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MIXING PROCESS BY MOLECULAR DIFFUSION AND NATURAL CIRCULATION OF TWO COMPONENT GASES IN A REVERSED U-SHAPED CHANNEL

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Abstract

This study is to investigate an effect of natural convection (or natural circulation) on a mixing process by molecular diffusion in a reverse U-shaped channel consisting of two component gases. It is confirmed that these phenomena appear when the depressurization accident occurs in the very high temperature reactor (VHTR). The experiment has been performed regarding the combined phenomena of molecular diffusion and natural circulation in a two parallel vertical slots filled with two component gases. The experimental results show that the transport phenomena by the molecular diffusion are influenced by the natural circulation of the gas mixture in the reverse U-shaped channel.

Introduction

The technological developments of the very high temperature reactor (VHTR) are currently interested worldwide. Besides its broad economical appeals resulting from unique high temperature capability, the reactor provides inherent and passive safety and aims at enhanced safety goal. Japan Atomic Energy Agency (JAEA) has successfully built and operated the 30MWt High Temperature Engineering Test Reactor (HTTR) and is now pursuing design and development of commercial systems such as the 300MWe gas turbine high temperature reactor GTHTR300C (Gas Turbine High Temperature Reactor 300 for Cogeneration)[1]. A depressurization accident is one of the design-basis accidents of the VHTR. When the pipe rupture accident occurs, air is expected to enter the reactor core from the breach and oxidize in-core graphite structures. In order to predict or analyse the process of air ingress during the depressurization accident of the VHTR, it is very important to develop computer programs and to validate them by experiments.

Previous studies focused mostly on molecular diffusion and natural circulation of the two-component gas mixture in a reverse U-shaped tube and in a simple test model of the HTTR [2]. In order to investigate the basic features of the flow behaviour of the multi-component gas mixture, consisting of helium (He), nitrogen (N_2), oxygen (O_2), carbon dioxide (CO_2), carbon monoxide (CO), etc., experimental and numerical studies were performed on the combined phenomena of molecular diffusion and natural circulation of the multi-component gas mixture with the graphite oxidation reaction in the reverse U-shaped tube [3]. The numerical results were in good agreement with the experimental ones regarding the density change of the gas mixture, the mole fraction change in the gas species and the onset time of natural circulation of air. Furthermore, the objective of these studies is to investigate the air ingress process and to develop the passive safe technology for the prevention of air ingress [4]. Recently, a density-gradient driven air ingress stratified flow was analysed using a CFD code for the NGNP, which is the U.S. VHTR [5, 6]. Authors have reported the mixing process by natural convection and molecular diffusion of two-component gases in a stable stratified fluid layer [7]. According to the report, the mixing process by molecular diffusion in the vertical stratified fluid layer was affected significantly by the localized natural convection induced by the slight temperature difference (about ten degree or more) between both vertical walls.

1. Basic feature of air ingress process in a reverse u-shape channel

Experimental and numerical studies were reported on the combined phenomena of molecular diffusion and natural circulation in a two-component (N_2/He) gas system in a reverse U-shaped tube [2, 3]. Figure 1 shows an example of the mole fraction and the ingress velocity changes of nitrogen in the reverse U-shaped tube. The solid line in the figure is the ingress velocity obtained by the hot-wire anemometer attached at the lower end of the high-temperature side pipe. The symbols (o) and (Δ), respectively, represent the mole fraction changes at the same distance (600 mm) from the lower ends of the both side pipes. The mole fraction of nitrogen at the high-temperature side pipe is higher than that at the low-temperature side pipe. This is because nitrogen is transported upward by molecular diffusion from the both ends of the pipes and by natural circulation of the gas mixture having the very slow velocity from the high-temperature side to the low-temperature side pipe.

