VALIDATION OF A MULTIPHASE CFD MODEL WITH MASS TRANSFER FOR XENON REMOVAL IN MOLTEN SALT REACTOR

BY

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THESIS

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ABSTRACT

Among the distinguishing features of molten salt reactors, the possibility of online fuel processing is especially promising. With removal of unfavorable fission products, the molten salt reactor could act as an effective converter reactor and could possibly operate in a load-following manner. The fuel life is also extended. This document is dedicated to developing a computational fluid dynamics (CFD) model as a design tool for the xenon removal system in fueled salt reactors. Beginning with the review of gaseous fission product removal, the possibility and limitation of CFD with multiphase species transfer modeling are discussed. The necessity of experimental validation and sensitivity studies is explained. Following this discussion, the details about the Eulerian-Eulerian two-fluid model and constitutive relations are presented. The detailed procedures and results of the three validation experiments are reported. Different phase interaction mechanisms, material properties, and constitutive relations are compared with each other and with the results from the experiments. These comparisons lead to a CFD model, which is further validated against different flow conditions. The CFD model can accurately predict the void fraction and velocity field in our experiment. The mass transfer model will be further developed and validated with the completion of the sparging experiments.
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# TABLE OF CONTENTS

Chapter 1: Introduction to Molten Salt Reactor ................................................................. 1
Chapter 2: History of Fission Product Removal ................................................................. 6
Chapter 3: Physical Model Description .................................................................................. 16
Chapter 4: Validation Experiments ......................................................................................... 46
Chapter 5: Simulation Results and discussion ...................................................................... 64
Chapter 6: Summary and Future Work .................................................................................. 96
REFERENCES .......................................................................................................................... 100
APPENDIX A: Uncertainty Analysis ...................................................................................... 107
APPENDIX B: UDFs ................................................................................................................. 114
Nomenclature .......................................................................................................................... 120
LIST OF FIGURES

Figure 3.1: Illustration of Two-Resistance Model ............................................................... 24
Figure 3.2: Illustration of One-Resistance Model ................................................................. 24
Figure 3.3: Density of FLiNaK ............................................................................................ 38
Figure 3.4: Viscosity of FLiNaK ......................................................................................... 38
Figure 3.5: Thermal Conductivity of FLiNaK ....................................................................... 40
Figure 3.6: Specific Heat Capacity of FLiNaK ......................................................................... 40
Figure 3.7: Solubility of Helium in FLiNaK with Pressure .................................................... 43
Figure 3.8: Solubility of Noble Gases in FLiNaK ................................................................. 43
Figure 4.1: Bubble Column with Water Box Used in Our Experiment ......................... 47
Figure 4.2: Bubble Column with Flange and the Molten Salt Xenon Experiment ............ 48
Figure 4.3: Obtained Image and Bubble Identification ........................................................... 50
Figure 4.4: The Convergence of Void Fraction with Increasing Frames ............................ 52
Figure 4.5: The Average Void Fraction at Different Superficial Gas Velocity ................ 52
Figure 4.6: Process of Obtaining the 2D Void Fraction Profile ........................................... 54
Figure 4.7: The Distribution Function $\eta$ and its Effect ...................................................... 54
Figure 4.8: A Pair of Images Taken 0.002s apart ................................................................. 55
Figure 4.9: The Masked Image and Instant Velocity Field ................................................... 58
Figure 4.10: Velocity Field and Axial Velocity from PIV Experiment at $U_g=2.5\text{mm/s}$ .... 58
Figure 4.11: Kinetic Energy of the Liquid Phase $k^e$ at $U_g=2.5\text{mm/s}$ ......................... 59
Figure 4.12: Dissolved Oxygen Concentration during Air Bubble Sparging .................. 62
Figure 4.13: Relation between $ka_i$ and $U_g$ .................................................................... 63
Figure 5.1: Grid Independence Study – Void Fraction ......................................................... 65
Figure 5.2: Sensitivity of Interfacial Forces ......................................................................... 67
Figure 5.3: Comparison of Drag Models – Void Fraction ..................................................... 69
Figure 5.4: Comparison of Lift Models – Projected Void Fraction ....................................... 71
Figure 5.5: Comparison of Turbulence Model – Axial Velocity ........................................... 73
Figure 5.6: Comparison of Turbulence Model (L: RNG, R: Realizable) – Void Fraction .... 74
Figure 5.7: Sensitivity of Bubble Induced Turbulence – Void Fraction ......................... 74
Figure 5.8: Sensitivity of Boundary Condition – Void Fraction .......................................................... 76
Figure 5.9: Sensitivity of Boundary Condition – Liquid Velocity ......................................................... 76
Figure 5.10: Sensitivity of Bubble Diameter – Void Fraction ............................................................... 78
Figure 5.11: Sensitivity of Viscosity ........................................................................................................ 81
Figure 5.12: Sensitivity of Density ........................................................................................................... 82
Figure 5.13: Sensitivity of Temperature ................................................................................................... 82
Figure 5.14: Comparison of Predicted Averaged Void Fraction with Experiment ................................. 84
Figure 5.15: Void Fraction Profile 6.8 cm Above the Inlet at 2.5mm/s $U_g$ ............................................. 85
Figure 5.16: Void Fraction Profile 3.8 cm Above the Inlet at 2.5mm/s $U_g$ ............................................. 85
Figure 5.17: Void Fraction Profile 9 cm above the Inlet at 2.5mm/s $U_g$ ................................................. 86
Figure 5.18: Comparison of Liquid Velocity Field at 2.5mm/s $U_g$ .................................................... 86
Figure 5.19: Comparison of Liquid Axial Velocity, $U_z$ [m/s], at 2.5mm/s $U_g$ .................................... 88
Figure 5.20: Comparison of Liquid Axial Velocity at 3.8cm ............................................................... 88
Figure 5.21: Comparison of Liquid Axial Velocity at 6.8cm ............................................................... 89
Figure 5.22: Comparison of Liquid Axial Velocity at 9cm ................................................................. 89
Figure 5.23: Validation of Diffusion within Phases ................................................................................. 91
Figure 5.24: Comparison of Different Species Transport Model at Free Surface ............................... 92
Figure 5.25: Concentration Profile During the Simulation ................................................................. 94
Figure 5.26: Comparison of the Dissolved Oxygen Concentration ..................................................... 95
LIST OF TABLES

