculated from (1), where  $\Delta H_r = 129.162$  kJ/mol,  $\xi = 0.5$  Kmol/day, and  $C_{p,s} = 134$  J/mol.K [8]. For a hydrogen production rate of 100 tonne/day, where  $\dot{n}_s$  is 50000 kmol/day,  $\dot{Q}$  will be 87 MW. For a HTGR, the nuclear reactor exit temperature is about 1000°C. The inlet temperature of the heating fluid in the jacket (same as the exit temperature of the intermediate heat exchanger (IHX)) is taken to be 900°C based on [17]. The exit temperature from the jacket is assumed to be 540°C (because the decomposition temperature is 530°C). For a hydrogen production rate of 100 tonne/day that is divided to a number of reactors  $N$ , the total thermal resistance of the oxygen reactor that is required when using HTGR ( $R_{HTGR}$ ) can be calculated from (2) as;

$$R_{HTGR} = \frac{1}{UA} = 1.15 \times 10^{-6} N \text{ K/W} \quad (18)$$

For CANDU SCWR, where the nuclear reactor exit temperature is about 625 °C [18], [19], the inlet and outlet jacket temperatures are assumed to be about 600 °C and 540 °C respectively, and the total thermal resistance required is  $R_{SCWR} = 3.55 \times 10^{-7} N \text{ K/W}$  for a hydrogen production rate of 100 tonne/day. The physical properties of the heating fluid in the service side of the oxygen reactor (13)-(17) are calculated at the mean temperature of the fluid, which is for a HTGR equal to 720 °C and for a SCWR is equal to 570 °C. For a helium gas with a pressure range of 1-7 MPa and a temperature range of 570-720 °C, the compressibility factor range is 1.001426-1.009594 [16]. This factor varies from unity by less than 1% which means that helium gas can be regarded as an ideal gas. The equation of sound for gases is;

$$c = \sqrt{\gamma RT}, \quad (19)$$

where  $\gamma$  is an adiabatic index and  $R$  is the gas constant. For a helium gas,  $\gamma_{He} = 1.6667$  and  $R_{He} = 2077$  J/kg.K. By using (19), the theoretical maximum speed of a helium gas (which is 1/3 of speed of sound) for a HTGR (T=720°C) is 618 m/s and for a SCWR (T=570°C) is equal to 570 m/s. In this paper, to be more conservative for getting incompressible flow, the operating speed of the helium gas is considered to be 400 m/s in the half pipe jacket for both HTGR and SCWR. The operating speed of the CuCl molten salt is assumed to be 4 m/s for the half pipe

jacket. These values are within the range of molten salt nuclear reactors reported [20], [21]. When the volume of oxygen reactor inventories is very large, a number of parallel reactors with specific volumes can be used. In this case, it is necessary to specify the dimensions of the reactor. The parameters used in the calculations of heat balance are summarized in Table II.

TABLE II  
 DETAILS OF PARAMETERS USED IN THE HEAT BALANCE CALCULATIONS

Parameter	Description
Material of the reactor wall	Stainless steel 321 <sup>a</sup>
Maximum allowable working stress ( <i>S</i> ) at T=649°C	3600 psi [22]
Thermal conductivity ( <i>k<sub>w</sub></i> ) for stainless steel 321 at T=816°C	22.1 W/m.K [23]
Corrosion allowance ( <i>t<sub>c</sub></i> )	8.9 mm <sup>b</sup> [24]
Joint efficiency ( <i>E</i> )	0.8
Internal diameter of the half pipe ( <i>d<sub>ci</sub></i> )	<i>D<sub>R</sub></i> /40
Spacing distance between pipes	<i>d<sub>ci</sub></i> /4
Operating pressure ( <i>P<sub>o</sub></i> ) of the process side	1 bar
Pressure of helium gas in the service side	7.5 MPa
Reactor diameter ( <i>D<sub>R</sub></i> )	4 m
Reactor aspect ratio ( <i>AR</i> )	2

<sup>1</sup> Stainless steels 321 is used because of its high working temperature range from 427 to 816°C.

<sup>b</sup> This value is used for the oxygen reactor because of the relative high corrosion susceptibility of CuCl molten salt.

In order to calculate the thermal resistance due to fouling in the oxygen reactor, fouling factor must be known or estimated. According to Tubular Exchanger Manufacturers Association (TEMA), the fouling factor for molten heat transfer salts is equal to 0.000088 m<sup>2</sup> K/W [25]. In this work, to be more conservative, twice of this value, 0.000176 m<sup>2</sup> K/W, is used to be the fouling factor for molten salt CuCl. Also, it is assumed that the fouling inside oxygen reactor comes from CuCl only, because it represents the major content as a quantity inside the oxygen reactor.

