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DOUBLE PHASE COAXIAL GAS HEATER FOR MOLTEN CARBONATE FUEL CELL

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ABSTRACT

An original small size, modular molten carbonate fuel cell (MCFC) has been patented (patent n°: PG 2003 A 0019) by researchers of the University of Perugia. The proposed MCFC may be conveniently employed for µCHP residential applications because of:

- high efficiency;
- long life;
- natural gas supplying.

Because of the small size and compact geometry of the proposed MCFC stack, a particular system for natural gas and water introduction is required: natural gas must be heated (stack temperature is 650°C) and mixed with steam in order to attain anodic humidification which is very important to improve MCFC working life [1].

In this work, a double fluid (water, natural gas), phase transition heat exchanger which is suitable to stack geometrical constraints is proposed and studied; it is composed by two coaxial pipes. Two different solutions have been analyzed with different steam and fuel gas arrangements. By formulating and solving an original equations system, the geometrical configuration that minimizes the exchanger dimension has been identified.

INTRODUCTION

The MCFC electrolyte is a molten carbonate salt mixture, usually lithium carbonate and potassium carbonate. The electrolyte is kept in ceramic matrix. Cell operating temperatures range is 600-800°C. High-temperature molten carbonate fuel cells can be fed by hydrogen attained by a variety of hydrocarbons using either an internal or external reformer. MCFC can tolerate higher amounts of carbon monoxide with respect to low temperature fuel cells, which makes natural gas more attractive for this type of fuel cell [1].

In this work a double fluid phase transition heat exchanger for an original small size and internal reformer MCFC is studied. Because of the peculiar MCFC stack geometry water and gas natural introduction must be carried out by two coaxial pipes. Pipes are soaked into uniform temperature environment: MCFC anode compartment. Heat exchanger aim is to:

- warm methane up to 650°C;
- vaporize water;
- warm steam up to 650°C;
- mix gases (methane and steam) for reforming.

By formulating and solving an original equations system, the geometrical configuration that minimizes the exchanger dimension has been identified.

THEORETICAL ANALYSIS

In order to fit small size fuel cell dimensions, the only geometrical solution allowed is a single pipe where heat exchange occurs. A coaxial pipe solution is adopted; the pipe is made up by two sections:

- the first section is constituted by two coaxial pipes where fluids flow separately, one inside internal pipe and the other inside external pipe;
- the second section is constituted by a single pipe where two fluids, both in gas phase, are mixed.

In the first section, heat is absorbed in part by fluid that flows in the external annular section, in part is transmitted to inner pipe fluid. Coaxial pipe characteristics are sketched in Fig. 1.

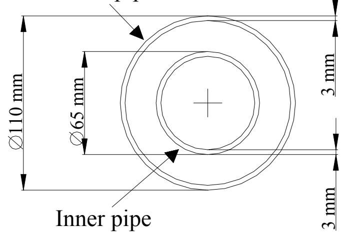

Figure 1: Coaxial pipe characteristics.

Two different gas arrangements have been analyzed:

- Case 1: water in the outer pipe and methane in the inner pipe;
- Case 2: methane in the outer pipe and water in the inner one.

Case 1 analysis

In Fig. 2 the coaxial pipe scheme is sketched; three segments may be observed:

- segment L1, where water heats from room temperature (section 0) up to saturated liquid condition at 100°C (section 1);
- segment L2, where water phase change takes place (section 2);
- L3 segment, where mixture (CH_4 + H_2O) is heated up to 650°C.

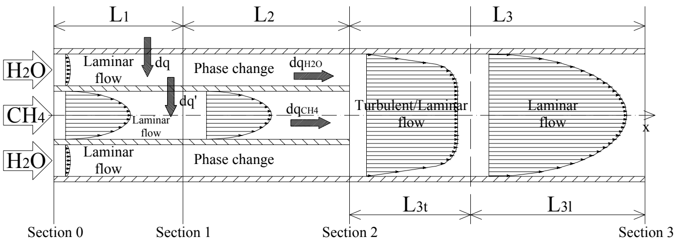

Figure 2: Coaxial pipe scheme.