Table 3.1: Models of drag coefficient $C_D$ ................................................................. 31
Table 3.2: Density Correlation of FLiNaK ................................................................. 37
Table 3.3: Viscosity Correlation of FLiNaK ............................................................... 39
Table 3.4: Thermal Conductivity Correlation of FLiNaK .............................................. 41
Table 3.5: Specific Heat Capacity Correlation of FLiNaK .............................................. 42
Table 3.6: Surface Tension Correlation of FLiNaK ....................................................... 42
Table 3.7: Solubility of Noble Gases in FLiNaK ........................................................... 45
Table 5.1: Summary of Models and Setups for Base Case ........................................... 64
Table 5.2: Grid Independence Study with Averaged Void Fraction ................................ 65
Table 5.3: Simulation and experimental volumetric mass transfer at $U_g=2.5mm/s$ ........... 93
CHAPTER 1: INTRODUCTION TO MOLTEN SALT REACTOR

Molten Salt Reactor (MSR) refers to the kind of nuclear reactors whose primary loops are filled with molten salt. Considering the fuel form of existing design, MSR could be subdivided into two different types as the liquid-fueled MSR and the solid-fueled MSR. The liquid-fueled MSR, unlike most reactor designs, uses a liquid fuel form, where fissile material is dissolved in the molten salt [1, 2]. This unique design introduces several distinguishing features compared with conventional LWR (Light Water Reactor). The solid-fueled MSR design is introduced in recent years, with the ambition to overcome some technical difficulties with the liquid-fueled design while retaining favorable features of molten salt as a heat transfer fluid. The main characteristics of molten salt are their high boiling point, low vapor pressure, low chemical reactivity, and similar thermal conductivity, viscosity and volumetric heat capacity to water. Low vapor pressure allows the reactor to operate at a much higher temperature than LWR with its loop pressure only slightly higher than atmosphere [3], which increases the thermal efficiency of the reactor without the risk introduced in a high pressure system. The comparable heat capacity and thermal conductivity retain the exceptional safety margin of LWRs during accidents. Moreover, fluoride based molten salt does not react violently with water, structure metal or air at high temperature, therefore steam explosion and hydrogen explosion encountered in commercial light water reactors are less likely to happen for a molten salt reactor.