Fig. 3 shows a comparison of the number of reactors required between HTGR and SCWR. This comparison is for each section of the half pipe jacketed oxygen reactor with a helium gas in the service side. From Fig. 3, it can be seen that the maximum number of reactors required comes from the wall of the reactor. This is because of the large thickness of the wall material, which is due to the large size of the reactor and the high static pressure produced inside the reactor. It can also be seen that the number of oxygen reactors for SCWR are higher than that for HTGR by more than three times. This is because of the higher temperature difference between the service and process sides in the HTGR. This means that using HTGR is more efficient for producing heat to the oxygen reactor than SCWR.

Fig. 4 shows the total number of jacketed oxygen reactors with each heat source of nuclear reactors for both helium gas and CuCl molten salt. From this figure, it can be seen that there are no significant differences between high pressure helium gas and molten CuCl in heating the jacketed oxygen reactor. Fig. 5 shows the comparison of number of oxygen reactors versus the hydrogen production rate between material and heat balances

for both HTGR and SCWR for helium gas service fluid. From Fig. 5, it can be seen that the numbers of reactors calculated from heat balance are much greater than that calculated from material balance. That means the design of the oxygen reactor size is affected mainly by the heat balance.

Fig. 6 shows the effect of the reactor diameter and aspect ratio on the number of oxygen reactors calculated from heat balance. From this figure, it can be seen that, when increasing the reactor diameter from 2 m to 4 m, the number of oxygen reactors decreases by 60.5% when *AR* = 2 and decreases by 42.6% when *AR* = 4. Also, it can be seen that, when increasing the aspect ratio from 2 to 4, the number of oxygen reactors decreases by 47.9% when *D<sub>R</sub>* = 2 m and decreases by 24.3% when *D<sub>R</sub>* = 4 m. From these results, it can be concluded that the effect of the reactor diameter on the number of oxygen reactors is more significant than the effect of aspect ratio. Moreover, Fig. 6 shows that, by increasing the diameter or aspect ratio of the reactor, their effects on the number of reactors will decrease.

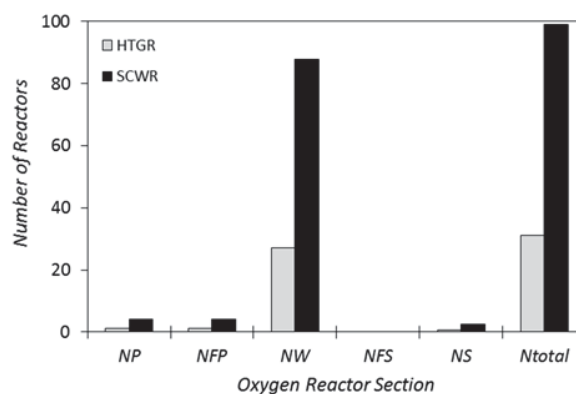


Fig. 3 Number of reactors for each section of jacketed oxygen reactor system heated by 7.5 MPa helium gas for both HTGR and SCWR

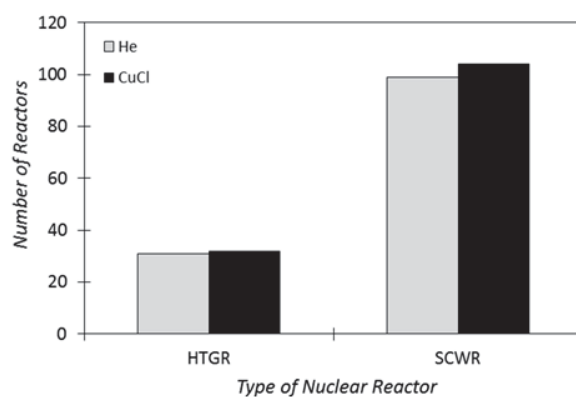


Fig. 4 Total number of jacketed oxygen reactors with each nuclear reactor for both 7.5 MPa helium gas and CuCl molten salt