Heat balance has been found assuming the following hypotheses:

1. steady state: fluid speed and temperature are constant in each point;
2. fluid temperature is uniform along each section;
3. environmental temperature is uniform and constant;
4. density ρ, specific heat C_P and fluid conductivity are temperature independent;
5. pipes material is homogeneous and isotropous;
6. no chemical reactions occur;
7. thin pipe walls;
8. radiative heat transfer rate is negligible.

The infinitesimal heat flux transmitted from water to gas through inner pipe wall is given by:

$$dq' = H' \cdot \left(T_{H2O} - T_{CH4}\right) \cdot 2\pi r_i \cdot dx \tag{1}$$

Similarly, heat transmitted by outer environment to water through outer pipe wall is:

$$dq = H \cdot \left(T_f - T_{H2O}\right) \cdot 2\pi r_e \cdot dx \tag{2}$$

dq is partially absorbed by H_2O (dq_{H2O}) and partially absorbed by methane (dq_{CH4}). In L1 section differential heat fluxes are respectively:

$$dq_{H2O} = C_{PH2O} \cdot m_{H2O} \cdot dT_{H2O} \tag{3}$$

$$dq_{CH4} = C_{PCH4} \cdot m_{CH4} \cdot dT_{CH4} \tag{4}$$

In L2 segment the differential heat flux rate absorbed by methane is given by (4), while the heat flux absorbed by water is:

$$q_{H_2O} = m_{H2O} \cdot r^* \tag{5}$$

For hypothesis 1) along L1 and L2 segments the following identities are considered:

$$dq = dq_{H2O} + dq' \tag{6}$$

$$dq' = dq_{CH4} \tag{7}$$

In L1 segment, both fluids temperatures vary; substituting (1), (2), (3) and (4) into eq. (6) and (7), the followings first order differential equations are obtained:

$$\begin{cases} \dfrac{dT_{CH4}}{dx} = \dfrac{H_1' 2\pi r_i}{C_{P1CH4} m_{CH4}} \cdot (T_{H2O} - T_{CH4}) \\ \dfrac{dT_{H2O}}{dx} = \dfrac{H_1 2\pi r_e}{C_{P1H2O} m_{H2O}} \cdot (T_f - T_{H2O}) - \dfrac{H_1' 2\pi r_i}{C_{P1H2O} m_{H2O}} \cdot (T_{H2O} - T_{CH4}) \end{cases} \tag{8}$$

Given wall transmittances H_1 and H_1' and pipes radiuses r_i, r_e, $T_{H2O}(x)$ and $T_{CH4}(x)$ can be obtained by solving (8). L1 length is calculated by imposing water temperature T^*_{H2O} = 100°C in Section 1. In L2 only methane temperature is x dependent while water temperature is constant because of phase change. The energy balance become:

$$H_2' \cdot (T^*_{H2O} - T_{CH4}) \cdot 2\pi r_i dx = C_{P2CH4} \cdot m_{CH4} \cdot dT_{CH4} \tag{9}$$

solving previous expression with respect to $T_{CH4}(x)$:

$$T_{CH4}(x) = T^*_{H2O} - (T^*_{H2O} - T_{CH4/x=L_1}) \cdot e^{-\frac{H_2' 2\pi r_i}{C_{PCH4} m_{CH4}} x} \tag{10}$$

where terms $T_{CH4/x=L1}$ is given by eq. (8) solution. To calculate L2 length heat balance (6) has been employed:

$$H_2 (T_f - T^*_{H2O}) 2\pi r_e dx = m_{H2O} r^* + H_2' (T^*_{H2O} - T_{CH4}) 2\pi r_i dx \tag{11}$$

Substituting $T_{CH4}(x)$ into (11) and integrating between section 1 (x = 0) and 2 (x = L_2) an equation with an only unknown (L_2) has been obtained:

$$\int_0^{L_2} 2\pi r_e H_2 \left(T_f - T^*_{H2O}\right) dx + \int_0^{L_2} H_2' \left(T^*_{H2O} - T_{CH4/x=L_1}\right) e^{-\frac{H_2' 2\pi r_i}{C_{P2CH4} m_{CH4}} x} 2\pi r_i = m_{H2O} r^* \tag{12}$$

L_2 has been numerically found.