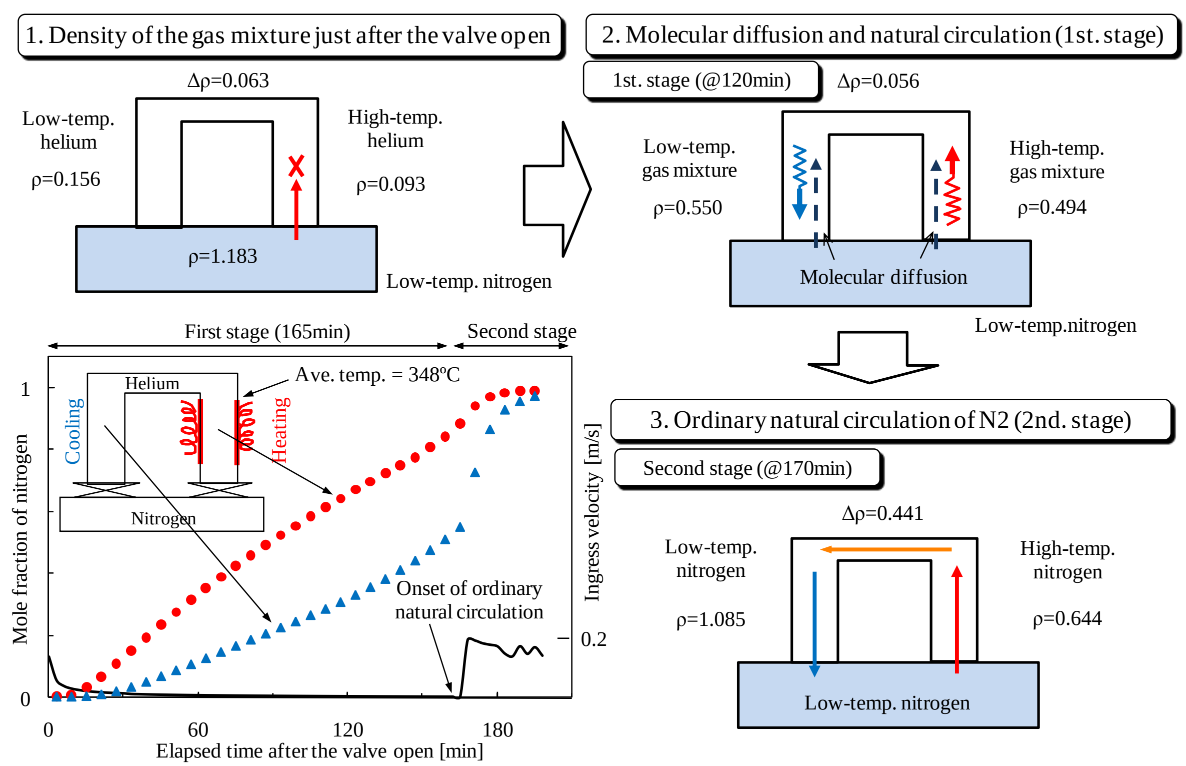

Figure 1 Basic feature of nitrogen ingress process

According to the results obtained, the density of the gas mixture in the reverse U-shaped tube gradually increases as nitrogen enters as a result of molecular diffusion and because of a very weak natural circulation of the gas mixture. The calculated velocity of this very weak natural circulation of the gas mixture is about 10^{-6}~10^{-3}m/s. The Reynolds number, Re, based on the tube diameter is about 10^{-4}<Re<1. The ordinary natural circulation of nitrogen, which is about 0.2m/s (Re≈200) in

Fig.1, occurs suddenly throughout the reverse U-shaped tube, because the buoyancy force has risen to such an extent as to bring about the natural circulation. The time required from the first stage to the second stage is about 10 seconds in the ingress process. Therefore, the production process of the second stage is a rapid phenomenon because the duration time of the first stage is about 165 minutes in the reverse U-shaped tube.

In the previous paragraph, the ingress process of nitrogen when helium was filled in the reverse U-shaped tube composed of the high-temperature and the low-temperature passages was described. However, it is necessary to investigate the influences of the localized natural circulation in the parallel channels onto the process of nitrogen ingress during the primary-pipe rupture accident because more than three parallel channels with different temperatures exist in the prismatic type VHTR core. According to the Ref.[8], if the wall temperature of the parallel channels is equal to the gas temperature, the localized natural circulation of the gas mixture will not occur in the parallel channels. Therefore, the mole fraction change and the ingress process of nitrogen are the same as in the reverse U-shaped tube experiment. The mole fraction changes of nitrogen in the parallel channels are almost the same because of the localized natural circulation generated in the parallel channels. Therefore, the mole fraction of nitrogen in the channels and plenum almost equals. The duration time of the first stage becomes short when the localized natural circulation is generated between the parallel channels. On the other hand, the duration time becomes long when the temperature of the parallel channels is equal. The duration time of the first stage is affected not only by the average temperature of the entire channels but also strongly by the localized natural circulation in the parallel channels. Therefore, it is necessary to evaluate quantitatively the amount of transported air not only by molecular diffusion but also by natural circulation in the parallel channels with different heat input when the air ingress accident occurs in the VHTR.

