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Heat Transfer Analysis of a Multiphase Oxygen Reactor Heated by a Helical Tube in the Cu-Cl Cycle of a Hydrogen Production

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Abstract**—**In the thermochemical water splitting process by Cu-Cl cycle, oxygen gas is produced by an endothermic thermolysis process at a temperature of 530°C. Oxygen production reactor is a three-phase reactor involving cuprous chloride molten salt, copper oxychloride solid reactant and oxygen gas. To perform optimal performance, the oxygen reactor requires accurate control of heat transfer to the molten salt and decomposing solid particles within the thermolysis reactor. In this paper, the scale up analysis of the oxygen reactor that is heated by an internal helical tube is performed from the perspective of heat transfer. A heat balance of the oxygen reactor is investigated to analyze the size of the reactor that provides the required heat input for different rates of hydrogen production. It is found that the helical tube wall and the service side constitute the largest thermal resistances of the oxygen reactor system. In the analysis of this paper, the Cu-Cl cycle is assumed to be heated by two types of nuclear reactor, which are HTGR and CANDU SCWR. It is concluded that using CANDU SCWR requires more heat transfer rate by 3-4 times than that when using HTGR. The effect of the reactor aspect ratio is also studied and it 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. Comparisons between the results of this study and pervious results of material balances in the oxygen reactor show that the size of the oxygen reactor is dominated by the heat balance rather than the material balance.

Keywords**—**Heat transfer, Cu-Cl cycle, hydrogen production, oxygen, clean energy.

I. Introduction

COPPER-CHLORINE (Cu-Cl) cycle is considered as one of the promising lower temperature cycles used for the sustainable production of hydrogen in large quantities [1], [2]. It can be linked with clean sources of heat such as nuclear reactors or solar sources to achieve lower environmental impact and lower costs of hydrogen production than existing technologies. The Cu-Cl cycle consists of three reactions, two thermal and one electrochemical. The three reaction steps of the Cu-Cl cycle are [3];

Step 1: $CuCl(a) + 2HCl(g) \rightarrow CuCl_2(a) + H_2(g)$

Step 2: $2CuCl_2(s) + H_2O(g) \leftrightarrow Cu_2OCl_2(s) + 2HCl(g)$

Step 3: $Cu_2OCl_2\ (s) \rightarrow 2CuCl\ (l) + ½\ O_2\ (g)$

where a, s, l and g denote to aqueous, solid, liquid and gas respectively. The reaction steps of the Cu-Cl cycle have been explained in details in [3] and [4].

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The continuous stirred tank reactor (CSTR) is a multiphase reactor that can be used for the multiphase reactions that have fairly high reaction rates such as oxygen production reaction in the Cu-Cl cycle [4]. In CSTR, heating can be achieved by using an internal helical tube that is immersed inside the oxygen reactor.

In the literature, numerous studies have been undertaken on hydrogen production using Cu-Cl cycles. Serban et al. [2] have studied the production of oxygen from the decomposition of an equimolar mixture of CuO and $CuCl_2$ in a vertical reactor at 500°C. They have concluded that the evolution of oxygen gas is limited by the decomposition of $CuCl_2$, and the decomposition is not completed until it reaches 600°C. Ikeda and Kaye [5] and Trevani et al. [6] have developed methods of synthesis to study the thermochemical properties of copper oxychloride (Cu_2OCl_2), because of its commercial unavailability. Ikeda and Kaye [5] have used stoichiometric amounts of CuO and $CuCl_2$ instead of Cu_2OCl_2. Trevani et al. [6] have used a more easily scalable method that can be used to produce large amounts of copper oxychloride. Orhan has studied Aspen Plus simulations of the copper oxychloride decomposition process in the Cu-Cl cycle. In his simulations, he has predicted the behavior of process reactions and steps using standard engineering relationships, mass and energy balances, as well as phase and chemical equilibrium data [7].