In L3 segment, the heat flux dq_3 that is transferred through pipe wall causes mixture temperature growing. dq_3 and dq_M are given by:

$$dq_3 = H_3 \cdot (T_f - T_M) \cdot 2\pi r_e \cdot dx \quad (13)$$

$$dq_M = C_{PM} \cdot m_M \cdot dT_M \quad (14)$$

Since hypothesis 1):

$$dq_3 = dq_M \quad (15)$$

By (13), (14) and (15) a differential equation is obtained. Equation solution is:

$$\ln(T_f - T_M) = -\frac{H_3 2\pi r_e}{C_{PM}\, m_M} \cdot x + K \quad (16)$$

K is calculated by imposing the followings constraints:

- mixture temperature at L2 final section is the same as L3 input section temperature;
- mixture temperature at L3 final section is 650°C.

Transmittance calculation

Thanks to 7) hypothesis, the transmittance of the outer pipe wall along L1 segment is given by:

$$H_1 = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{1H2O}}} \quad (17)$$

Fluid motion characteristic has been identified by means of Grashof (*Gr*) and Rayleigh (*Ra*) adimensional numbers.

The convection coefficient h_f has been obtained by means of Churchill-Chu equation [2].

$$Nu = \left\{ 0.60 + \frac{0.387 \cdot Ra^{1/6}}{\left[1 + \left(\frac{0.599}{\mathrm{Pr}} \right)^{9/16} \right]^{8/27}} \right\}^2 \quad (18)$$

For h_{1H2O}, Nusselt number has been calculated by Chen-Hawkins-Solberg equation for laminar fluid motion inside annular sections [2]:

$$Nu = 1.02 \cdot \mathrm{Re}^{0.45}\, \mathrm{Pr}^{0.5} \left(\frac{d_{eq}}{L_1} \right)^{0.4} \left(\frac{d_e}{d_i} \right)^{0.8} \left(\frac{\mu_{T_m}}{\mu_{T_p}} \right)^{0.14} Gz^{0.05} \quad (19)$$

In eq. (19) an arbitrary value of L_1 has been introduced as first attempt value. Ultimate L_1 has been determined by means of an iterative process which gives:

$$L_1 = 0.01 \text{ m} \quad (20)$$

By 7) hypothesis, the transmittance of the outer pipe wall for L1 segment is given by:

$$H_1' = \frac{1}{\frac{1}{h_{1H2O}'} + \frac{s_i}{\lambda_i} + \frac{1}{h_{1CH4}'}} \quad (21)$$

In order to determine h'_{1H2O}, wall temperature of the inner pipe has been assumed as the average value of water temperature along L1 segment (T_P = 57°C). For h'_{1CH4}, *Nu* has been found by means of Sieder-Tate equation for laminar fluid motion [2]:

$$Nu = 1.86 \cdot Gz^{1/3} \left(\frac{\mu_{Tm}}{\mu_{Tp}} \right)^{0.14} + 0.87 \cdot \left(1 + 0.015 \cdot Gz^{1/3} \right) \quad (22)$$

In L2 segment the transmittance of outer pipe wall is given by:

$$H_2 = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{2H2O}}} \quad (23)$$

h_f is supposed to be the same as for L1. In the annular section, water motion is turbulent near outer wall because of boiling.

In order to determine h_{2H2O} and h'_{2H2O}, Bromley equation has been used, obtaining the convection coefficient for film evaporation [3].

$$h = 4.306 \cdot \left[\frac{\lambda_{vs}^3 (\rho_{ls} - \rho_{vs}) \rho_{vs} g}{\mu_{vs} d_{eq} (T_p - T_m)} \right]^{1/4} \quad (24)$$

where T_m is water boiling temperature and T_p, is considered the external temperature.

Inner pipe wall transmittance is given by:

$$H_2' = \frac{1}{\frac{1}{h_{2H2O}'} + \frac{s_i}{\lambda_i} + \frac{1}{h_{2CH4}'}} \quad (25)$$

h'_{2CH4} has been obtained by (22) assuming, as first attempt value, an arbitrary L_2. Ultimate L_2 has been determined by means of an iterative process which gives:

$$L_2 = 0.70 \text{ m} \quad (26)$$

Along L_3 segment two different types of flow can be observed: turbulent in the initial part corresponding to fluids mixing, laminar in the final part.