In general, mixing processes of two component gases in a vertical stable stratified fluid layer is often governed by molecular diffusion. When a stable stratification is formed in a vertical slot with two component gases which is different density, a rate of transportation will be different by a mutual diffusion coefficient. On the other hand, it is expected that natural convection will occur in the vertical slot when one side wall is heated and the other side wall is cooled. When the stable stratification is formed with the two component gases and the two vertical parallel walls of the slot is kept at different temperature, the transport process of the gases becomes more complex. In this case, the heavier gas diffuses into the lighter gas. In addition to that these gases will also be transported by natural convection. Both phenomena may produce at the same time during the air ingress process of the primary pipe rupture accident. According to the previous experiments [2,4], molecular diffusion and natural convection would have occurred simultaneously in the annular passage between the inner barrel and the water-cooled jacket. The range of Rayleigh number based on the width of the annular passage is about $0 < Ra_d < 3.26\times10^5$ [2] and $Ra_d < 1.56\times10^6$ [4] respectively. Rayleigh number based on the width of the annular passage of the HTTR or the GTHTR-300C will be two order or bigger than that of the simulated apparatus. Therefore, it is necessary to know which phenomenon becomes dominant for the mixing processes of the two component gases in the vertical stable stratified fluid layer. It is also important to quantitatively evaluate an influence of natural convection on the mixing processes by molecular diffusion.

There are many studies regarding natural convection of the vertical slot to obtain the flow characteristics of the density stratified layer by changing of the fluid temperature. Eckert and Carlson [9] had carried out an experiment regarding natural convection of air in a vertical slot ($2.1 <$ height/width (H/d) <46.7). They have classified the flow characteristics to conduction, transition, and boundary layer regions by using Grashof number, *Gr*, and the aspect ratio, *H/d*. In addition to that, Elder [10, 11] had performed an experiment and analysis regarding natural convection in a vertical slot. The range of the aspect ratio is $1<H/d<60$ and Prandtl number, *Pr*, is 10^3. If the Rayleigh number, *Ra*, exceed the critical number, $3.0\times10^5\pm30\%$, it is found that a unicellular convection change to a multi-cellular convection.

On the other hand, there is a double diffusive convection [12] as similar phenomena related with coexisting of natural convection and molecular diffusion. In this phenomenon, natural convection will occur by the buoyancy of density difference between two mediums or one medium and temperature distribution. Especially, it is called thermo-salt convection if the two elements are the temperature and salt. There are a lot of studies in the field of the ocean physics. However, the most of those are the experimental studies using liquid. Usually the density change along with transporting of medium is small in case of liquid. Then, not only the density difference but also the diffusion coefficient is also small compared with gas system.

According to the previous analytical study [13], the buoyancy term of the momentum-conservation equation may be expressed by a linear combination of thermal buoyancy and concentration buoyancy term when the density of the binary gas mixture is approximated by Eqn.(1).

$$1/\rho = 1/\rho_0 \{1+\beta_T(T-T_0)+\beta_C(X_B-X_{B\min})\} \tag{1}$$

Where β_T and β_C are given by $\beta_T=1/T_0$, $\beta_C=1-M_B/M_A$. Here, β_T and β_C are the thermal coefficient of volumetric expansion and concentration coefficient of volumetric expansion, respectively. Then, M_A and M_B are the molecular weight of species *A* and *B*. X_B is the mole fraction of species *B*. The subscript *A* refers to the heavy gas and the subscript *B* to the light gas. Both gases are assumed to be ideal. In the binary gas system, however, it will be difficult to divide the two buoyancy terms if the differences of the temperature and gas density are so large.

From the view point of buoyancy forces which arise from a combination of temperature and species concentration effects of comparable magnitude [14], the density of the mixture will indicate in generally as follows. We expand ρ in a Taylor series in *T* about some reference temperature T_0 [15].

$$\rho = \rho_0 + \left.\frac{\partial\rho}{\partial T}\right|_{T0}(T-T_0)+\cdots\cdots = \rho_0 - \rho_0\beta_0(T-T_0)+\cdots\cdots \tag{2}$$

Here, we have introduced ρ_0 the density at temperature T_0, and the coefficient of volume expansion β_T, also evaluated at T_0. If the pressure effect negligible, the coefficient of volume expansion is defined by the following equation.

$$\beta_T = -(1/\rho)(\partial\rho/\partial T)_p = -1/T_0 \tag{3}$$

In systems with concentration inequalities, as well as temperature inequalities, we also expand ρ in a double Taylor series in *T* and X_B for a two-component system,

$$\rho = \rho_0 + \left.\frac{\partial\rho}{\partial T}\right|_{T0,XB\min}(T-T_0)+\left.\frac{\partial\rho}{\partial X_B}\right|_{T0,XB\min}(X_B-X_{B\min})+\cdots\cdots \tag{4}$$

$$\rho = \rho_0 - \rho_0\beta_T(T-T_0)-\rho_0\beta_C(X_B-X_{B\min}) \tag{5}$$

Where $\beta_C = -(1/\rho)(\partial\rho/\partial X_B)_T$ is a quantity (defined analogously to β_T) that indicates how much the density varies with composition.

Then, $\beta_C = -(1/\rho)(\partial\rho/\partial X)_T = -(1 - M_A/M_B)$.