The liquid fueled MSR not only shares the heat transfer features brought by utilizing molten salt, as discussed above, it also has several distinct virtues which make it favorable as an advanced reactor. One important feature of a liquid fueled MSR is the intrinsic safety against core melting. In case of power surge, the fuel salt will be drained from the primary loop into a storage
tank where it is subcritical. Moreover, the liquid fuel possesses a larger thermal expansion coefficient compared with solid fuel, which leads to larger negative temperature coefficient for reactivity. Another inspiring design feature of a liquid fueled MSR is online processing of the fuel salt. Since fissile material is dissolved in the molten salt, it is transported out of the reactor core region during circulation. By diverting a part of the fuel salt from the primary loop, unfavorable fission product could be removed, and the processed salt would reenter the primary loop. This feature, with online refueling, enables many interesting fuel-cycle designs, one of which is the Thorium 233-Uranium cycle. With different fuel compositions, the specific reactor could be a once-through convertor reactor, a self-sustainable reactor or a breeder reactor based on the conversion ratio. The neutrons could either be in the fast spectrum or the thermal spectrum based on specific reactor design [4]. However, the fact that the fission product is transported out of the pressure vessel into the whole primary loop may not be favorable in terms of radiation protection and plant maintenance. The radioactivity of the extracted fission product should be properly handled.

The ideas of using molten salt and dissolved fissile material were first proposed by ORNL as part of its Aircraft Nuclear Propulsion project (ANP) in 1949, which aimed at creating a nuclear-powered aircraft for military use [5]. As the project went on, ORNL proposed the Aircraft Reactor Experiment (ARE) to build a low power test reactor, which was successfully operated with a maximum power of 2.5MW for 9 days in 1954. The design of a liquid-fueled reactor was successfully demonstrated in this project. However, due to the cost and the development of intercontinental ballistic missile, an aircraft with unlimited range lost its military importance. As the interest into a nuclear-powered aircraft began to diminish, ORNL decided to adapt the ARE design into a civilian nuclear reactor [6]. These efforts led to the conceptional design of a Molten
Salt Breeder Reactor (MSBR) operating on a thorium-uranium fuel cycle. The MSBR design is a two-region, two-fluid system. The fuel salt with uranium fluoride is mostly for power generation, which is surrounded by the blanket salt with thorium fluoride for breeding. On site fuel recycling is considered, combining the possibility of online processing and refueling. To further develop this idea, ORNL began its Molten Salt Reactor Experiment (MSRE) in 1960. The MSRE was designed to use essentially the same material as in the MSBR, with a different fuel salt component as LiF − BeF$_2$ − ZrF$_4$ − UF$_4$ (65.0 − 29.1 − 5.0 − 0.9 mole%). For economy and simplicity, breeding was not part of the experiment. Thus, the only fissile material dissolved in the fuel salt was uranium, rather than a mixture of thorium and uranium. The blanket salt loop is also not included, since breeding is not a part of the experiment. The MSRE reactor first reached criticality on June 1, 1965 with addition of highly enriched $^{235}\text{U}$ and was finally shutdown in December 1969. Various experiments were done during this period on reactor dynamics, material corrosion, fuel inspection, operation with $^{233}\text{U}$, xenon stripping, fission product deposition, tritium behavior, plutonium additions and others [2].

Despite the successful operation of MSRE, subsequent proposal for MSBR was rejected as the US decided to focus on the research of liquid metal fast breeder reactor (LMFBR). The development status of MSBR is summarized in a later report [7]. Nevertheless, the development of breeder reactor wasn’t successful globally and became uneconomic, partly because of the seized increment of nuclear power after the two major nuclear accidents. Consequently, light water reactor and other energy types took over the market. There were still some small-scale researches being carried out in Japan and other countries, though a full power reactor was never built.