Abdulrahman et al. [8] have investigated the material balance of the oxygen reactor in the Cu-Cl cycle and calculated the size of the oxygen reactor for different hydrogen production rates and different residence times. They have also studied the factors that affect the size of the oxygen reactor, such as; solid particles characteristics (size, shape and concentration), oxygen bubbles, reactor heating rate and fouling. Abdulrahman [4] has examined the scale-up of the multiphase thermolysis oxygen reactor in the Cu-Cl cycle that is heated by using a half pipe jacket. He has concluded that using a jacket for heat transfer in the oxygen reactor leads to a large number of reactors, because of the high thermal resistance of the reactor wall that is due to the large thickness of the wall.

Helical tubes that have thinner walls because of their smaller tube diameter, are used widely for different applications. A review of heat transfer through helical tubes has been performed by Pawar [9]. His review was in terms of various dimensionless numbers, friction factors, different coil curvature ratios, fluid types, validity, and effect of geometry, as well as effect of laminar and turbulent flow on heat transfer rate. He has concluded that most of the previous studies have developed correlations based on experimental data, and these correlations are applicable either to work within acceptable range only or limited range of parameters.

In this paper, the thermal scale-up analysis of the multiphase thermolysis oxygen reactor is studied by using an internal helical tube to transfer the heat required for the endothermic decomposition reaction inside the reactor. Helical tubes have many advantages such as; compact size, low cost, large surface area, and passive heat transfer enhancement techniques due to high turbulence and velocity. Fig. 1 shows a schematic diagram for the system of the oxygen reactor with the internal helical tube. In this paper, heat balances of the oxygen reactor system with a helical tube, are examined for different hydrogen production rates. The effect of the thermal resistance of each section in the system on the heat balance is calculated to identify which one has the dominant contribution on heat transfer. Also, the effect of the service side fluid type on heat transfer is investigated by studying two types of fluids which are the high pressure helium gas and the molten salt (CuCl). Furthermore, the effect of the heat source type on heat transfer in the oxygen reactor is determined by investigating two types of nuclear reactor heat sources such as; CANDU SCWR and HTGR. Moreover, the effect of the oxygen reactor aspect ratio on the size of the oxygen reactor is studied from the perspective of heat balance.

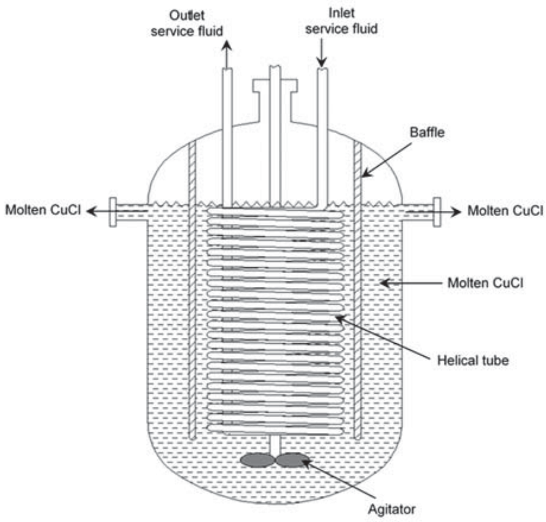

Fig. 1 Schematic diagram of the oxygen reactor with an internal helical tube

II. Modeling of Heat Balance in the Oxygen Reactor

The total amount of the heat required for the decomposition process in the oxygen reactor ($\dot{Q}$), 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;

$$\dot{Q} = \Delta H_r \, \dot{\xi} + \dot{n}_s \int_{375}^{530} Cp_s \, dT, \tag{1}$$

where ΔH_r is the reaction heat in (J/kg.K), $\dot{\xi}$ 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 Cp_s is the specific heat of copper oxychloride solid particles in (J/mol.K). In the oxygen reactor, the temperature inside the reactor (T_c) is assumed to be constant and equal to 530°C. Therefore, there is no effect of heat transfer configuration (parallel or counter flow) in the equation of the log mean temperature difference, and the heat flow can be expressed as;