Finally, the density of mixture indicate in Eqn.(1). The buoyancy term in the momentum-conservation equation can be obtained as follows.

$$(\rho_0 - \rho)/\rho_0 = \beta_T(T - T_0) + \beta_C(X_B - X_{B\min}) \tag{6}$$

As mentioned above, it will be difficult to divide the two buoyancy terms if the differences of the temperature and gas density are so large. Each gas species A and B can be obtained as follows assuming these gases as an ideal.

$$1/\rho_A = 1/\rho_{A0}\{1 + \beta_T(T - T_0)\} \tag{7}$$

$$1/\rho_B = 1/\rho_{B0}\{1 + \beta_T(T - T_0)\} \tag{8}$$

Therefore, $$\rho = \rho_A X_A + \rho_B X_B = \frac{1}{1 + \beta_T(T - T_0)}(\rho_{A0} X_A + \rho_{B0} X_B) \tag{9}$$

Finally, the density of the gas mixture can be obtained in the following equation.

$$\rho = \rho_0 \frac{1 - \beta_C X_B}{1 + \beta_T(T - T_0)} \tag{10}$$

In addition to that, there are few experimental studies using two component gases because it will be difficult to perform experiment from the density measurement view point. As the first stage of this study, especially, the experiments have performed using several component gases under the regions of the conduction and transition. This paper describes the results of the experiment and the mixing processes of two component gases in the vertical slot.

2. Experimental apparatus and method

Figure 2 and 3 show a schematic drawing of an experimental apparatus consisting of two vertical slots. The vertical slot of the left hand side consists of the heated wall and the cooled wall in Fig.2. The vertical slot of the right hand side consists of the two cooled walls. Each slot is connected at the top of the slot. The two vertical slots are composed the reverse U-shaped channel. The bottom ends of the both slots are connected to a gas storage tank. The vertical walls of the heated side slot are made of copper with thickness of 3mm. A stainless sheath heater and a water cooling pipe made of copper are attached to the heated wall and the cooled wall, respectively. These walls are covered by an insulator which is 50mm in thickness. The dimension of the walls is 598mm in height (H) and 208mm in width. The width (d) and the aspect ratio (H/d) of the heated side slot are 20mm and 25, respectively. The vertical walls of the cooled side slot are made of stainless steel. The dimension of the cooled side slot is same as the heated side slot. The dimension of the gas storage tank is 248mm × 548mm × 398mm. The both slots and the gas storage tank are separated by a partition plate. The wall and gas temperatures are measured by a K-type thermocouple. The 8 thermocouples which is 1.0mm in diameter are inserted into the slot from the cooled wall side to measure the gas temperature. The numbers of the measurement points are 8 on the surface of the heated wall, 6 on the surface of the cooled wall, 3 on the surface of the cooled slot and 2 in the gas storage tank. The numbers of the measurement points were decided by referring of the previous experimental results [16]. The locations of the thermocouples are provided in Table 1.

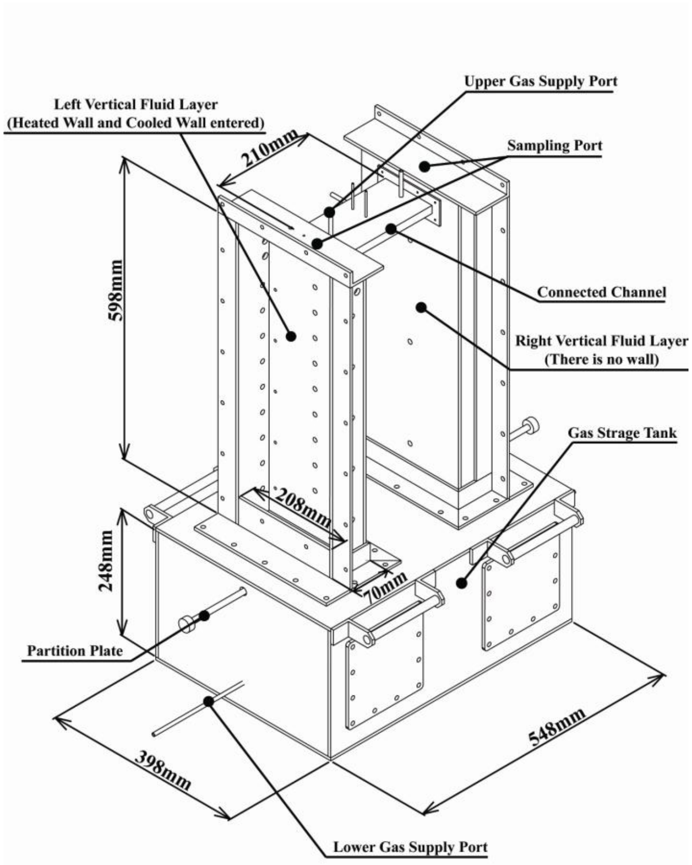

Figure 2 Experimental apparatus (All units are in cm)