However, over the years, the development of light water reactor reaches the point where major improvement is unlikely, when nuclear power needs to be more economically competitive
while meeting the safety regulation. Thus, numerous existing nuclear reactor concepts were revisited and examined by the Generation-IV International Forum (GIF) initiated by the US Department of Energy in 2000. As a result, molten salt reactor was selected as one of the six Generation-IV reactor concepts, and the interest has grown since. The research interest in the Europe has been focused on using molten salt reactor for burning the transuranic elements in spent fuel and molten salt breeder reactor. In their study, it is pointed out that a fast neutron reactor without graphite moderation possess a higher breeding ratio and could reduce the requirement for fuel processing rate. They proposed the MOSART design as a burner of spent fuel and the MSFR design for a fast spectrum breeder reactor. In US, the solid fueled Fluoride salt-cooled High-temperature Reactor has been studied by many universities and national labs. Generally, this type of reactor design utilizes solid fuel, and the fluoride salt is only used as a heat transfer liquid. The fluoride salt allows for high temperature operation with low pressure system, which is very favorable for reactor safety. The KR-FHR design by Kairos Power is in the early stage for licensing with NRC. The liquid fueled MSR also received many research interests in the recent years, especially on the topic of fuel processing. In China, the design of FHR and liquid fueled MSR has been evaluated by the Chinese Academy of Science. Forced convection test loops and natural circulation loop were built for thermal hydraulic study. Recently, a 2MWt liquid fueled experimental reactor, TMSR-LF1, has been approved for construction by the regulation committee. For a detailed review of the recent development of MSR, one could refer to the reviews by Serp et al. [8] and Locatelli et al. [9].

As pointed out by Rosenthal et. al. [10], while the molten salt reactors offer many attractive features, there are still many aspects to be studied before a full-scale reactor could be built. The remaining challenges for material and chemistry are better understanding of the behavior of fuel
salt mixture, the technique for large-scale fuel processing, better structure material that could handle the radiation and salt corrosion satisfactorily at the same time, along with graphite with low permeability and longer service life. The engineering development of large-scale components is also important. Though the MSRE has demonstrated the workability of many small components for the MSR, some of those may not be easily adopted in a commercial scale reactor. The steam generator compatible with molten salt, the control rod system for insertion into the salt, remote maintenance system and the xenon removal system are some examples.

In this document, the efforts towards the design analysis of a xenon removal system is presented. In chapter 1, the features, development and open questions for molten salt reactors are briefly discussed. The limited experience with xenon removal system for commercial sized molten salt reactor is highlighted. In chapter 2, a literature review on gaseous fission product removal and multiphase mass transfer is carried out. The motivation of xenon removal is briefly discussed, and the difficulty of designing a xenon removal system is reviewed, which leads to the usage of CFD simulation. In Chapter 3, the basics of Eulerian two-fluid model and species mass transfer are discussed in detail. Various constitutive relations are introduced, which are later tested in Chapter 4. A series of sensitivity studies and validation experiments are presented in Chapter 4. The difference between different models is shown in the sensitivity studies. The void fraction, velocity field and mass transfer data from the validation experiment serve as a guidance for model selection and a benchmark of the final CFD model. In Chapter 5, the future steps of this work are briefly discussed.
CHAPTER 2: HISTORY OF FISSION PRODUCT REMOVAL

As a reactor type that is still being actively studied, many aspects of the molten salt reactor are still under investigation. The idea of fission product removal is among one of these aspects. In this chapter, the development of fission product removal, along with the associated modeling efforts are reviewed.

2.1. Experience with Gaseous Fission Product Removal

The liquid-fueled MSR, unlike most reactor designs, uses a liquid fuel form, where fissile materials (typically $^{235}U$, or $^{233}U-^{232}Th$ for breeding reactors) [1, 2] are dissolved in the molten salt. This feature brings several benefits over the solid-fueled design. Continuous removal of fission products and refueling, for example, are two of them [10]. Among all the nuclides produced during fission, $^{135}Xe$ is of special importance because it is the major neutron poison produced in reactor operation, with a thermal neutron absorption cross section of $2.6 \times 10^6$ barns. It is mostly produced by decay of $^{135}I$ as [11]

$$^{135}I \rightarrow ^{135}Xe + \beta^-.$$  \hspace{1cm} (2.1)

During normal operation of the nuclear reactor, the concentration of xenon is at equilibrium. Shortly after reactor shutdown, the concentration of xenon will increase significantly by $^{135}I$ decay for a short period, introducing extra negative reactivity. This phenomenon is known as iodine pit. A similar jump of xenon concentration also appears when the reactor is subjected to power change, which can dramatically slow down this process. By removing xenon continuously from the fuel salt, the power level of the reactor could vary more flexibly. This feature makes it possible for MSR to operate in a load-following manner. With load-following capacity, MSR can be more competitive in the economic aspect. Another reason for constantly removing xenon is its
influence on the fuel cycle. The molten salt reactor was initially designed as a breeder reactor. With its liquid fuel form, the fission material could be processed and extracted during the operation. In the two-fluid two-region MSBR design, the fission material extracted from the blanket salt could be directly added into the fuel salt. However, the breeding ratio for a thermal neutron MSR is relatively low [12]. To make the reactor an effective breeder, the fission products must be removed simultaneously. Xenon, as a thermal neutron poison, is the most influencing element.