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

where U is the overall heat transfer coefficient, A is the heat transfer area, and $T_{H_{in}}$ and $T_{H_{out}}$ are the inlet and outlet service side temperatures respectively. The term UA in (2), can be calculated from the total thermal resistance (R_t) as;

$$UA = \frac{1}{R_t}, \tag{3}$$

where R_t is the sum of the thermal resistances in each section of the reactor as indicated in Fig. 2, and is written as;

$$R_t = R_P + R_{FP} + R_W + R_S + R_{FS}, \tag{4}$$

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. Table I shows the thermal resistances equations for each section of the oxygen reactor system. In Table I, h is the heat transfer coefficient, f is the fouling factor, d_{hi} and d_{ho} are the inside and outside diameters of the helical tube, L_h is the length of the helical tube, and k is the thermal conductivity. The subscripts P, S, and W denote to the process, service, and wall sections respectively.

TABLE I
Thermal Resistances Equations for each Section of the Oxygen Reactor System with Internal Helical Tube

R_W	R_{FP}	R_{FS}	R_S	R_P
$\frac{\ln\left(\frac{d_{ho}}{d_{hi}}\right)}{2\,\pi\,k_w\,L_h}$	$\frac{f_P}{\pi\,d_{ho}\,L_h}$	$\frac{f_S}{\pi\,d_{hi}\,L_h}$	$\frac{1}{h_S\,\pi\,d_{hi}\,L_h}$	$\frac{1}{h_P\,\pi\,d_{ho}\,L_h}$

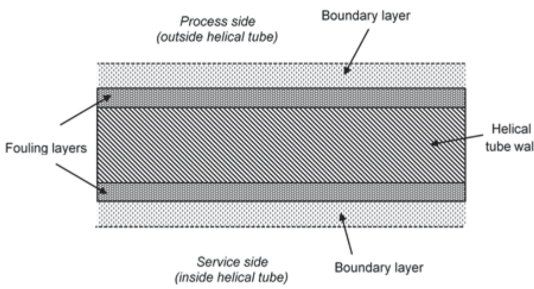

Fig. 2 Thermal resistances through the helical tube wall sections

According to the ASME Code, the wall thickness (t) (in inches) for a cylindrical pipe under internal pressure is [10];

$$t = \frac{PR_o}{SE - 0.6P} + t_c\ ,t \leq 0.25D_R, P \leq 0.385\ SE, \quad (5)$$

where t_c is the corrosion allowance in inches, R_o is the outside radius of the cylindrical tube in inches and S is the maximum allowable working stress in psi, P is the design pressure, and E is the joint efficiency, where its value ranges between 0.6 and 1. To calculate the design pressure, the operating pressure is usually increased by 10% or 0.69-1.7 bar, whichever is greater. Therefore, the design pressure, P in (5) is taken as 1.7 bar greater than the normal total pressure. The normal total pressure is the sum of the operating pressure (P_o) and the static pressure (P_{st}). Thus the design pressure is;

$$P = P_{st} + P_o + 1.7\ (bar), \quad (6)$$

where ρ is the density of the slurry.

In the service side, the helical coil length, L_c that is needed to make n turns is [11];

$$L_c = n\sqrt{(\pi D_c)^2 + p_c^2}\ , \quad (7)$$

where p_c is the pitch of the coil and D_c is the diameter of the helical coil. The volume occupied by the coil, V_c is;

$$V_c = (\pi/4)\ d_{co}^2\ L_c\ , \quad (8)$$

where d_{co} is the outside diameter of the helical tube. The total volume available for the oxygen reactor will be;

$$V'_{\text{total}} = V_{\text{total}} - V_c, \quad (9)$$

where V_{total} is the effective total volume of the reactor calculated from material balance. The shell-side equivalent diameter of the coiled tube is;

$$D_e = \frac{4\ V'_{\text{total}}}{\pi\ d_{co}\ L_c}. \quad (10)$$

Nusselt number inside the helical tube can be calculated by using the Sieder Tate correlation for pipe flow heat transfer [12], [13];