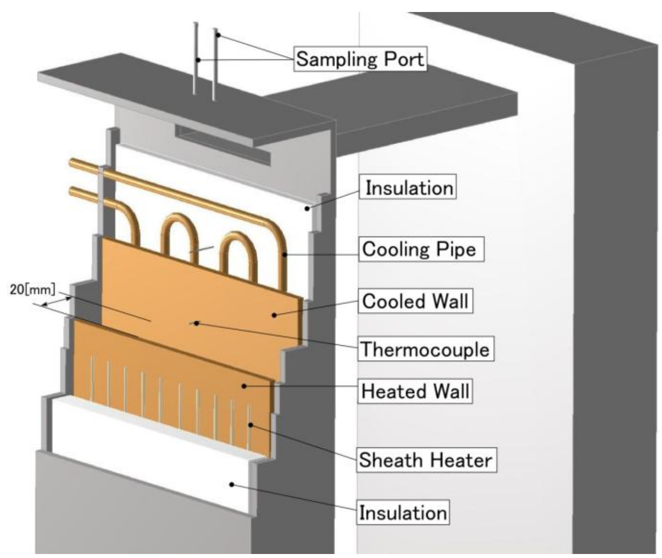

Figure 3 Heated side slot

Combination of nitrogen/argon(N_2/Ar), neon/argon(Ne/Ar), etc. is used as the two component gas system. The gas mixture is sampled from the top of the slot. The sampled gas is returned to the top of the slot. The density change of the gas mixture and the gas temperature distribution in the channel are obtained. Considering the errors induced by the thermocouples, the scanner junction and the digital data acquisition control unit, the entire accuracy of the temperature measurement was within ±0.5ºC. The mole fraction of argon and the density of the gas mixture are obtained as follows. The two component gases and the gas mixture are assumed to be ideal gas. The velocity of sound can be calculated by the following equation [17].

$$v_s = \sqrt{\gamma RT / M} \tag{11}$$

Thus, the molecular weight of the gas mixture is also obtained by the following equations.

$$M = \gamma RT / v_s^2 \tag{12}$$

Finally, the mole fraction of argon can be obtained by the molecular weight by assuming the gas mixture obey the linear combination of the mixing rule [18].

$$X_A = (M - M_B) / (M_A - M_B) \tag{13}$$

The gas mixture was sampled from the top of the heated side slot. The velocity of sound was measured and the mole fraction of argon was obtained. The relative uncertainties in the density of the gas mixture and in the mole fraction of argon are found to be ±10%.

The experiment was carried out to obtain the distribution and the fluctuation of the gas temperature, and the density change of the gas mixture. The experimental procedure of the N_2/Ar experiment is as follows. At first, the two vertical slots and the gas storage tank were filled with nitrogen gas as the light gas. Argon gas as the heavy gas was poured into the bottom part of the gas storage tank and nitrogen gas was released from the top of the storage tank. The one side wall was heated and the other side was cooled of the heated side slot. The gas pressures in the slots and the gas storage tank were kept at the atmospheric pressure. After the temperature reached the steady state condition (within ±0.2 ºC/h), the partition plate was removed and the experiment was started.

Table 1 Location of thermocouples

T/C position (H-Gas)		T/C position (H-Wall)		T/C position (C-Wall)		T/C position (C-Gas)	
No.	Height(mm)	No.	Height(mm)	No.	Height(mm)	No.	Height(mm)
1	490	10	490	19	440	23	475
2	440	11	440	20	340	24	275
3	390	12	390	21	240	25	75
4	340	13	340	22	140		
5	290	14	290	Sampling port (H)		Sampling port (C)	
6	240	15	240	No.	Height(mm)	No.	Height(mm)
7	190	16	190	26	598	27	598
8	140	17	140	28	50		
9	90	18	90				

3. Experimental results and discussions

Figures 4 to 7 show the typical experimental results of N_2/Ar experiment. Figure 4 shows the temperature change of the gas mixture with time elapsing at the heated side slot in the N_2/Ar experiment. The temperature difference between the heated wall and cooled wall was 30K. The temperature change of the gas mixture with time elapsing at the cooled side slot is shown in Fig.5. From the figures, it was confirmed that the temperature of the gas mixture in the heated side slot decreased and the one in the cooled side slot increased. The change in temperature occurs at about 12000 seconds after the experiment starts. This is because the ordinary natural circulation through the reverse U-shaped channel will occur suddenly at this time. This phenomenon is almost same as the reversed U-shaped tube experiment [2].

