Heat exchanger for latent heat recovery

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Abstract
The exhaust gas from energy system consists of non-condensable gas and steam with sensible and latent heat, respectively. When a lot of latent heat is included in the exhaust gas, its recovery is very important to improve the system efficiency. For condensation from steam-gas mixture, analogy correlation of heat and mass transfer has been used as an approximation. The analogy method is eagerly expected for the design of heat exchangers where the heat transfer correlation has been empirically established. However, at the high concentration of steam, some modification is necessary on the mass transfer correlation because it is originated and estimated from the heat transfer correlation without condensation. Condensation heat transfer on a row of horizontal stainless steel tubes was investigated experimentally and new analogy relation taking account of the mass absorption effect on the wall was proposed. Based on this basic study, a thermal hydraulic prediction method for latent heat recovery exchangers was proposed. Furthermore two kinds of compact heat exchanger with staggered banks of small bare tubes were designed with the prediction method. The more compactness was obtained with the smaller tubes at a designed heat recovery. The thermal hydraulic behavior in the compact heat exchangers was experimentally studied with air-steam mixture gas. The experimental results agreed well with the prediction proposed in this study and the more compactness with the smaller tubes was proved. Finally the prediction method was used in the commercial design of home hot water supply system using natural gas and the thermal efficiency was raised by 17% with the latent heat recovery technology.

Keywords: Latent heat, Heat exchanger, Non-condensing gas, Prediction, Modified analogy, Compactness

1. Introduction
The most part of energy losses in the energy system is due to the heat released by the exhaust flue gas to atmosphere. The released heat consists of sensible and latent one. Recently, for a biological and environmental safety, clean fuel such as natural gas is widely used in the system such as boiler or fuel cell. As the clean fuel includes a lot of hydrogen instead of carbon, the exhaust flue gas includes a lot of steam accompanying with the latent heat. So the latent heat recovery from the flue gas is very important to improve the recent system efficiency. Furthermore to reduce the toxic products of combustion and total amount of exhaust gas, it is preferable to use oxygen instead of air. As a next generation boiler, the oxy-fuel combustion boiler was planned and developed in Japan (Osakabe et al., 2001, 2002). The oxy-fuel combustion flue gas has the larger concentration of steam than that in the air-fuel combustion flue gas including nitrogen. So the latent heat recovery from the flue gas is very effective and important to compensate the additional energy to separate oxygen from air and improve the boiler efficiency.

Shown in Fig.1 is a relation between the boiler efficiency and the exhaust gas temperature in the oxygen and air combustion system (Osakabe, 2000b). The efficiency is larger than 100% at the lower exhaust temperature as the boiler efficiency is defined with a lower heating value of fuel. The efficiency in the oxygen combustion is much higher than that in the air combustion due to the lack of heat loss carried out with the nitrogen. The dew point of the oxy-fuel combustion flue gas including the larger amount of steam instead of nitrogen is higher than that in the air-fuel combustion flue gas. The steep increase of efficiency can be observed at the lower temperature region below the dew points. In this lower temperature region, the latent heat in the flue gas is recovered and the efficiency is increased significantly.
The pioneering basic studies were conducted for the latent heat recovery from wet air by Fujii et al. (1984) and Taniguchi et al. (1986). In the ensuing basic studies (Osakabe et al., 1998, 1999a, 1999b, 1999c), the condensation heat transfer on horizontal stainless steel bare or finned tubes was investigated experimentally by using actual flue gas from natural gas boiler. The experiments were conducted using single and 2 stages of tubes at different air ratios and steam mass concentrations of the flue gas in a wide range of tube wall temperature. The condensation heat transfer was well predicted with the simple analogy correlation in the high wall temperature region. In the low wall temperature region less than 30°C or the high steam mass concentration presuming the oxy-fuel combustion, the total heat transfer was higher than that predicted by the simple analogy correlation. For the high steam mass concentration, the modified Sherwood number (Osakabe et al., 1999b, 2006) taking account of the mass absorption effect on the wall was proposed.

A prediction method was proposed for the design of heat exchanger to recover the latent heat in the flue gas (Osakabe, 1999c, 2000b). In the prediction, the flue gas was treated as a mixture of CO₂, CO, SO₂, O₂, N₂ and H₂O, and its property was estimated with special combinations of each gas property proposed by the previous studies (Lindsay & Bromley, 1950)(C.R.Wilke, 1950). The mass diffusivity of steam in flue gas was estimated with the well-known mass diffusivity of steam in air (Fujii et al., 1977). The detail of the estimation method for the properties is summarized in Appendix. The one-dimensional heat and mass balance calculation was conducted along the flow direction of flue gas in the heat exchanger. For the finned tubes, the fin efficiency at the condensing region was calculated with a semi-empirical correlation (Osakabe et al., 2000a, 2002). The heat and mass transfer on tubes was evaluated with the modified analogy correlation and the thermal resistance of the condensate film on tubes. In the calculation, it was possible that the gas temperature coincided with the dew point which was the saturation temperature corresponding to the partial pressure of steam in the flue gas. When the gas temperature decreased below the dew point, the condensation of steam in the flue gas took place and the latent heat increased the gas temperature until the gas temperature coincided with the dew point.

The compactness could be obtained in the heat exchanger for the latent heat recovery when bare tubes of small diameter were used instead of conventional finned tubes (Osakabe et al., 2000a, 2002). The compact countercurrent cross-flow heat exchanger using small bare tubes of SUS304 was designed and constructed to prove its high ability (Osakabe et al., 2003). Two kinds of compact heat exchanger with staggered banks of bare tubes of 10.5 or 4mm in outer diameter was designed with the prediction method. The experimental study varying the steam concentration of mixture gas, feed water temperature and flow rate was conducted and the predictions agreed well with the experimental results (Osakabe et al., 2007, 2009). The pressure loss in gas side was slightly smaller and that in water side was significantly larger in case of the smaller tube. By adapting the single header instead of conventional multi header, the pressure loss in the water side could be significantly reduced but the reduction rate of heat recovery was only between 40 to 10% (Osakabe, 2011).