$$Nu_S = 0.027\ Re^{4/5}\ Pr^{1/3}\left(\frac{\mu}{\mu_w}\right)^{0.14}\left(1 + 3.5\frac{d_{ci}}{D_{cHC}}\right), \quad (11)$$

where; $0.7 < Pr < 16{,}700$ and $Re > 10^4$. In (11), Re is the Reynolds number, Pr is the Prandtle number, μ and μ_w are the dynamic viscosities at the bulk and wall temperatures respectively, d_{ci} is the helical tube inner diameter, and D_{cHC} is the center line diameter of the helical coil. In this paper, two types of fluids are examined as a working fluid in the oxygen reactor; CuCl molten salt and high-pressure helium gas (He). Abdulrahman [4] has discussed in details the substantial differences between CuCl molten salt and high-pressure helium gas that must be considered in selecting the working fluid as a heating medium in the oxygen reactor.

The process side of the oxygen reactor contains different phases of solid particles (Cu_2OCl_2), molten salt (CuCl) and oxygen gas (O_2). Since the density of the oxygen gas is significantly less than that of the molten CuCl, it is assumed in this study that the oxygen gas will leave the reactor immediately after it is formed. Therefore, the presence of the oxygen gas is neglected. Also, it is assumed that the solid particles and molten salt are well mixed and form a homogeneous slurry, because the difference in densities between them is small, where the density of the Cu_2OCl_2 solid is 4080 kg/m^3 and that of the CuCl molten salt is 3692 kg/m^3. This well mixed homogeneous slurry has a more uniform temperature profile inside the oxygen reactor. Therefore, the temperature profile is assumed to be constant and equal to 530^{o}C. The correlation describing the Nusselt number for heat transfer to the slurry in the oxygen reactor with mechanical agitation and heated by submerged helical coil is as [14];

$$Nu_P = 0.87\ Re^{0.62}\ Pr^{0.33}\left(\frac{\mu}{\mu_w}\right)^{0.14}, \quad (12)$$

where 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 dynamic viscosity of the slurry (μ_{sl}) is expressed in terms of the liquid dynamic viscosity (μ_l) as;

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

where μ_r is the relative dynamic viscosity (dimensionless). The relative viscosity can be calculated from the volumetric solid concentration (C_s). According to Einstein [15], when the solid concentration is very low, its effect can be neglected and $\mu_r = 1$. For higher solid concentrations, the equation of Guth and Simba [16] can be used;

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

Other thermophysical properties of the slurry mixture can be calculated from the volume percent of the solid and molten salt as;

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

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

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

where ρ, C_p and k are the density, specific heat and thermal conductivity respectively. The subscripts s and l represent the solid and liquid phases respectively. In the calculations of the thermal resistances due to the fouling effects, R_{FS} is expected to be negligible since there should be no build-up associated with clean dry helium.

III. Results and Discussion

Table II summarizes the parameters used in the calculations of heat balance. For a hydrogen production rate of 100 tonne/day, where $\dot{n}_s$ is 50000 kmol/day [8], the total amount of heat required in the oxygen reactor ($\dot{Q}$) is calculated from (1) to be 87 MW, and the total thermal resistance of the oxygen reactor that is required when using HTGR (R_{HTGR}) can be calculated from (2) as;

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

where N is the number of reactors. For CANDU SCWR, the total thermal resistance required is $R_{SCWR} = 3.55 \times 10^{-7}\ N$ K/W for a hydrogen production rate of 100 tonne/day. The physical properties of the heating fluid inside the helical tube 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.