As discussed in Chapter 1, the idea of MSR originates from the ANP project at ORNL. During the operation of Aircraft Reactor Experiment, a steady state xenon poisoning experiment was carried out. The reactor was maintained at constant power for 25 hours, and the xenon poisoning was measured by the change of control rod value to maintain the reactor in steady state. The measured xenon poisoning is only 5% of the theoretical value if all xenon is contained in the fuel salt [5]. This deviation was attributed to the swirling of the fuel at the fuel pump, where the fission-product gases could have escaped to the cover gas. This argument was reinforced by the fission-product gases detected in a leak of the gas systems. In ORNL’s subsequent proposal for Aircraft Reactor Test [13], this feature was utilized, and a xenon removal system by helium scrubbing at the mixing chamber was considered.

As a transition from the military based project to a civilian power reactor, the MSBR design and the MSRE project were proposed as the interest in a nuclear-powered aircraft diminished. Many design considerations in the MSRE were inherited from the ANP project, including the xenon removal system at the fuel pump [14]. During the operation of MSRE, a series of experiments and studies were carried out on xenon poisoning. It is concluded that most of the xenon poisoning result from neutron absorption by $^{135}$Xe within the core graphite [15]. One
solution is to use graphite with lower xenon diffusivity and void fraction. Additionally, this xenon poisoning could be reduced by processing of the fuel salt. With careful analysis of the system, an empirical model for steady state xenon poison calculation was established [16]. In this model, xenon poisoning is estimated based on balance between source and sink terms. The source terms are identified as decay of $^{135}$I and small portion of direct generation from fission, which are functions of reactor power level. The sink terms, on the other hand, are more complicated. Five primary sink terms are considered, including burnup, decay, migration to core graphite, transfer to helium bubble and removal via the xenon stripper. The xenon stripper, or the xenon removal system, refers to the special design of MSRE’s fuel pump, where a portion of the discharged fuel salt is sucked from the pump and sprayed into the cover gas in the fuel pump bowl through a spray ring. Large interfacial area is formed for xenon stripping in this process, whose performance is approximated by a stripping efficiency coefficient, defined as the percentage of xenon transferred to the cover gas in the diverted fuel salt. Another important sink term is the transfer to helium bubbles. Helium bubbles are formed at the fuel pump bowl by helium bubbler and salt spray. Some of the bubbles are further carried into the loop where xenon could escape from the salt to the bubbles. These circulating bubbles could escape to the cover gas as passing through the fuel pump again, and the xenon absorbed in these bubbles could level the system. The helium bubble renewal efficiency, which is defined as the percentage of circulating bubbles that are renewed in the fuel pump, is used to describe this process. The model was first evaluated using a gas containing $^{85}$Kr tracer in a preliminary experiment. Some of the model constants were determined in these tests. Good agreement was found between the model and experiment. By applying the model to xenon poisoning, it is concluded that the transfer to helium bubbles term has a huge impact on the final derived xenon poisoning. With 1% of helium void fraction, xenon poisoning would decrease by
around 80%, if the stripping efficiency and helium bubble renewal efficiency are both as assumed to be 10%. During their derivation, the bubble size was assumed to be 0.254 mm in diameter and the mass transfer coefficient was taken as $1.7 \times 10^{-4} m/s$. However, certain disagreement was shown when compared with the actual $^{135}$Xe poisoning later in the MSRE operation, which could partly come from the uncertainty in the model parameters.