This paper mainly summarizes the 20 years research activities of present author for the shell & tube heat exchanger to recover the latent heat. Recently the new type of heat exchanger, in which flue gas flows inside tubes and water is shell-side, was proposed to develop the performance and compactness of shell & tube type heat exchanger for latent heat recovery.
heat recovery (Yamashita et al., 2013). The heat exchangers using the finned tubes were also studied by Kang et al. (2000), Joardar et al. (2008), Shi et al. (2011) and Hwang et al. (2010) in spite of the low fin efficiency due to the high heat transfer of condensation. The appropriate fin height is very important for the compact design. The experimental studies were conducted for the plate-fin type heat exchanger to recover the latent heat (Kawaguchi et al., 2006) and to design heat pump system (Osada et al., 1999). The compactness can be obtained also in these heat exchangers but the fouling degradation due to the dirty flue gas is difficult to avoid.

The condensation in the exhaust flue gas strongly depends on the steam mass transfer affected with the wall temperature and the steam mass concentration. However in some studies, only the heat transfer correlations were experimentally obtained without the concept of steam mass transfer described in this paper. To use these correlations of heat transfer, the applicable range for the wall temperature and the steam mass concentration should be carefully considered.

**Fig. 2** Heat resistance of condensate film

### 2. Constitutive equations for prediction

#### 2.1 Heat resistance of condensate

Though a part of condensate falls down between the tubes and on the duct wall, it is assumed that all the condensate generated at the upper stage flows on the tubes as a laminar film. The momentum balance dominated by viscous and gravity force gives the velocity distribution at $\theta^\circ$ from the tube top in Fig.2:

$$\begin{align*}
    u &= \frac{(\rho_L - \rho_G)g \sin \theta}{\mu_L} \left( y\delta - \frac{y^2}{2} \right) \\
    &= \frac{(\rho_L - \rho_G)g \sin \theta}{\mu_L} \left( y\delta - \frac{y^2}{2} \right) \\
    &= \frac{(\rho_L - \rho_G)g \sin \theta}{\mu_L} \left( y\delta - \frac{y^2}{2} \right)
\end{align*}$$

(1)

where $\rho$ is the density, $g$ the acceleration due to gravity, $\mu$ the viscosity, and $\delta$ the film thickness. The subscript $L$ and $G$ indicate the liquid and gas phases, respectively. Integrating the above velocity profile and using the condensate mass flow rate per unit of tube length, $m$, yields

$$\delta = \left[ \frac{1.5 \mu_L m}{\rho_L (\rho_L - \rho_G) g \sin \theta} \right]^{1/3}$$

(2)

The overall heat transfer coefficient of film $K$ is

$$K = \frac{\lambda_L}{\delta} = \left[ \frac{\lambda_L^3 (\rho_L - \rho_G) g \sin \theta}{1.5 \mu_L m} \right]^{1/3}$$

(3)

where $\lambda_L$ is the heat conductivity of film. Equation (3) gives the heat flux through the film when the temperature difference between the film is multiplied. The average overall heat transfer coefficient from $\theta = 0^\circ$ to $\theta = \pi$ is
The average heat resistance of film is defined as the inverse of the above average overall heat transfer coefficient. The average film thickness is

\[
\bar{\delta} = \frac{\lambda_f}{K}
\]  

In the calculation, the mass flow rate, \( m \), at a certain stage includes the condensate generated at the stage for the conservative estimation.

### 2.2 Analogy correlations of heat and mass transfer

The condensation with a certain amount of non-condensing gas is frequently observed in various industrial applications. The condensation heat transfer at a small amount of non-condensing gas has been studied for the air leakage problem in the condenser of steam turbine system. For a large amount of non-condensing gas, experimental and theoretical studies have been conducted for the latent heat recovery from the exhausted flue gas and the cooling of nuclear containment vessel. As the previous studies of condensation heat transfer focused on a limited range of non-condensing gas concentration, the proper estimation method covering the wide range of non-condensing gas concentration is needed for the various applications.

When the flow induced with the condensation is negligibly small in the two-dimensional flow field, the heat transportation equation is

\[
\frac{\partial \tilde{T}}{\partial \tilde{t}} + \tilde{u} \frac{\partial \tilde{T}}{\partial \tilde{x}} + \tilde{v} \frac{\partial \tilde{T}}{\partial \tilde{y}} = \frac{1}{\text{Re}_f \text{Pr}_f} \left( \frac{\partial^2 \tilde{T}}{\partial \tilde{x}^2} + \frac{\partial^2 \tilde{T}}{\partial \tilde{y}^2} \right)
\]  

where \( \text{Re} \) is Reynolds number \([=ud/\nu_m]\), \( \text{Pr} \) is Prandtl number \([=\nu_m/\kappa]\), \( u \) the velocity of \( x \) direction, \( v \) the velocity of \( y \) direction, \( d \) the representative length such as the tube diameter, \( \nu_m \) the kinematic viscosity. The non-dimensional temperature, time, velocities and coordinates are defined as,

\[
\tilde{T} = \frac{T - T_i}{T_f - T_i}, \quad \tilde{t} = \frac{t \mu_m}{d}, \quad \tilde{u} = \frac{u}{u_m}, \quad \tilde{v} = \frac{v}{u_m}, \quad \tilde{x} = \frac{x}{d} \quad \text{and} \quad \tilde{y} = \frac{y}{d}
\]

where the subscript \( f \) and \( i \) indicate the mixture gas and interface, respectively, and \( u_m \) is the representative velocity such as an average velocity. The mass transportation equation can also be described as,

\[
\frac{\partial \tilde{w}}{\partial \tilde{t}} + \tilde{u} \frac{\partial \tilde{w}}{\partial \tilde{x}} + \tilde{v} \frac{\partial \tilde{w}}{\partial \tilde{y}} = \frac{1}{\text{Re}_f \text{Sc}_f} \left( \frac{\partial^2 \tilde{w}}{\partial \tilde{x}^2} + \frac{\partial^2 \tilde{w}}{\partial \tilde{y}^2} \right)
\]

where \( \text{Sc} \) is Schmidt number \([=\nu_m/\alpha]\), \( \alpha \) the mass diffusivity, \( w \) the mass concentration per fluid of an unit mass. The non-dimensional mass concentration is defined as,

\[
\tilde{w} = \frac{w - w_i}{w_f - w_i}
\]

Equations (6) and (7) indicate that the distribution of mass concentration can be estimated with that of temperature by replacing \( \text{Pr} \) with \( \text{Sc} \). So when the heat transfer can be described by,

\[
\text{Nu}_f = \left[ \frac{\partial \tilde{T}}{\partial \tilde{y}} \right] = f(\text{Re}_f, \text{Pr}_f)
\]

where \( \text{Nu} \) is Nusselt number \([=h_d/\lambda]\), \( h \) the heat transfer coefficient. The mass transfer can be described by replacing...
Pr with Sc as,

\[
\text{Sh}_f = \left[ \frac{\partial w}{\partial y} \right] = f(\text{Re}_f, \text{Sc}_f)
\]  

where Sh is Sherwood number \(= h_d/\alpha\), \(h_d\) the mass transfer coefficient.