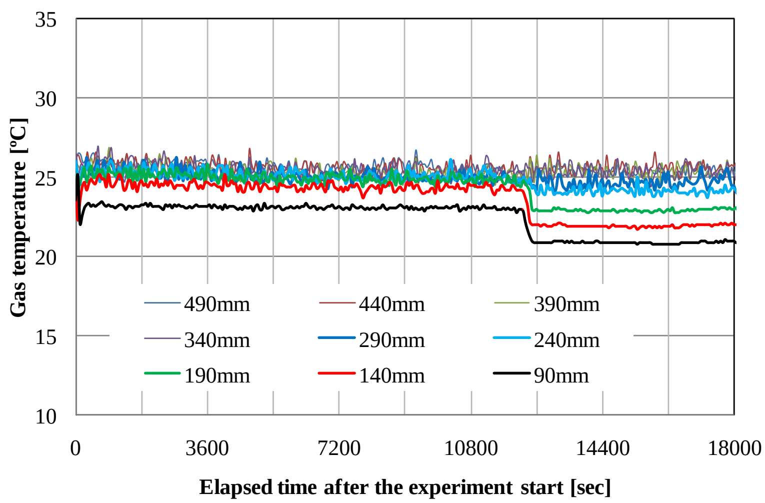

Figure 4 Gas temperature change with time in the heated side slot

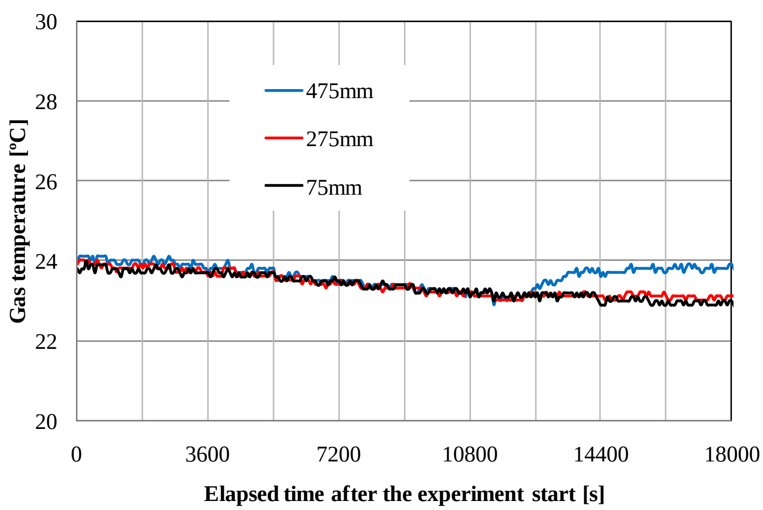

Figure 5 Gas temperature change with time in the cooled side slot

Figures 6 and 7 show the temperature change of the gas mixture when the temperature difference was 50 ºC. The onset time of the ordinary natural circulation in the reverse U-shaped channel will depend not only on the average temperature of the heated side slot but also on the temperature difference between the both channels. The temperature of the gas mixture in the heated side slot decreased stepwise at about 9000 seconds.