Though in the MSRE project, the xenon removal system is integrated in the fuel pump. For a large scale MSR at higher power, separate xenon removal device is required to achieve the same level of xenon removal. Based on the study mentioned before and several other researches [5, 17], physical removal of fission product by inertial gas sparging raised special interest because of its simplicity and compatibility with the fuel salt system [18]. In these reports, the lack of experimental xenon diffusivity and equilibrium ratio data were identified as a significant limitation, and further experiments were suggested. A prototypic gaseous fission product removal system was designed [7, 19] based on these studies, which includes a gas generator to inject helium bubbles and a gas separator to remove them. The separator adopted a rotational flow pattern to push the bubbles into the center of the pipe, and subsequently exit from the outlet located at the center. The liquid entrainment and pressure drop are the limiting factors for this design. In view of xenon removal and neutronics, the separation efficiency of the separator should be as large as possible. In order to design a gaseous fission product removal system, accurate prediction of mass transfer coefficient, void fraction and interfacial area concentration is necessary. However, these quantities were assumed to be uniform in the preliminary design and were calculated based on experiments with simplified geometry and flow conditions. Thus, considerable uncertainty could be expected for these results.
After the research of MSR was ceased in the US, few studies have been carried out on fission product removal, until recently. With the effort to develop more advanced and safer Gen-IV nuclear reactors, the MSR once again raises global research interest, and the possibility of fission product removal with inert gas sparging is being inspected again. Rubio et al. [20] carried out a scaled experiment to study tritium extraction by inert gas sparging and ultrasonic enhancement, where the Schmidt number of the scaling fluid is adjusted to the Schmidt number of FLiBe by changing its composition. In this experiment, oxygen was dissolved in the scaling fluid and was removed by inert gas sparging. The mass transfer coefficient is calculated based on the oxygen concentration change with time. The experiment is carried out in a co-current pipe flow. The bubbles are injected upstream to the ultrasonic affected region and the oxygen concentration is measured downstream. After the measurement point, the bubbles are separated in a vertical cylinder. The upper region of the cylinder is filled with gas and connected to the outlet. The mixture enters the cylinder in the middle and flows downwards. The bubbles are driven by buoyancy and exit the cylinder at the top. From the result, it is concluded that by applying ultrasonic wave onto the two-phase mixture, the mass transfer could be enhanced considerably, which is attributed to the increased interfacial area because of breakup. Kanai et al. [21] studied two-phase mass transfer in molten salt for bubble column application. The salt composition used in the experiment was a combination of Li₂CO₃, Na₂CO₃ and K₂CO₃. The concentration of CO₂ in the off gas was measured with an infrared detector and the concentration of CO₂ in the liquid was calculated based on mass balance and time integration. The sensitivity of temperature and gas injection rate on the mass transfer rate is reported. A decrease of mass transfer coefficient is found with increasing temperature, which is different from the observation in common air-water bubble columns. Chen et al. studied the effect of salt properties on the volumetric mass transfer coefficient.
with computational fluid dynamics [22]. Apart from the studies of mass transfer, the design of a pipeline gas separator is also studied. Contrary to the pipeline separator design, Funahashi et al. proposed a separator with pick-off rings and a vertical cylinder [23]. Zheng et al. carried out a series of experiments with an inclined tube [24]. Moreover, experiments and analysis were carried out regarding the general thermal hydraulics of molten salt. Bardet et al. performed an analysis for possible scaling fluid in MSR development [25]. Salt loops were built at University of Wisconsin to study the corrosion, heat transfer characteristics and natural circulation of molten salt [26, 27]. Forced convection loops with FLiNaK and nitrates, and a natural circulation loop with nitrate were constructed and tested at Shanghai Institute of Applied Physics as part of their TMSR program [28]. A natural circulation experiment with FLiNaK and heat pipes was conducted by Liu et al. [29] at Xi’an Jiaotong University.

2.2. Modeling of Mass Transfer in Multiphase Flow

In order to design a xenon removal device, information on local distribution of species concentration, relative velocity and void fraction are needed to determine the overall transport rate. Due to the challenge and cost of carrying out experiment with molten salt and xenon, designing and testing a gaseous fission product removal device completed based on experiment is continually becoming less practical. Therefore, simulation approach becomes a natural choice for verifying the feasibility of this technology. With the help of computational fluid dynamics (CFD), a design analysis of fission product removal system could be carried out in a cost-effective way. Typically, in dealing with large scale two-phase flow phenomenon, Eulerian-Eulerian two fluid model is utilized.