It should be noted that Nu and Sh are the interfacial gradients of non-dimensional temperature and concentration distributions, respectively, when the flow induced with the condensation is negligibly small. This useful and simple relation is called as the simple analogy relation and the condensation heat transfer at large amount of non-condensing gas can be well described with the relation. The analogy method is preferable for the design of heat exchangers where the heat transfer correlations have been empirically established.

For small amount of non-condensing gas, the vigorous condensation can be expected and the effect of flow towards the condensing surface (mass absorption effect) has to be considered. The non-condensing gas flows towards the surface with the condensing gas at the normal velocity \(v_i\) and is removed with the diffusion as shown in Fig.3. The continuous equation of the non-condensing gas gives

\[ v_i = -\frac{\alpha_f}{1-w_i} \left[ \frac{\partial w}{\partial y} \right] \]

\[ \text{Condensing} \quad \text{Non-condensing} \]

\[ \rho_f (1-w_i) v_i \]

\[ \rho_f \alpha_f \left[ \frac{\partial w}{\partial y} \right] \]

Fig. 3 Behavior of non-condensing gas

As a first approximation, the mass absorption effect is considered only in the mass transfer equation, not in the heat transfer equation. The integral equations for the mass and energy conservation are

\[ \frac{d}{dx} \int \rho_f u (w_f - w) dy - \rho_f v_i (w_f - w_i) = \rho_f \alpha_f \left[ \frac{\partial w}{\partial y} \right] \]

\[ \frac{d}{dx} \int \rho_f u (T_f - T) dy = \rho_f \kappa_f \left[ \frac{\partial T}{\partial y} \right] \]

where \(\kappa\) is thermal diffusivity. By using the non-dimensional temperature and mass concentration, Eqs (11) and (12) become

\[ \frac{d}{dx} \int \rho_f u (1-w) dy = \frac{1-w_f}{1-w_i} \rho_f \alpha_f \left[ \frac{\partial w}{\partial y} \right] \]

\[ \frac{d}{dx} \int \rho_f u (1-T) dy = \rho_f \kappa_f \left[ \frac{\partial T}{\partial y} \right] \]
\[ \kappa_f = \frac{1 - w_f}{1 - w_i} \alpha_f \]  

is presumed, the distribution of non-dimensional mass concentration coincides with that of non-dimensional temperature. Equation (15) can be modified as

\[ \text{Pr}_f = \frac{1 - w_i}{1 - w_f} \text{Sc}_f \]  

When Nu is described with function \( f \)

\[ \left[ \frac{\partial T}{\partial y} \right] = \frac{1}{d} \text{Nu}_f = \frac{1}{d} f (\text{Re}_f, \text{Pr}_f) \]  

Equations (16) and (17) give the gradient of non-dimensional mass concentration as

\[ \left[ \frac{\partial w}{\partial y} \right] = \frac{1}{d} f (\text{Re}_f, \frac{1 - w_i}{1 - w_f} \text{Sc}_f) \]  

The condensation rate \( m_c \) can be described with considering the normal velocity to the surface and the diffusion as

\[ m_c = -\rho_f v_i w_i + \rho_f \alpha_f \left[ \frac{\partial w}{\partial y} \right] \]  

The normal velocity \( v_i \) given by Eq(10) and Eq(19) give

\[ m_c = -\rho_f v_i w_i - \rho_f v_i (1 - w_f) = -\rho_f v_i \]  

The condensation rate can be described with the simple multiplication of mixture density and the normal velocity. When the normal velocity \( v_i \) given by Eq(10) is again used in Eq(20),

\[ m_c = -\rho_f v_i \frac{\rho_f \alpha_f}{1 - w_i} \left[ \frac{\partial w}{\partial y} \right] = \frac{w_f - w_i}{1 - w_i} \rho_f \alpha_f \left[ \frac{\partial w}{\partial y} \right] \]  

Equations (18) and (21) give Sh for small amount of non-condensing gas as,

\[ \text{Sh}_f = \frac{m_c D}{\rho_f \alpha_f (w_f - w_i)} = \frac{1}{1 - w_i} f (\text{Re}_f, \frac{1}{\omega} \text{Sc}_f) \]  

where \( \omega = \frac{1 - w_f}{1 - w_i} \)

So the mass transfer equation can easily be derived if the heat transfer function of Nu is known. These correlations gave good predictions when the non-condensing gas concentration was more than 75% in single and multiple stages of heat transfer tubes using actual flue gas. But in the analogy relation, the mass transfer correlation was originated and estimated with the heat transfer correlation without the condensation; i.e. no mass absorption effect. So the estimation error can be expected as the heat transfer equation is affected with the mass absorption effect when the significant condensation occurs at the lower concentration of non-condensing gas.

2.3 Heat and mass transfer in gas side

The total heat flux \( q_w \) consists of the convection heat flux \( q_V \) and the condensation heat flux \( q_C \) as
The convection heat flux is expressed as
\[ q_V = h_V(T_f - T_i) \] (24)

where \( h_V \) is the heat transfer coefficient. The condensation heat flux \( q_C \) can be expressed as,
\[ q_C = h_C L_w \rho_f (w_f - w_i) \] (25)

where \( w_i \) is the mass concentration of saturated steam at the interfacial temperature \( T_i \), \( h_C \) the mass transfer coefficient, \( L_w \) the latent heat. Based on the previous studies[12], the Nusselt number \( \text{Nu}_f \) for the average convective heat transfer coefficient in the range of \( 10^3 < \text{Re}_f \leq 2 \times 10^5 \) is
\[ \text{Nu}_f = c \text{Re}_f^{a} \text{Pr}_f^{b} (\text{Pr}_f / \text{Pr}_w)^{0.25} \] (26)