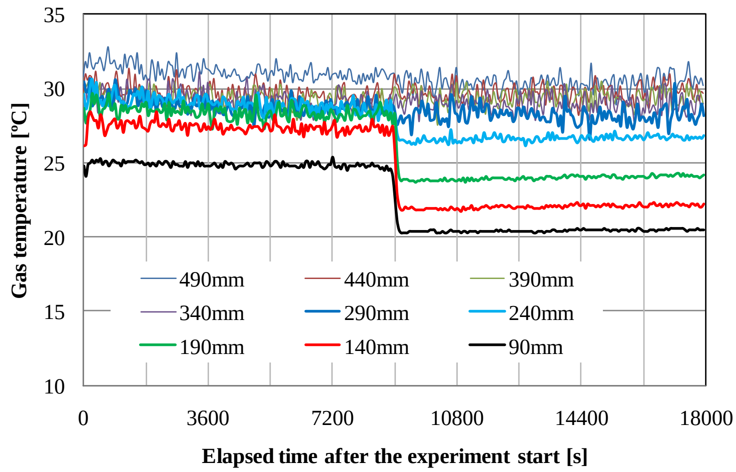

Figure 6 Gas temperature change with time in the heated side slot

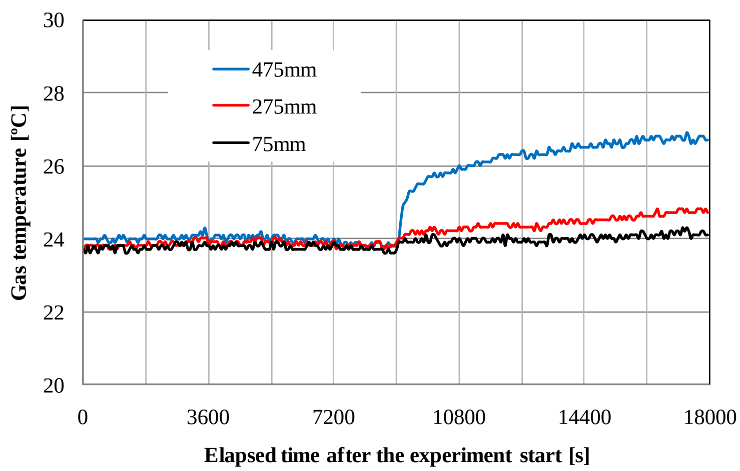

Figure 7 Gas temperature change with time in the cooled side slot

Figure 8 shows the mole fraction change with time in the top of the heated side slot. A vertical axis shows a mole fraction of argon gas and a horizontal axis shows an elapsed time after the experiment starts. The black solid line in the figure indicates the density change of the gas mixture in the isothermal experiment (ΔT=0ºC). The mixing process of the two component gases under the isothermal condition is governed by molecular diffusion. The onset of increasing the density of the gas mixture is about 60 minutes after the opening of the partition plate. Other solid lines indicate the density change in which the vertical walls kept at different temperatures. The rise rate of the density of the gas mixture increases with increasing of the temperature difference of the two vertical walls. The mole fraction of argon gas at the top of the heated side slot will begin to increase in about 10 min in the case of heated condition. Figure 9 shows the mole fraction change with time in the top of the cooled side slot. The mole fraction of argon gas at the top of the cooled side slot will increase slowly compared with the one at the top of the heated side slot. This is because the argon gas will transport by only molecular diffusion between the heated and cooled side slot.

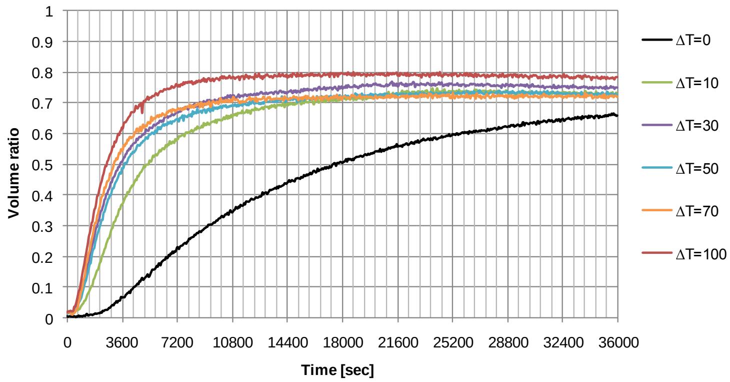

Figure 8 Mole fraction change with time at the top of the heated side slot

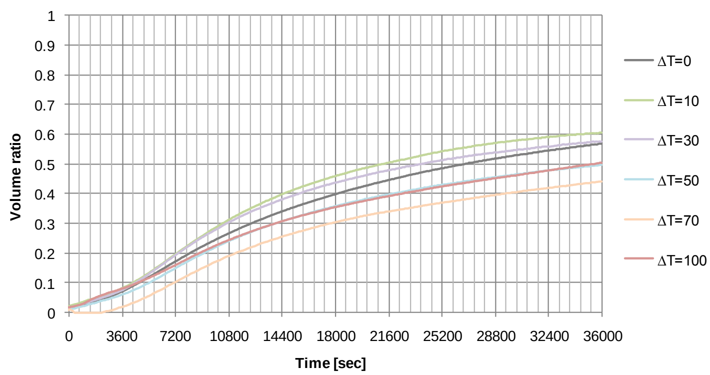

Figure 9 Mole fraction change with time at the top of the cooled side slot

Figure 10 shows the density change of the gas mixture with time elapsing at the top of the heated side slot. A vertical axis shows a density of the gas mixture and a horizontal axis shows the elapsed time after the experiment starts. The density of the gas mixture at the top of the heated side slot began to increase in about 60 min in the case of molecular diffusion. In the case of heating, the time when the density of the gas mixture begins to increase has been shortened compared with the case of molecular diffusion. It was confirmed the density change of the gas mixture increasing with time elapsing at the top of the heated side slot was same as the previous experiment [16] qualitatively.

Figure 11 shows the mole fraction change with time at the top of the heated side slot in the Ne/Ar experiment. A vertical axis shows a mole fraction of argon gas and a horizontal axis shows an elapsed time after the experiment starts. This result is almost the same as the N_2/Ar experiment.