TABLE II
DETAILS OF PARAMETERS USED IN THE HEAT BALANCE CALCULATIONS

Parameter	Description
ΔH_r	129.3 kJ/mol [17]
$\dot{\xi}$	0.5 Kmol/day
Cp_s	134 J/mol.K [18]
T_c	530°C [4]
$T_{H_{out}}$	540°C [4]
$T_{H_{in}}$ (HTGR)	900°C [4], [17]
$T_{H_{in}}$ (SCWR)	600°C [4]
Material of the reactor wall	Stainless steel 321[a]
Maximum allowable working stress (S) at T=649°C	3600 psi [19]
Thermal conductivity (k_w) for stainless steel 321 at T= 816°C	22.1 W/m.K [20]
Corrosion allowance (t_c)	8.9 mm[b] [21]
Joint efficiency (E)	0.8
Operating pressure (P_o) of the process side	1 bar
Pressure of helium gas in the service side	7.5 MPa
Reactor diameter (D_R)	4 m
Reactor aspect ratio (AR)	2
Helical tube diameter (D_c)	$D_R/2$
Inside helical pipe diameter (d_i)	$D_R/30$
Helical tube pitch (p_c)	1.5 d_{co}
Fouling factor of process side (f_P)	0.000176 m^2 K/W [4]
operating speed of the helium gas in the service side	400 m/s [4]
operating speed of the CuCl in the service side	4 m/s [4]

[a] Stainless steels 321 is used because of its high working temperature range from 427 to 816°C.
[b] This value is used for the oxygen reactor because of the relative high corrosion susceptibility of CuCl molten salt.

Since the dimensions of the helical tube are calculated relative to the reactor diameter, it is expected that the speed of the fluid in the service side will be more than the limit that is specified previously (see Table II). In order to keep the values of the helical tube dimensions and speeds of the fluids inside the helical tube as constants, different numbers of reactors are needed to carry the required amount of heat that is necessary for oxygen reactor decomposition process. In this case, there are two different numbers of reactors, one is calculated from the heat balance by calculating the thermal resistances and the other is calculated from the amount of heat by fixing the dimensions of the helical tube and the speeds of the fluids inside the helical tube. Then, the biggest value of the number of reactors is considered in the results. For mass production of hydrogen, it is necessary to use a number of parallel reactors with specific dimensions.

Table III shows the values of the numbers of oxygen reactors with helical tubes required for each section of the oxygen reactor for different heating fluids and different nuclear reactors. From this table, it can be shown that the least number of reactors can be obtained when using CuCl molten salt inside the helical tube for both HTGR and SCWR. Fig. 3 shows a comparison of the number of reactors required between HTGR and SCWR. This comparison is for each section of the oxygen reactor system with a CuCl molten salt. From Table III and Fig. 3, it can be seen that the maximum number of reactors required comes from both the wall of the helical tube and the service side inside the helical tube. When the pressure inside the helical tube is 2MPa, the thermal resistance of the service side is higher than that of the helical tube wall, but when the pressure inside the tube is increased to 7.5MPa, the thickness of the tube has to be increased which leads to a higher thermal resistance for the tube wall and the thermal resistance inside the tube decreases because of the higher pressure. It can also be seen from Table III and Fig. 3 that the number of oxygen reactors for HTGR is less than that for SCWR by more than three times. This is due to the higher service fluid temperature in the HTGR. In this way, using HTGR is considered more efficient than SCWR in producing heat to the oxygen reactor.

Fig. 4 displays the total number of oxygen reactors with helical tubes of each nuclear reactor for both 7.5MPa helium gas and CuCl molten salt. From this figure, it can be seen that the number of reactors using 7.5MPa helium gas is higher, by more than 40%, than that using molten CuCl as a heating fluid inside the helical tube. Fig. 5 compares the number of oxygen reactors with helical tubes versus the hydrogen production rate between material balance [8] and heat balances for both HTGR and SCWR for a residence time of 2 hours and for CuCl molten salt service fluid. From Fig. 5, it is shown that the numbers of reactors calculated from heat balance by using internal helical tubes with CuCl molten salt, is higher than that calculated from material balance by more than two times for HTGR and more than eight times for SCWR. That means the design of the oxygen reactor size with the internal helical tube is affected mainly by the heat balance.