Zukauskas(1972) proposed \( a=0.6, b=0.36 \) and
\[ \text{For } s_1/s_2 < 2 \quad c = 0.35 (s_1 / s_2)^{0.2} \] (27)
\[ \text{For } s_1/s_2 \geq 2 \quad c = 0.40 \] (28)

for a staggered bank. In the equation, \( s_1 \) and \( s_2 \) are span-wise and flow-directional pitch of the tubes as shown in Fig.10, respectively. For the condensation of steam on heat transfer tubes, the modified analogy relation of Eq.(22) gives
\[ \text{Sh}_f = M_f c \text{Re}_f^{a} \text{Sc}_f^{b} (\text{Sc}_f / \text{Sc}_w)^{0.25} \] (29)

where \( M_f = \frac{1}{1-w_f} \left( \frac{1}{\omega_f} \right)^b \)

The Sh number increases sharply at the steam mass concentration of 1 in Eq.(29). This indicates the mass transfer at the pure steam condition is enough high to neglect the interfacial resistance of mass transfer. In the calculation for pure steam without non-condensing gas, the modification factor \( M_f \) of 100 is used to avoid the calculation error divided by zero.

The one-dimensional heat and mass balance calculation along the flow direction of flue gas was conducted. The steam mass concentration and the flue gas temperature at N+1th stage can be calculated from those at Nth stage as shown in Fig.4.
The heat and mass balance equations are;

\[
 w_f(N+1) = \frac{m_f w_f(N) - q_c A_w / L_w}{m_f - q_c A_w / L_w} \tag{30}
\]

\[
 T_f(N+1) = T_f(N) - \frac{q_v A_w}{C_{pf} m_f} \tag{31}
\]

where \( A_w \) is the heat transfer area per a stage, \( C_{pf} \) the specific heat of flue gas.

It is possible that the gas temperature merges with the dew point which is the saturation temperature corresponding to the partial pressure of steam in the flue gas. When the gas temperature decreases below the dew point, the condensation of steam in the flue gas takes place and the latent heat increases the gas temperature until the gas temperature coincides with the dew point. In this case, the energy balance gives the relation between the increase of the gas temperature, \( \Delta T_f \), and the decrease of steam concentration, \( \Delta w_f \), as;

\[
 \Delta T_f = \frac{L_w}{C_{pf}} \Delta w_f \tag{32}
\]

### 2.4 Heat conduction in tube

The heat conductivity for the inconel or austenite stainless steel is given with the following approximate correlation (Osakabe, 1989).

\[
 \lambda_t = 13.2 + 0.013 T_t \quad \text{W/(m K)} \tag{33}
\]

where \( T_t \) is the average temperature of tube as,

\[
 T_t = \frac{T_w - T_{wi}}{2} \tag{34}
\]

where \( T_w \) and \( T_{wi} \) are the outer and inner wall temperatures, respectively. The heat flux at the outer wall is,

\[
 q_w = \frac{2 \lambda_t (T_w - T_{wi})}{d \ln (d / d_i)} \tag{35}
\]

where \( d \) is the outer diameter of tube, \( d_i \) the inner diameter of tube.

### 2.5 Heat transfer in water side

Heat transfer correlation by Dittus-Boelter taking account of the pipe inlet region is used. The coefficient by McAdams(1954) was used for the modification.

\[
 Nu = 0.023 \left( \frac{d}{L} \right)^{0.8} \left( \frac{d_i}{L} \right)^{0.4} \left[ 1 + \left( \frac{d_i}{L} \right)^{0.7} \right] \tag{36}
\]

where \( L \) is the heating length of tube.

### 2.6 Pressure loss calculation

The pressure loss per a stage of tube in gas side is,

\[
 \Delta P = 2 \rho_f u^2 \tag{37}
\]

For the staggered bank of bare tube, Jacob(1938) proposed the following coefficient \( f \).
This correlation can be used also in the range of $10^3 < Re_f \leq 2 \times 10^5$. The pressure loss per a tube in water side is expressed as followings assuming the inlet and outlet pressure loss coefficient of 1.5,

$$\Delta p = \left( 1.5 + 4f \frac{L}{d_i} \right) \frac{\rho u^2}{2}$$

where the friction coefficient $f$ is,

$$f = \frac{16}{Re} \quad \text{for laminar flow,}$$

$$f = 0.079 Re^{-0.25} \quad \text{for turbulent flow.}$$

3. Single row experiment

Shown in Fig.5 is a schematic of experimental apparatus. The steam of approximately 120°C mixed with air as non-condensing gas was supplied to the test section of atmospheric pressure. Steam and air was well mixed with the special nozzles where air and steam jets had different rotational directions. The steam was supplied from a couple of steam boilers and the mass flow rate was measured with a vortex-shedding flow meter or V-cone mass flow meter. The error was within ±2% of the measured mass flow rate. The airflow rate was measured with several rotor flow meters. The mixture gas temperatures just above and below the test tubes were measured with sheathed T-type thermocouples of 0.5 mm in diameter.

Water cooled test tubes of SUS316L were installed in a transparent polycarbonate duct with a cross-section of 160mm×101mm as shown in Fig.5. The water flow rate was measured with a rotor flow meter. The connecting pipes outside the test section were thermally insulated with a glass wool. Three tubes of 21.7mm in diameter were arranged horizontally at a pitch of 33.7mm. The pitch to diameter ratio $s/d$ was 1.55 that was typical for the conventional heat exchanger. Sheathed T-type thermocouples of 0.5 mm in diameter were imbedded at circumferential locations of 0, 45, 90, 135 and 180° from the top of 3 tubes to obtain the average wall temperature. The inlet and outlet water temperatures of the test tubes were measured also with the sheathed K-type thermocouples of 0.5 mm in diameter. The outlet temperature was measured at a mixing chamber to obtain a well-mixed bulk temperature.

The temperature difference of the cooling water through the tubes was kept approximately at 1.7~15.7K. The heat flux of the test tubes was calculated with the flow rate and the temperature difference of cooling water through the tubes. The measurement error of the heat flux was estimated to be ±12% as the measurement error of the temperature.
by thermocouples was within ±0.1 K. The thermocouple signals were transferred to a personal computer by a data logger and analyzed.