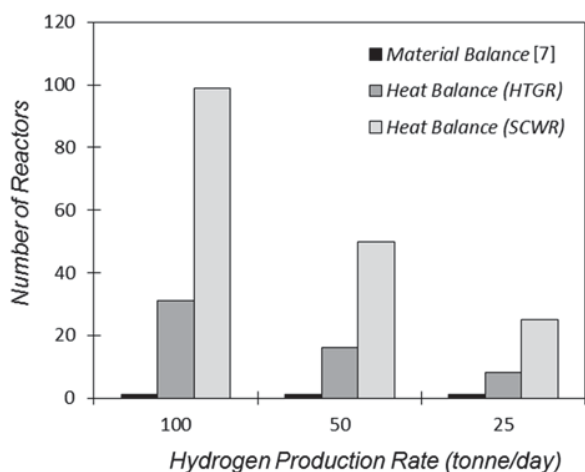


Fig. 5 Number of oxygen reactors calculated by material balance [1] and heat balances of both HTGR and SCWR versus hydrogen production rate

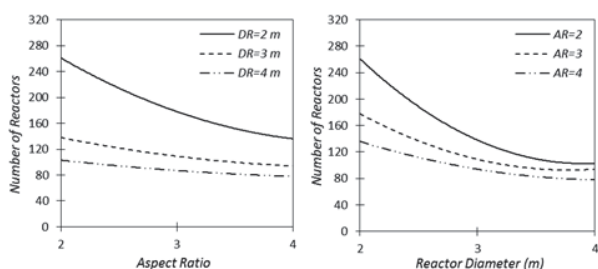


Fig. 6 Effect of reactor diameter and aspect ratio on the number of oxygen reactors calculated from heat balances for a hydrogen production rate of 100 tonne/day

#### V. CONCLUSION

In this paper, the scale up analysis of the jacketed oxygen reactor in the Cu-Cl cycle of hydrogen production, was investigated from the perspective of heat balance. In the heat balance, a half pipe jacket was used to transfer heat to the process side of the oxygen reactor. The thermal resistance of each section in the jacketed reactor was examined, and it was found that the main contribution to the total thermal resistance that dominates the overall heat transfer coefficient is from the wall.

Different types of working fluids were used in the service side such as helium gas and CuCl molten salt and the comparison between them was studied. It was determined that there is no significant difference in the size of the jacketed oxygen reactor when using helium gas or CuCl molten salt. In general, there are technical risks in the use of either helium gas or CuCl molten salt, and both should be retained and studied until these technical issues are resolved and an optimal choice can be made.

The effects of the reactor diameter and aspect ratio on the size of the oxygen reactor were determined from the perspectives of heat balance. It was concluded that the size of the oxygen reactor decreases non-linearly with increasing the reactor diameter or aspect ratio, and the effect of the reactor diameter on the reactor size is more significant than that of the

aspect ratio. The rate of decrease in the size of the oxygen reactor was found to be decreased by increasing the reactor diameter or aspect ratio.

Two types of nuclear reactor heat sources were examined in the scale up analysis of the oxygen reactor, which are CANDU SCWR and HTGR. It was shown that the overall thermal resistance of the oxygen reactor ( $R_t$ ) when using HTGR is less than that of SCWR by 3-4 times. That means a better heat transfer rate has to be provided for SCWR than HTGR, because of the higher exit temperature of the HTGR which in turn produces higher temperature differences between the service and process sides than for the SCWR.

From the comparison between the results of material and heat balances, it was found that the size of the oxygen reactor is determined by the heat balance that requires larger size of the oxygen reactor. The comparison study also indicated that using a jacket for heat transfer, leads to a large number of reactors to provide enough heat transfer area for the large amount of heat required for the decomposition process.