It was mentioned in the previous paragraph, the transport process of the gases becomes more complex when the vertical parallel walls is kept at different temperature under the stable stratified fluid layer. In this case, the heavier gas diffuses into the lighter gas. In addition to that these gases will also be transported by natural convection. Both phenomena for air ingress may produce at the same time during the primary pipe rupture accident. According to the previous experiments using the simulated apparatus of the reactor [2, 4], molecular diffusion and the localized natural convection would have occurred simultaneously in the annular passage between the inner barrel and the water-cooled jacket. The annular passage simulated the annular channel between the permanent reflector and the pressure vessel. Thus, it is necessary to know which phenomenon becomes dominant for the mixing processes of helium and air in the vertical stable stratified fluid layer. However, Rayleigh number based on the width of the annular passage of the HTTR or the GTHTR-300C is bigger than that of the simulated apparatus. It will be difficult to perform the experiment simulated under the condition of Rayleigh number for real reactor. Therefore, it will be important to carry out the numerical simulation by the analysis code which was verified by the experiment. These experimental results will be useful for the verification of the analysis code.

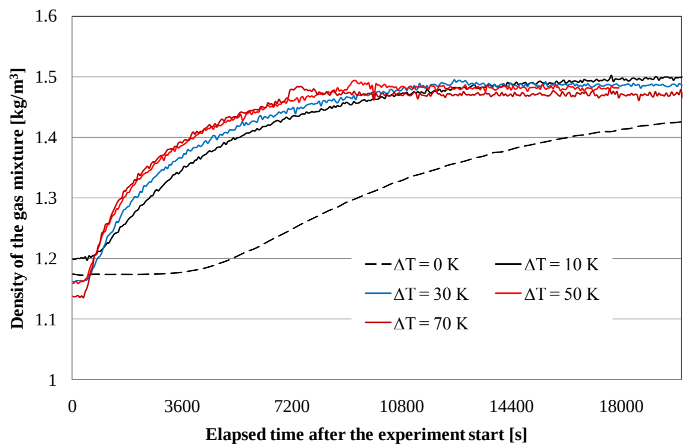

Figure 10 Gas density change with time in the top of the heated side slot

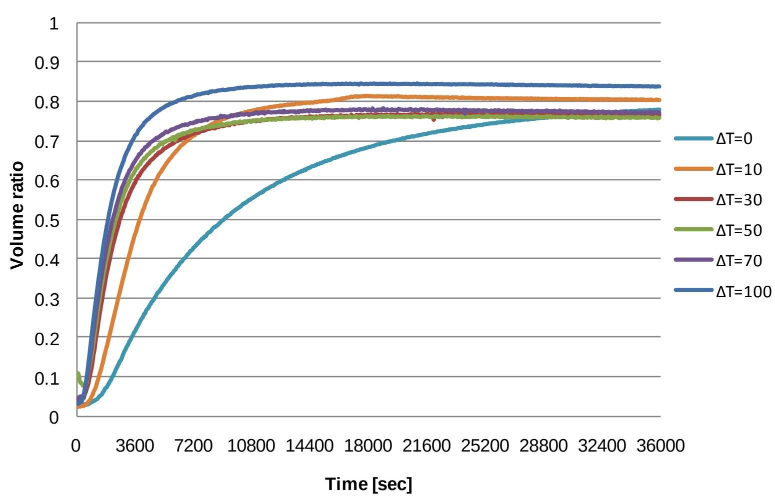

Figure 11 Mole fraction change with time at the top of the heated side slot

4. Conclusion

An experiment on the mixing processes of the two component gases in the reverse U-shaped channel has been carried out to research the effectiveness of natural convection on the mixing processes by molecular diffusion in the stratified density layer. The following results were obtained.

It was found that the mixing process by molecular diffusion was affected significantly by the natural convection induced by the slight temperature difference between vertical walls. The amount of transported argon gas was mainly limited by natural convection in the N_2/Ar and Ne/Ar experiment. So, the influence of natural convection on the transport phenomena of molecular diffusion clearly appeared to the experiments.

It was also confirmed that the temperature of the gas mixture in the heated side slot decreased and the one in the cooled side slot increased. The change in temperature occurs at about 12000 seconds after the experiment starts in this apparatus. This is because the ordinary natural circulation through the reverse U-shaped channel will occur suddenly at this time. This phenomenon is almost same as the previous reversed U-shaped tube experiment. The localized natural convection may affect to the onset time of the natural circulation. The experimental results showed that the transport phenomena by the molecular diffusion were influenced by the localized natural convection and natural circulation of the gas mixture. Therefore, it is necessary to evaluate quantitatively not only the localized natural convection in the slot but also natural circulation through the channel.

We are now carrying out the helium/argon and helium/nitrogen experiments. In order to evaluate the amount of air ingress, the two component gas transport analysis using the CFD code will be carried out in the future.

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