Table 1 Experimental condition

<table>
<thead>
<tr>
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<th>1350 ~ 11300</th>
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<tr>
<td>$Re_f$</td>
<td></td>
</tr>
<tr>
<td>Air mass ratio $I/W_f$</td>
<td>0 ~ 0.78</td>
</tr>
<tr>
<td>Mixture gas temp. $T_f$ (°C)</td>
<td>73.0 ~ 129.1</td>
</tr>
<tr>
<td>Average wall temp. $T_w$ (°C)</td>
<td>28.8 ~ 91.0</td>
</tr>
<tr>
<td>Inlet cooling water temp. (°C)</td>
<td>8.0 ~ 74.0</td>
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The experiments were conducted at atmospheric pressure. The mixture Re number, the non-condensing gas concentration and average surface temperature of tubes were varied for the parametric study. The major test conditions are shown in Table 1.

The average interfacial temperature $T_i$ can be described with the average wall temperature $T_w$ and heat flux $q_w$ as,

$$T_i = \frac{q_w}{K} + T_w$$

(40)

The heat flux $q_w$ can be calculated both with Eq.(40) and the heat & mass transfer equation mentioned above in the mixture gas side which strongly depends on the interfacial temperature $T_i$. The iteration method was used to obtain the heat flux and interfacial temperature. In the present study of single stage, the average flow rate of condensate on unit of tube length is used as the condensate mass flow rate, $m$. These equations which give the average conductivity based on the condensate mass flow rate are very important to calculate the inundation effects on the tubes in the multi-stages tube bank.

Based on the previous studies, the Nusselt number $Nu_f$ for the average convective heat transfer coefficient is

$$Nu_f = c Re_f^a Pr_f^b (Pr_f / Pr_w)^{0.25}$$

(41)

where subscript $f$ and $w$ indicate the physical values defined at mixture gas and wall, respectively. Zukauskas(1972) proposed $a = 0.6$, $b = 0.37$, $c = 0.26$ for the first row tubes of an in-line bank with $s/d = 1.6$, which is approximately the same as that in the present study, in the range of $10^3 < Re_f \leq 2 \times 10^5$.

The analogy relation Eq.(22) gives the Sherwood number describing the average mass transfer coefficient as,

$$Sh_f = M_f c Re_f^a Sc_f^b (Sc_f / Sc_w)^{0.25}$$

(42)

where $M_f = \frac{1}{1-w_f} \left( \frac{1}{\omega} \right)^b$

The mixture gas was treated as the mixture of N$_2$, O$_2$ and H$_2$O and its property was estimated with special combinations of each gas property proposed by the previous studies mentioned in Appendix.

The heat resistance of condensate dominates the heat transfer in the pure steam condition as the convective and diffusive resistance in the gas side is negligible. So the heat resistance of condensate was studied experimentally using pure steam before the mixture gas experiments. When the laminar film of condensate without the interfacial shear is assumed on the heat transfer tube, the average heat transfer coefficient can be described with the following Nusselt correlation.

$$Nu = \frac{h d}{\lambda_L} = 0.728 \left( \frac{Ra}{Ja} \right)^{1/4}$$

(43)

where $Ra = \frac{g \left( \rho_L - \rho_f \right) Pr_L d^3}{\rho_L \nu_L^2}$, $Ja = \frac{C_{ml} (T_{sat} - T_w)}{L_w}$. 

where the subscript sat and w indicate the saturated condition and tube wall, respectively. Though Eq.(4) was obtained with the constant flow rate of condensate, the actual flow rate of condensate increases as flowing down from the top to the bottom of tube. When 0.424 times of the total generated condensate is used as \( m \) in Eq. (4), Eq.(43) coincides with Eq.(4). The average flow rate of condensate for the evaluation of heat transfer is slightly smaller than the half of total generated condensate on the tube.

On the other hand, Fujii(1981) proposed the following empirical correlation for the condensation heat transfer on a single tube placed in a wide space taking account of the effect of cross flow velocity \( u_\infty \).

\[
\text{Nu} = 0.96X^{0.2}\sqrt{\text{Re}_L} 
\]

(44)

where \( X = \frac{\text{Pr}_L}{\text{FrJa}} \), \( \text{Fr} = \frac{u_\infty^2}{gd} \), \( \text{Re}_L = \frac{u_\infty d}{v_L} \),

in the applicable range of \( 0.03 < X < 600 \). In the above correlation, \( u_\infty \) is the main flow velocity approaching to the single tube.

*Fig. 6 Condensation without non-condensing gas*

Shown in Fig.6 is the relation of steam Re and non-dimensional heat flux divided by Nusselt and Fujii’s predictions. The key \( \Delta \) is the non-dimensional heat flux divided by the heat flux obtained with the average conductivity of laminar film, Eq.(4), where 0.424 times of the total generated condensate is used as the average condensate mass flow rate \( m \). The total mass of generated condensate was estimated with the measured heat flux. In the present experimental range, the experimental data agree well with the prediction assuming the laminar film of zero interfacial shear. As the Fujii’s correlation was obtained with horizontal single tube experiments crossed by the horizontal flow of low-pressure steam, the lower heat transfer in the present experiment is considered to be due to the different experimental geometry.

The observation showed that the heat transfer surface was covered always with the thin film of condensate at the lower mass concentration of air but the several dry spots appeared at the higher concentration of air. In the present prediction, the laminar film of condensate was assumed as a first approximation. The heat flux was calculated with Eq.(40) and the heat & mass transfer correlations in the gas flow side. As mentioned above, 0.424 times of the total generated condensate was used as the average condensate mass flow rate \( m \). The total generated condensate was calculated with the mass transfer determined by the predicted Sh number in the mixture gas side. As the film of condensate was not continuous and several dry spots could be observed at the higher mass concentration of air, the assumption of continuous laminar film in the present prediction is not appropriate in the strict sense. However, the heat transfer in this region was dominated with the convection and diffusion of the mixture gas side and the film thickness of condensate was negligibly small. So it was considered that the effect of non-continuous film appeared as the dry...
spots could be neglected.

Shown in Fig.7 is the relation of non-dimensional heat flux and air mass concentration ratio between main flow and interface, $\omega = (1-W_f)/(1-W_i)$. The parameter $\omega$ is the important in previous correlations by Rose(1980) and Fujii(1981), and approximately proportional to $1-W_f$ in the present experiment. The experimental heat flux approximately tends to agree with the prediction at the lower $\omega$ less than 0.2 because the heat transfer in this region is dominated with the heat conductivity of film, and increases with an increase of $\omega$. The non-dimensional heat flux takes a maximum and gradually decreases to 1 approximately at $\omega=0.8$ indicating the proper prediction.