Fig. 6 compares the number of reactors for each section of the oxygen reactor system between the half pipe jacket system serviced by 7.5MPa helium gas [4] and the internal helical tube system serviced by CuCl molten salt. This comparison has been performed for HTGR heat source and a hydrogen production rate of 100 tonne/day. This figure shows that there are no significant differences in the number of reactors for each section of oxygen reactor except for the wall section, where the number of half pipe system is higher, by more than eight times, than that of internal helical tube. In addition, Fig. 6 shows that the total number of reactors needed by using internal helical tube with CuCl molten salt is less, by more than three times, than that needed by using half pipe jacket system with 7.5MPa helium gas.

Fig. 7 shows the number of reactors versus reactor aspect ratio. From this figure, it can be seen that the number of reactors decreases with increasing the aspect ratio. It is also shown that the rate of decrease in the number of reactors decreases with increasing the aspect ratio. The number of reactors decreases by approximately 24.2% when increasing the aspect ratio from 2 to 3, while the number of reactors decreases by approximately 9.5% when increasing the aspect ratio from 4 to 5.

TABLE III
NUMBER OF REACTORS FOR EACH SECTION OF THE OXYGEN REACTOR SYSTEM FOR DIFFERENT HEATING FLUIDS AND NUCLEAR REACTORS

Nuclear reactor type	Fluid type	P (MPa)	N_P	N_{FP}	N_W	N_{FS}	N_S	N_{total}
HTGR	He	2	1.3	0.86	4.6	0	7.3	15
		7.5		0.7	8.5		2.1	13
	CuCl	0.1		0.9	3.2	1.1	1.8	9
SCWR	He	2	4.6	2.8	14.9	0	37.3	60
		7.5		2.3	27.6		10.6	46
	CuCl	0.1		3	10.2	3.5	10.9	33

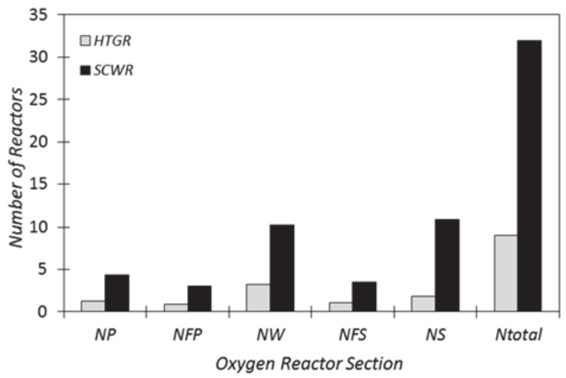

Fig. 3 Number of reactors for each section of the oxygen reactor system heated by CuCl molten salt for both HTGR and SCWR

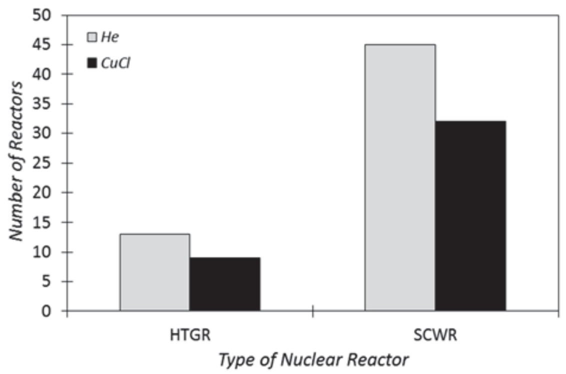

Fig. 4 Total number of oxygen reactors of each nuclear reactor for both 7.5 MPa helium gas and CuCl molten salt

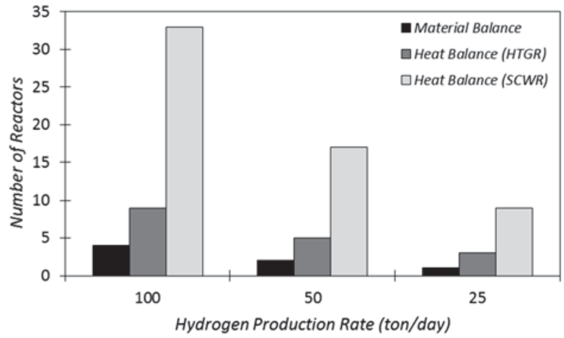

Fig. 5 Number of oxygen reactors calculated by material balance [8] and heat balances of both HTGR and SCWR versus hydrogen production rate for CuCl molten salt and residence time of 2 hours