Laminar segment length has been calculated imposing:

$$\mathrm{Re} = \frac{4 \cdot m_M}{\pi \cdot d_e \cdot \mu} = 2000 \tag{27}$$

Mixture viscosity (μ_{2000}) has been determined by eq. (27). Given μ_{2000} and gases molar fraction, mixture temperature has been calculated by means of Keyes equation by an iterative process [4]:

$$\mu = \frac{a_0 \cdot \sqrt{T}}{1 + \frac{a}{T} \cdot 10^{-a_1/T}} \tag{28}$$

Given T_{2000}, the laminar segment length (L_{3l}) is given by [5]:

$$L_{3l} = \frac{C_{PMl} \cdot m_M}{H_{3l} \cdot 2\pi r_e} \cdot \ln\left(\frac{T_f - T_{2000}}{T_f - T_M^f}\right) = 1.47 \text{ m} \tag{29}$$

The "not laminar" segment length (L_{3t}) has been calculated by means of the previous equation, considering both turbulent and laminar condition:

$$L_{3t} = \frac{C_{PMt} \cdot m_M}{H_{3t} \cdot 2\pi r_e} \cdot \ln\left(\frac{T_f - T_M^*}{T_f - T_{2000}}\right) = 0.19 \text{ m} \tag{30}$$

where H_{3t} is given by:

$$H_{3t} = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{3tM}}} \tag{31}$$

h_f is attained by Churchill-Chu equation. h_{3tM} has been calculated twice:

A) on "laminar conditions" by eq. (22);

B) on "not laminar conditions" by eq. (32) [3].

$$Nu = \frac{0.021 \cdot \mathrm{Re}^{0.8}\, \mathrm{Pr}^{0.4}}{\left(T_p / T_m\right)^{0.29 + 0.0019(L_{3t}/d)}} \tag{32}$$

Heat exchanger length is given by (33) both on A) and B) conditions due to the small difference between Nu obtained respectively by A) and B).

$$L_{tot} = L_1 + L_2 + L_{3l} + L_{3t} = 2.37 \text{ m} \tag{33}$$

Case 2 analysis

In Fig. 3 the coaxial pipe scheme is sketched. The problem solution is attained by the same method as case 1; coaxial pipe heat exchanger is again composed by three segments: L1, L2 and L3 (see case 1).

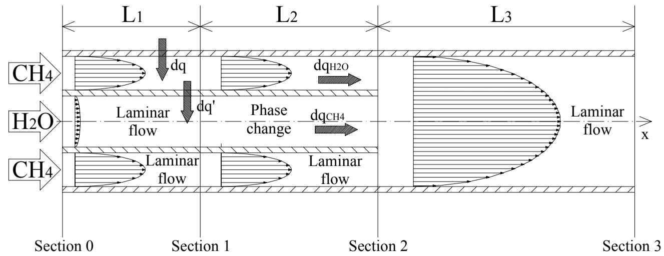

Figure 3: Coaxial pipe scheme.

The infinitesimal heat flux transmitted by outer environment to methane through outer pipe wall is given by:

$$dq = H \cdot (T_f - T_{CH4}) \cdot 2\pi\, r_e \cdot dx \tag{34}$$

while the heat transmitted from gas to water through inner pipe wall is:

$$dq' = H' \cdot (T_{CH4} - T_{H2O}) \cdot 2\pi\, r_i \cdot dx \tag{35}$$

Energy absorbed by fluids along L1 and L2 segments are given respectively by (3), (4) and (5), while equations (6) and (7) become:

$$dq = dq_{CH4} + dq' \tag{36}$$

$$dq' = dq_{H2O} \tag{37}$$

By substituting equations (3), (4), (34) and (35) into (36) and (37), the followings first order differential equations are obtained:

$$\begin{cases} \dfrac{dT_{H2O}}{dx} = \dfrac{H_1' \cdot 2\pi r_i}{C_{P1H2O} \cdot m_{H2O}} (T_{CH4} - T_{H_2O}) \\ \dfrac{dT_{CH4}}{dx} = \dfrac{H_1 \cdot 2\pi r_e}{C_{P1CH4} \cdot m_{CH4}} (T_f - T_{CH4}) - \dfrac{H_1' \cdot 2\pi r_i}{C_{P1CH4} \cdot m_{CH4}} (T_{CH4} - T_{H2O}) \end{cases} \tag{38}$$