When $\omega$ is larger than 0.8, the modification of correlation is not necessary as the flow induced with the condensation becomes negligibly small. The previous experimental data at the non-condensing gas concentration larger than 75% ($\omega > 0.75$) also agreed well without the modification. The non-dimensional heat flux can be described with the following correlation except the region of low and high non-condensing gas concentration.

$$q_w/(q_w)_{Pred.} = 2 - 1.2 \omega \quad (45)$$

The mass transfer correlation was estimated from the convective heat transfer correlation without condensation in the analogy relation described with Eqs.(41) and (42). So the mass transfer correlation underestimates the experimental result when the convective heat transfer is enhanced due to the significant condensation at the low mass concentration of non-condensing gas. However, the convective heat flux is negligibly small compared to the condensation heat flux when the enhancement due to the condensation appears. So the modification of convective heat transfer correlation due to the mass absorption effect is not important compared to that of the mass transfer correlation.

On the other hand, when the enhancement due to the mass absorption effect can be neglected at the high concentration of non-condensing gas, the modification of heat transfer correlation is considered to be also unnecessary. As a first approximation, the heat transfer correlation without the mass absorption effect was used and only the mass transfer correlation was modified with Eq.(45) as,

$$Sh_f = M_f h e^{a_f} S_c^b (Sc_f)/(Sc_w)^{0.25} \quad (46)$$

where

$$M_f = \frac{Max(1, 2 - 1.2 \omega)}{1 - w_f} \left( \frac{1}{\omega} \right)^b$$

![Fig. 7 Relation of heat flux divided by conventional predictions and $\omega = (1-W_f)/(1-W_i)$](image-url)
Considering the previous study of the non-condensing gas concentration more than 75%, the larger value of $1$ or $2-1.2\omega$ is given by the Max function in Eq.(46). The estimated uncertainty for the $Sh$ number is considered to be within $\pm20\%$.

Shown in Fig.8 is the comparison of experimental and predicted heat flux with the previous and new analogy relation. The underestimation with the previous analogy relation is successfully improved in the new analogy relation. The slight underestimation at the higher heat flux region is considered to be due to the conservative assumption of laminar film with zero interfacial shears.

To verify the new analogy relation in the different geometry is important, the relation was applied to a single tube placed in a wide space. Rose(1980) proposed the following correlation for a single tube placed in a wide space and crossed by steam with non-condensing gas.

$$Sh_f = \frac{1 + 2.28Sc_f^{1/3}(\frac{1}{\omega} - 1)}{2(1 - \omega)} \sqrt{\frac{Re_{fc}}{(1 - w_f)}} \left(\frac{1}{1 - w_f}\right)$$

(47)

where $\omega = \frac{1 - w_f}{1 - w_i}$.

For the convective heat transfer with the condensation is expressed with,

$$Nu_f = \frac{0.57\sqrt{Re_{fc}}Pr_f^{1/3}}{1 + \beta Pr_f} \sqrt{Re_{fc}}$$

(48)

where $\beta = \frac{Sh_f(1 - w_f)(1 - \omega)}{\sqrt{Re_{fc}Sc_f}}$. The applicable range of the equation is $10 < Re_{fc} < 10^4$.

Fujii(1981) proposed the following correlation instead of Eq.(47) to fit to their experimental results.

$$Sh_f = 0.73(1 + 0.0028\sqrt{Re_{fc}})\frac{Sc_f^{1/3}}{\sqrt{\omega(1 + \omega)}} \frac{\sqrt{Re_{fc}}}{1 - w_f}$$

(49)
In the correlations by Rose and Fuji, the physical values were defined at the mean temperature of mixture gas and interface; i.e. film temperature.

In Rose and Fujii’s correlations, the base correlation of heat transfer to estimate the mass transfer correlation is,

\[ \text{Nu}_f = 0.57 \text{Re}_f^{1/2} \text{Pr}_f^{1/3} \]  \hspace{1cm} (50)

The new analogy relation in the present study simply and successfully estimates the mass transfer correlation as,

\[ \text{Sh}_f = \frac{\text{Max}(1, 2 - 1.2 \omega)}{1 - w_f} \left( \frac{1}{\omega} \right)^{1/3} 0.57 \text{Re}_f^{1/2} \text{Sc}_f^{1/3} \]  \hspace{1cm} (51)

![Fig. 9 Comparison of present new analogy, Rose and Fujii correlations for single tube](image)

Shown in Fig.9 is the comparison of this correlation with the Rose and Fujii’s correlations. In the Rose and Fujii’s correlations, \( \text{Sc}_f \) of 0.8 and \( \text{Re}_f \) of 5000, respectively, were used as the typical values. The mass transfer correlation given by the new analogy relation agrees well with the Rose and Fujii’s correlations. The new analogy relation is useful not only for the tube bank but also for the single tube.

When the single tube correlation has been applied to the heat exchanger geometry, the special velocity taking account of the blockage effect in the restricted duct has been empirically used. These empirical methods have been applied though the flow in the heat exchanger is apparently different from that around the single tube in a wide space. The application of these methods to the different geometries of heat exchangers such as using fins or spacers to enhance the heat transfer of tube is very difficult. Also when the flow slantingly crosses the tubes or the much smaller tubes of diameter than the conventional tubes are used, the application of these methods should be careful. Even when the heat transfer characteristics are significantly different from that of the conventional single tube in wide space, the present new analogy relation can give the mass transfer correlation if the heat transfer correlation is empirically given. So the wide applications of the new analogy relation are expected on the future design of condensing heat exchangers of complex geometries.

4. Compact heat exchanger

In the following design and prediction of compact heat exchanger, the new analogy relation mentioned above was
used. Shown in Fig. 10 is a schematic of compact heat exchanger. Heat transfer tubes were installed in a rectangular duct of 205x205mm. The tubes at each stage were connected with a header to maintain the same flow rate of feed water. The feed water was supplied at the downstream of gas flow and flows counter-currently to the upstream. In the present study, two kinds of heat transfer tubes with the different diameter were used. The height \( L \) of the heat exchanger necessary to recover a desired heat strongly depends on the diameter of heat transfer tubes. The arrangement of heat transfer tubes is also shown in Fig. 10. The staggered tube bank with the same flow-directional and span-wise pitch was adopted. Two kinds of bare tubes of 10.5 or 4mm in outer diameter were installed in the rectangular duct. The heat exchanger with 10.5 mm tubes was called as “Large” and that with 4mm was called as “Small”.