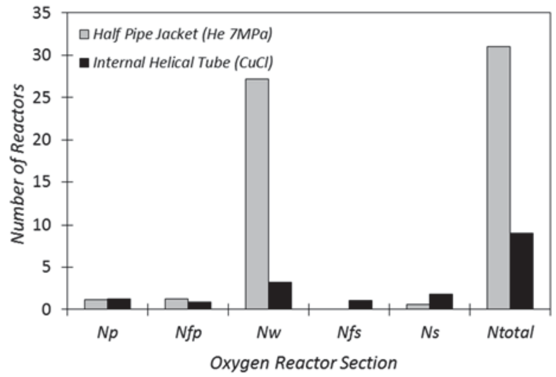

Fig. 6 Comparison of number of reactors for each section of oxygen reactor system between the half pipe jacket with 7.5 MPa helium gas [4] and the internal helical tube with CuCl molten salt for HTGR, hydrogen production rate of 100 tonne/day and a residence time of 2 hours

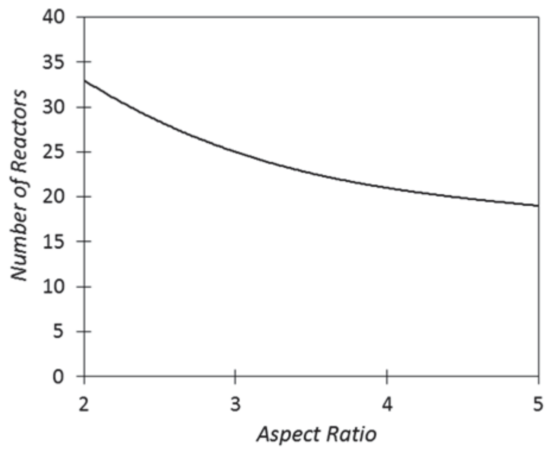

Fig. 7 Effect of reactor aspect ratio on the number of oxygen reactors for a molten CuCl, SCWR, and a hydrogen production rate of 100 tonne/day

IV. CONCLUSION

This paper is aimed at making a contribution in the development of the thermochemical Cu-Cl cycle for hydrogen production. For this purpose, the scale-up analysis of the multiphase oxygen production reactor was examined from the perspective of heat transfer. This research has mainly investigated the problem of providing the required heat for the thermal decomposition process inside the oxygen reactor that is driven by nuclear reactors, by using an internal helical tube. The thermal resistance of each section in the oxygen reactor system was investigated, and it was found that the main contributions to the total thermal resistance that dominate the overall heat transfer coefficient are from the tube wall and the service side.

Two kinds of heat sources, which are HTGR and CANDU SCWR, were examined in the heat transfer analyses of the oxygen reactor. It was found 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. The type of the working fluid used inside the helical tube was also studied in this paper, and it was determined that using CuCl molten salt in the service side is thermally better than using helium gas because of the reduction in the number of oxygen reactors.

The results of heat transfer analyses of the oxygen reactor by using an internal helical tube were compared with the previous results of material balances, as well as the results of heat transfer analyses by using a half pipe jacket. It was found that the size of the oxygen reactor is determined by the heat balance rather than the material balance, because of the larger size that is required in the heat balance. Also, it was shown that using an internal helical tube inside the oxygen reactor is more efficient than using a half pipe jacket in transferring heat to the oxygen reactor. The effect of the reactor aspect ratio was also investigated, and it was found that the number of reactors decreases when the aspect ratio increases, and the rate of decrease in the number of reactors decreases by increasing the aspect ratio.

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