Solving (38) with respect to $T_{H2O}(x)$ and $T_{CH4}(x)$, the following L1 length has been attained:

$$L_1 = 0.19 \text{ m} \tag{39}$$

Along L2 only methane temperature is x dependent while water temperature is constant. L3 length has been calculated by the same method as case 1.

Transmittance calculation

Thanks to hypothesis 7) the transmittance of the outer pipe wall along L1 is given by:

$$H_1 = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{1CH4}}} \quad (40)$$

where h_f is calculated by Churchill-Chu equation. Methane motion characteristic along L1 segment has been individuated by means of *Re*. h_{1CH4} has been obtained by means of Chen-Hawkins-Solberg equation.

By 7) hypothesis, the transmittance of the inner pipe wall is given by:

$$H_1^{'} = \frac{1}{\frac{1}{h_{1CH4}^{'}} + \frac{s_i}{\lambda_i} + \frac{1}{h_{1H2O}^{'}}} \quad (41)$$

For h'_{1CH4} eq. (19) has been employed; wall temperature T_p has been assumed as the average value of the water temperature along L1. h'_{1H2O} has been calculated by means of Hausen equation:

$$Nu = 3.66 + \frac{0.085 \cdot Gz}{1 + 0.047 \cdot Gz^{2/3}} \left(\frac{\mu_{T_m}}{\mu_{T_p}} \right)^{0.14} \quad (42)$$

In L2 segment, transmittance of the outer pipe wall is given by:

$$H_2 = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{2CH4}}} \quad (43)$$

where h_f and h_{2CH4} have been calculated respectively by means of Churchill-Chu and Chen-Hawkins-Solberg equations. Transmittance of the inner pipe wall is:

$$H_2^{'} = \frac{1}{\frac{1}{h_{2CH4}^{'}} + \frac{s_i}{\lambda_i} + \frac{1}{h_{2H2O}^{'}}} \quad (44)$$

To calculate h'_{2CH4} equation (19) has been employed. h'_{2H2O} has been obtained by means Bromley equation assuming film boiling inside the inner pipe. For L2 length the following value has been found:

$$L_2 = 1.90 \text{ m} \quad (45)$$

In L3 segment, transmittance of the pipe wall is given by:

$$H_3 = \frac{1}{\frac{1}{h_f} + \frac{s_e}{\lambda_e} + \frac{1}{h_{3M}}} \quad (46)$$

where h_f is given by Churchill-Chu equation. To calculate h_{3M} Hausen equation has been employed. The L3 length has been obtained by means of case 1 method:

$$L_3 = 1.32 \text{ m} \quad (47)$$

In this case fluid motion is laminar all along L3 segment and heat exchanger length results:

$$L_{tot} = L_1 + L_2 + L_3 = 3.41 \text{ m} \quad (48)$$

Gases arrangement of case 2 determines:

- heat exchanger length longer than case 1;
- mixture motion along L3 is laminar and gases mixing is inhibited.