Shown in Fig. 11 are photograph of the two heat exchangers designed with the same heat recovery rate. The height of “Large” heat exchanger with the larger tubes was 820mm, on the other hands, that of “Small” with the smaller tubes was only 160mm.

Generally the heat transfer is described as,

\[
\text{Nu} \approx \text{Re}^m
\]

(52)

where \( m \) is between 0 and 0.8. So the heat transfer coefficient \( h \) can be expressed as,

\[
h \approx \frac{1}{d^{1-m}}
\]

(53)

The smaller diameter \( d \) of tube results as the higher heat transfer coefficient and the analogy relation gives the higher mass transfer coefficient. The more compactness of heat exchanger can be obtained with the smaller heat transfer tubes.

---

**Fig. 10 Schematic of tube bank and array**

**Fig. 11 Photograph of two HXs**
Table 2 shows the major dimensions. The total number of heat transfer tubes was 380 in the “Large” and 500 in the “Small”. The total weight of heat transfer tubes was 21.6kg in the “Large” and 7.65kg in the “Small”. The tube weight of “Small” was approximately 1/3 of “Large”. The heat transfer area at the gas side of “Small” was approximately the half of “Large”. The compactness was achieved with the smaller tubes.

Table 3 Major test conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Steam mass flow rate kg/h</td>
<td>21.6 ~ 86.9</td>
</tr>
<tr>
<td>Steam mass concentration $w_f$</td>
<td>0.2 ~ 1</td>
</tr>
<tr>
<td>Mixture gas temp. $T_f$ (°C)</td>
<td>77.0 ~ 113.2</td>
</tr>
<tr>
<td>Cooling water flow rate kg/h</td>
<td>360 ~ 614</td>
</tr>
<tr>
<td>Inlet temp. of cooling water (°C)</td>
<td>8.7 ~ 26.8</td>
</tr>
</tbody>
</table>

Fig. 12 Comparison of recovered heat
The temperature distributions of water and flue gas in the heat exchanger were measured with sheathed thermocouples. The thermocouple signals were transferred to a personal computer with a GPIB line and analyzed. The measurement error of the temperature in this study was within ±0.1 K. The parametric study varying the mixture gas flow rate, steam mass concentration, feed water flow rate and temperature was conducted. The major test conditions are shown in Table 3. The steam mass flow rate was varied between 21.6 to 86.9 kg/h and the air mass flow rate was controlled to obtain a desired steam mass concentration between 0.2 and 1. As the result of mixing of steam and air, the mixture gas temperature was between 77 and 113 °C.

Shown in Fig. 13 is the comparison of the experimental result and prediction at the steam mass concentration $w_f=0.35$ in “Large” heat exchanger. Feed water and steam mass flow rates are 600 and 23.5 kg/h, respectively. The key ○ and △ are the measured temperatures of gas and water, respectively. The lines in the figure are the predictions. The solid lines are the temperatures of gas and water in the tube bank. The a-dot-dashed line and the two-dots-dashed line are the interfacial temperature of condensate and the inner wall temperature of tubes, respectively. The dashed line which is the saturation temperature (dew point) corresponding to the partial pressure of steam in the mixture gas agrees well with the prediction of gas temperature. When the outer wall temperature is smaller than the dew point, the condensation on the wall takes place throughout the heat exchanger. The dew point and gas temperature decrease with increasing stages indicating the condensation of steam in the mixture gas.

In the present experiments, the entrained and dispersed condensate in the mixture gas tends to wet the thermocouples among the tube bank in spite of the small umbrella installed above the thermocouple. The wet thermocouples indicate the lower value than the actual gas temperature. Though the prediction for the gas temperature is slightly higher than the experimental result, the prediction for the water temperature agrees well with the experimental result indicating the proper prediction.

![Fig. 13 Temperature distribution at $w_f=0.35$ in “Large”](image-url)
Shown in Fig. 14 is the comparison of the experimental result and prediction at the steam mass concentration $w_f=0.84$ in “Large” heat exchanger. The temperature increase of cooling water is strongly depressed as the saturation temperature and the cooling water temperature is nearly equal at the upper heat exchanger. When the interfacial temperature becomes below the saturation temperature at 12th stage, the condensation on the tubes initiates and the temperature rise of cooling water significantly increases.
The saturation temperature merges with the gas temperature approximately at 25th stage and the condensation of steam occurs not only on the tubes but also in the mixture gas below this stage. The wet thermocouples also indicate the lower value than the actual gas temperature. Though the prediction for the gas temperature is slightly higher than the experimental result, the prediction for the water temperature agrees well with the experimental result indicating the proper prediction.

In the case of pure steam, \( w_f = 1 \), in “Large” heat exchanger as shown in Fig.15, the inlet gas temperature was equal to the saturation temperature of 100°C as the outlet of heat exchanger is open to the atmosphere. Heat resistance of condensate film governs the heat transfer process in this case. When the gas temperature sharply dropped approximately at 14th stage, all the steam has condensed above this height. Below the height, air can invade from the outlet of heat exchanger and the heat transfer was strongly depressed. So the temperature field is kept nearly at the inlet temperature of cooling water. Even in this case, the prediction using the modification factor \( M_f \) of 100 agrees well with the experiment.

Shown in Fig.16 is the comparison of the experimental result and prediction at the steam mass concentration \( w_f = 0.37 \) in “Small” heat exchanger using the smaller tubes when the steam flow rate is 24.1 kg/h. The tube outer diameter of “Small” is 4 mm and nearly equal to the droplets size falling from the upper condensing tubes. So the comparison of the experimental result and prediction assuming the uniform thin film of condensate is interesting and important for the further improvement of prediction method. Though the prediction for the gas temperature is slightly higher than the experimental result due to the wet thermocouples, the prediction for the water temperature agrees well with the experimental result indicating the proper prediction for the heat exchanger of smaller tubes.

Shown in Fig.17 is the comparison of the experimental result and prediction at the steam mass concentration \( w_f = 0.37 \) in “Small” heat exchanger when the feed water flow rate is reduced to 360 kg/h. The increase of cooling water temperature due to the reduced flow rate can be properly predicted. The reduced amount of condensate assures the correct measurement of gas temperature in this case. The proper prediction is also indicated in the comparison of the experimental results and prediction.