#### REFERENCES

- [1] M. W. Abdulrahman, Z. Wang, and G. F. Naterer, "Scale-up analysis of three-phase oxygen reactor in the Cu-Cl thermochemical cycle of hydrogen production," *EIC Climate Change Technology Conference (CCTC2013)*, Montreal, QC, Canada, 2013.
- [2] M. A. Lewis, M. Serban, and J. K. Basco, "Generating hydrogen using a low temperature thermochemical cycle," in *Proc. of the ANS/ENS Global International conference on Nuclear Technology*, New Orleans, 2003.
- [3] M. Serban, M. A. Lewis, and J. K. Basco, "Kinetic study of the hydrogen and oxygen production reactions in the copper-chloride thermochemical cycle," *American Institute of Chemical Engineers Journal*, Spring National Meeting, New Orleans, LA, pp. 2690-2698. 2004.
- [4] G. D. Marin, "Kinetics and transport phenomena in the chemical decomposition of copper oxychloride in the thermochemical Cu-Cl cycle (Doctoral dissertation)," *University of Ontario Institute of Technology*, Ontario, Canada. 2012.
- [5] G. F. Naterer, K. Gabriel, Z. L. Wang, V. N. Daggupati, and R. Gravelins, "Thermochemical hydrogen production with a copper-chlorine cycle. I: Oxygen release from copper oxychloride decomposition," *International Journal of Hydrogen Energy*, vol. 33, pp. 5439-5450, 2008.
- [6] B. M. Ikeda, and M. H. Kaye, "Thermodynamic properties in the Cu-Cl-O-H system. In *Proc. 7th International Conference on Nuclear and Radiochemistry*, Budapest, Hungary, 2008.
- [7] L. Trevani, "The copper-chloride cycle: synthesis and characterization of copper oxychloride," in *Proc. Hydrogen and Fuel Cells International Conference and Exhibition*, Vancouver, BC, Canada, 2011.
- [8] C. Zamfirescu, I. Dincer, and G. F. Naterer, "Thermophysical properties of copper compounds in copper-chlorine thermochemical water splitting cycles," *International Journal of Hydrogen Energy*, vol. 35, pp. 4839-4852, 2010.
- [9] J. R. Couper, P. W. Roy, J. R. Fair, and S. M. Walas, *Chemical process equipment selection and design*, 2<sup>nd</sup> ed. Gulf Professional Publishing Elsevier Inc., 2005.
- [10] A. K. Coker, *Modeling of chemical kinetics and reactor design*. Houston, Texas, USA, Gulf Professional Publishing, 2001.
- [11] W. R. Penny, "Guide to trouble free mixers," *Chemical Engineering Journal*, vol. 77, no. 12, p. 171, 1970.
- [12] A. Einstein, "A new determination of molecular dimensions (Eine neue Bestimmung der Moleküldimensionen)," *Annalen der Physik*, vol. 19, pp. 289-306, 1906.
- [13] E. Guth, and H. Simba, "Viscosity of suspensions and solutions: III Viscosity of sphere suspensions," *Kolloid-Z*, vol. 74, pp. 266-275, 1936.
- [14] K. Boomsma, and D. Poulikakos, "On the effective thermal conductivity of a three-dimensionally structured fluid-saturated metal foam," *International Journal of Heat and Mass Transfer*, vol. 44, pp. 827-836, 2001.

- [15] M. S. El-Genk, and J. Tournier, "On the use of noble gases and binary mixtures as reactor coolants and CBC working fluids," *Energy Conversion and Management*, vol. 49, pp. 1882-1891, 2008.
- [16] H. Petersen, "The properties of helium: Density, specific heats, viscosity, and thermal conductivity at pressures from 1 to 100 bar and from room temperature to about 1800 K," *Risø Report No. 224*. Danish atomic energy commission research establishment Risø, 1970.
- [17] K. Natesan, A. Moisseytsev, S. Majumdar, and P. S. Shankar, "Preliminary issues associated with the next generation nuclear plant intermediate heat exchanger design," *ANL/EXT-06/46*. Argonne National Laboratory, 2006.
- [18] C. K. Chow, and H. F. Khartabil, "Conceptual fuel channel designs for CANDU – SCWR," *Nuclear Engineering and Technology, Special issue on the 3rd international symposium on SCWR*, vol. 40, no. 2, 2007.
- [19] R. B. Duffey, and L. Leung, "Advanced cycle efficiency: generating 40% more power from the nuclear fuel," *World Energy Congress (WEC)*, Montreal, Canada, 2010.
- [20] J. P. Caire, and A. Roure, "Pre design of a molten salt thorium reactor loop," *Excerpt from the Proceedings of the COMSOL Users Conference Grenoble*, 2007.
- [21] W. R. Huntley, and M. D. Silverman, "System design description of forced-convection molten-salt corrosion loops MSR-FCL-3 and MSR-FCL-4," *Oak Ridge National Laboratory, Union Carbide Corporation for the Energy Research and Development Administration, Engineering Technology Division*, 1976.
- [22] E. J. Rozic, "Elevated temperature properties as influenced by nitrogen additions to types 304 and 316 austenitic stainless steels," *ASTM International, Steel, Stainless*, 1973.
- [23] P. A. Schweitzer, *Encyclopedia of corrosion technology*, 2<sup>nd</sup> ed. New York, Marcel Dekker, Inc., 2004.
- [24] K. Coker, *Ludwig's applied process design for chemical and petrochemical plants*, 4<sup>th</sup> ed. Gulf Professional Publishing, Burlington, Massachusetts, USA, 2007.
- [25] J. M. Chenoweth, "Final report of the HTRI/TEMA joint committee to review the fouling section of the TEMA standards," *Heat Transfer Engineering*, vol. 11, no. 1, pp. 73-107, 1990.