NOMENCLATURE

C_{P1CH4} methane specific heat along L1 segment (J/kgK)
C_{P1H2O} water specific heat along L1 segment (J/kgK)
C_{PCH4} methane specific heat (J/kgK)
C_{PH2O} water specific heat (J/kgK)
C_{PM} mixture specific heat (J/kgK)
C_{PMl} mixture specific heat for laminar flow (J/kgK)
C_{PMt} water specific heat for turbulent flow (J/kgK)
d pipe diameter (m)
dq heat flux transmitted through outer pipe wall (W)
dq' heat flux transmitted through inner pipe wall (W)
dq_3 heat flux transmitted through outer pipe wall along L3 segment (W)
dq_{CH4} heat flux absorbed by methane (W)
dq_{H2O} heat flux absorbed by water (W)
dq_M heat flux absorbed by mixture (W)
dr^* water vaporization heat (J/kg)
dT_{CH4} methane temperature gradient (K)
dT_{H2O} water temperature gradient (K)
dx infinitesimal segment (m)
Gz Graetz number (adimensional)
h'_{1CH4} methane-inner pipe convection coefficient along L1 (W/m^{2}K)
h_{1H2O} water-outer pipe convection coefficient along L1 (W/m^{2}K)
h'_{1H2O} water-inner pipe convection coefficient along L1 (W/m^{2}K)
h'_{2CH4} methane-inner pipe convection coefficient along L2 (W/m^{2}K)
h_{2H2O} water-outer pipe convection coefficient along L2 (W/m^{2}K)
h'_{2H2O} water-inner pipe convection coefficient along L2 (W/m^{2}K)

h_{3tM} mixture-outer pipe convection coefficient along L3 (W/m^2K)
h_{CH4} methane convection coefficient (W/m^2K)
h_f outside fluid convection coefficient (W/m^2K)
h_{H2O} water convection coefficient (W/m^2K)
H outer pipe transmittance (W/m^2K)
H' inner pipe transmittance (W/m^2K)
H_1 outer pipe transmittance along L1 (W/m^2K)
H'_1 inner pipe transmittance along L1 (W/m^2K)
H_2 outer pipe transmittance along L2 (W/m^2K)
H'_2 inner pipe transmittance along L2 (W/m^2K)
H_3 outer pipe transmittance along L3 (W/m^2K)
H_{3l} outer pipe transmittance along L3 for laminar flow (W/m^2K)
H_{3t} outer pipe transmittance along L3 for not laminar flow (W/m^2K)
K integration constant (adimensional)
L_1 L1 length (m)
L_2 L2 length (m)
L_{3t} not laminar segment length (m)
L_{3l} laminar segment length (m)
m_{CH4} methane mass flow rate (kg/s)
m_{H2O} water mass flow rate (kg/s)
m_M mixture mass flow rate (kg/s)
Nu Nusselt number (adimensional)
Pr Prandtl number (adimensional)
r pipe radius (m)
r^* heat of vaporization (J/kg)
Re Reynolds number (adimensional)
s pipe thickness (m)
T_{2000} mixture temperature when Re is 2000 (K)
T_{CH4} methane temperature (K)
T_f environment temperature (K)
T_{H2O} water temperature (K)
T^*_{H2O} water boiling point (K)
T_m bulk temperature (K)
T_M mixture temperature (K)
T^*_M mixture temperature at Section 3 (K)
T^f_M mixture temperature at Section 2 (K)
T_P fluid temperature near pipe wall (K)

Greek Letters
λ thermal conductivity (W/mK)
μ_{373} mixture viscosity at T_m (Ns/m^2)
μ_{923} mixture viscosity at T_m (Ns/m^2)
μ_{2000} mixture viscosity at T_m (Ns/m^2)
μ_{Tm} fluid viscosity at T_m (Ns/m^2)
μ_{Tp} fluid viscosity at T_P (Ns/m^2)

Subscripts
e outer
eq equivalent
i inside/inner

CONCLUSION

In this paper a gas heat exchanger for an original MCFC has been studied. Because of the anodic compartment peculiar geometry the only heat exchanger configuration allowed is a coil shape single pipe. The pipe is made up of two sections. In the first section, the coil is constituted by two coaxial pipes: one for each fluid. Here the methane is warmed up to 100°C while the water is warmed up to 100°C and then vaporized. In the second section the coil is constituted by a single pipe. The two fluids are mixed and warmed up to reforming process temperature (650°C).

The arrangement with water in the outer pipe and methane in the inner pipe has been found to be the one that minimizes the exchanger dimensions: the pipe length is equal to 2.37 meters, while the section occupied by the concentric pipes is 0.70 meters long.

Furthermore, such a configuration shows a “not laminar” transition region where gas mixing is enhanced.

The results have been achieved by proposing and solving new heat transfer differential equations which may also be useful for many other applications.

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