![Fig. 16 Temperature distribution at feed water flow rate of 600 kg/h and \( w_f = 0.37 \) in “Small”](image-url)
Fig. 17 Temperature distribution at feed water flow rate of 360 kg/h and $w_f=0.37$ in “Small”

Shown in Fig. 18 is the comparison of the experimental result and prediction at the steam mass concentration $w_f=0.49$ in “Small” heat exchanger when the steam flow rate is increased to 60.2 kg/h. The increase of cooling water temperature due to the increased steam flow rate can be properly predicted. The proper prediction is also indicated in the comparison of the experimental results and prediction.

Fig. 18 Temperature distribution at feed water flow rate of 600 kg/h and $w_f=0.49$ in “Small”
5. Conclusions

1. Condensation heat transfer on horizontal stainless steel tubes was investigated experimentally for the modification of mass transfer correlation. The experiment was conducted in a wide range of non-condensing gas concentration of 0~78% and the new analogy relation was proposed as,

\[
Nu = f(Re, Pr)
\]

\[
Sh = \frac{Max(1, 2 - 1.2\omega)}{1 - w_i} f\left(Re, \frac{1}{\omega} Sc\right)
\]

where function Max yields the larger value of 1 or 2\(^{-1.2\omega}\).

2. Based on the previous basic studies, a thermal hydraulic prediction method for latent heat recovery exchangers was proposed. For the condensation of steam on heat transfer tubes, the modified Sherwood number taking account of the mass absorption effect on the wall was used.

3. Two kinds of compact heat exchanger with staggered banks of bare tubes of 10.5 or 4mm in outer diameter was designed with the prediction method. The more compactness was obtained with the smaller tubes at a designed heat recovery.

4. The thermal hydraulic behavior in the compact heat exchangers of bare tubes of 10.5 or 4mm was experimentally studied with air-steam mixture gas. In the parametric experiments varying the steam mass concentration, the temperature distributions of cooling water and mixture gas were measured. The experimental results agreed well with the prediction proposed in this study and the more compactness with the smaller tubes was proved.

5. The developed prediction method was used in the design of hot water supply system at home (Motegi, 2007). Figure 19 is the equipment for the home bath using a natural gas and its thermal efficiency is raised by 17 % with the latent heat recovery. The heat output is approximately 15 kW. Now the same equipment of 3 million is used in Japan and 7700MW can be reduced with the latent heat recovery which is equivalent to the heat output from the large nuclear power plants of 2.5 units.
Appendix

Mixture gas is treated as a mixture of CO$_2$, CO, SO$_2$, O$_2$, N$_2$, and H$_2$O, and its property can be estimated with special combinations of each gas property proposed by the previous studies. Table A.1 is the molecular weight $M$ and Sutherland coefficient $S$. In the following equations, $T(K)$ is the temperature and $p(Pa)$ is the pressure.

<table>
<thead>
<tr>
<th></th>
<th>$N_2$</th>
<th>$CO_2$</th>
<th>$O_2$</th>
<th>$H_2O$</th>
<th>$CO$</th>
<th>$SO_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M$ (kg/kmol)</td>
<td>28.016</td>
<td>44.01</td>
<td>32</td>
<td>18.016</td>
<td>28.01</td>
<td>64.06</td>
</tr>
<tr>
<td>$S$ (K)</td>
<td>104</td>
<td>253</td>
<td>127</td>
<td>534</td>
<td>101</td>
<td>396</td>
</tr>
</tbody>
</table>

(1) Density
The molecular weight and fraction of each gas are defined as $M_i$ and $x_i$, respectively. The density of mixture gas can be calculated as

$$\rho = \frac{\sum_i M_i x_i}{22.4} \frac{273.15}{T} \frac{p}{1.013 \times 10^5}$$  \hspace{1cm} (A.1)

(2) Specific heat capacity

$$C_p = \sum_i C_{p,i} x_i$$  \hspace{1cm} (A.2)

(3) Viscosity

The viscosity is estimated with the methods by Wilke(1950).

$$\eta = \sum_i \frac{\eta_i}{1 + \frac{1}{x_i \sum_{j \neq i} x_j \phi_{ij}}}$$  \hspace{1cm} (A.3)

where

$$\phi_{ij} = \left[ 1 + \left( \frac{\eta_i}{\eta_j} \left( \frac{M_j}{M_i} \right)^{1/4} \right)^2 \right]^{1/2} \frac{4}{\sqrt{2}} \left[ 1 + \frac{M_i}{M_j} \right]^{1/2}$$  \hspace{1cm} (A.4)

(4) Heat conductivity

The heat conductivity is estimated with the methods by Lindsay & Bromley(1950).

$$\lambda = \sum_i \frac{\lambda_i}{1 + \frac{1}{x_i \sum_{j \neq i} x_j A_{ij}}}$$  \hspace{1cm} (A.5)

where

$$A_{ij} = \frac{1}{4} \left[ 1 + \left( \frac{\eta_i}{\eta_j} \left( \frac{M_j}{M_i} \right)^{3/4} \frac{1 + S_i / T}{1 + S_j / T} \right)^{1/2} \right] \left[ \frac{1 + S_i / T}{1 + S_j / T} \right]$$  \hspace{1cm} (A.6)

$$S_{ij} = \sqrt{S_i S_j}$$  \hspace{1cm} (A.7)
However when the gas includes the polar molecular such as H$_2$O or NH$_3$,

\[ S_{ij} = 0.733 \sqrt{S_i S_j} \quad (A.8) \]

(5) Diffusivity of steam

It is considered that a strong correlation exists between the thermal and mass diffusivities. As a first attempt, the mass diffusivity of steam in mixture gas was estimated with the well-known mass diffusivity of steam in air as

\[ \alpha = \alpha_{air} \left( \frac{\kappa}{\kappa_{air}} \right) \quad (A.9) \]

where \( \kappa \) and \( \kappa_{air} \) are the thermal diffusivities of flue gas and dry air, respectively. The diffusivity of steam in air can be expressed as (Fujii et al., 1977),

\[ \alpha_{air} = 7.65 \times 10^{-5} \frac{T^{11/6}}{p} \quad (A.10) \]

(6) Steam mass concentration

\[ w = \frac{p_{H_2O} M_{H_2O}}{\sum_i p_i M_i} \quad (A.11) \]

where \( p_i \) is the partial pressure of each gas in the mixture.

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