However, the modeling of two-phase flow mass transfer is far from mature. Despite extensive simulation and experimental research, a consensus on how to model the interfacial
forces, turbulence, bubble diameter and even interpretation of the governing equations is hardly reached. For a detailed discussion on this topic, one could refer to some existing literature reviews [30-32]. As for mass transfer, this issue becomes more subtle, since the rate of transfer not only relies on the mass transfer coefficient, but also relies on liquid velocity, void fraction distribution and interfacial area concentration. Without proper modeling of the quantities mentioned before, validation of mass transfer models is hardly convincing. Nevertheless, various mass transfer models have been used for CFD simulation in literature. In an early attempt to simulate mass transfer in bubble column, Krishna et al. [33] adopted a constant mass transfer coefficient of $4 \times 10^{-4} m^2/s$ and reported good agreement with experimental data. Wang et al. [34] examined mass transfer coefficient based on penetration theory and surface renewal model. Wiemann et al. [35] chose an empirical correlation of Sherwood number to calculate the coefficient. In general, the mass transfer models used in CFD simulation are mostly from the studies for single bubble mass transfer. The major improvement compared with the 1D or 0D mass balance analysis with theoretical or experimental correlation is that the local distribution of velocity, void fraction and turbulent quantities are considered. The transient behavior of the system could be properly captured. With physical constitutive models, the CFD simulation could be applied to more complicated flow conditions and geometries. Additional description of mass transfer modeling is included in work of Kulkarni [36] and Rzehak et al. [37].

In the field of nuclear engineering, ORNL carried out some studies on mass transfer in pipe flow. Preliminary experiments and analysis were carried out to study this two-phase mass transfer mechanism after the MSRE experiment [18, 38], the focus of which was to obtain an experimental correlation for mass transfer in bubbly pipe flow that can be further used in the design of a xenon removal system. Peebles [15] carried out a theoretical study for the mass transfer between liquid
and spherical bubble. The author first analyzed the simple case of a spherical bubble moving in a stagnant liquid, considering mobile and rigid bubble interface. Assumptions are made for the velocity field in order to arrive at a solution. However, these assumptions would not be correct in a turbulent liquid. Moreover, the relative velocity between phases used in the derivation is only applicable to laminar flow. In the large-scale xenon removal system, it is expected that the flow would be turbulent. In order to make an estimation, the author related the relative velocity between phases in turbulent cases to the fluctuation of liquid velocity. This assumption is questionable since the derived relative velocity is not related to the orientation of the flow. The derived turbulent relative velocity is plugged back into the laminar flow solution, which is expected to underestimate the mass transfer since the vortex near the bubble interface is not considered. After a review of the literature, the author stated that the knowledge at that time was not enough to arrive at a firm analysis of the mass transfer process, and further experiments were suggested. Subsequently, a more thorough study was carried out by Kress [38]. In this study, a comprehensive review of the single bubble mass transfer analysis and pipe flow experiment is first conducted. The scaling of pipe Sherwood number to Schmidt number is reported in the range of $1/3$ to $1/2$. The power dependence on Reynold number, on the other hand, is reported to be 0.9 to 1.1 for conduits, and 0.6 to 0.8 for stirred vessels. Dissolved oxygen experiments were carried out to measure the mass transfer coefficient and to separate the coefficient from the interfacial area concentration. The measurement of interfacial area was obtained from images taken at the region of interest. By assuming a fixed bubble distribution and estimating the gas void fraction from experimental correlation, the interfacial area is derived from the bubble number density. This is simply done by counting the number of bubbles in the images, which is much easier than measuring the size
directly from the image. From the experimental result, a correlation is proposed for the Sherwood number as

\[ \text{Sh}_L = 0.34 \cdot \text{Re}_L^{0.94} \cdot \text{Sc}_L^{0.5} \left( \frac{d}{L_c} \right), \]  

where the characteristic length \( L_c \) used here is the hydraulic diameter of the pipe. Additionally, with the usage of the surface renewal model, a similar relation is obtained as

\[ \text{Sh}_L \sim \text{Sc}_L^{0.5} \cdot \text{Re}_L^{0.92} \left( \frac{d}{L_c} \right)^{\frac{1}{3}}. \]  

In arriving at the experimental and theoretical results, many assumptions were made along with the usage of experimental correlation. The local distribution of flow variables is also ignored, which could be a source of uncertainty in the results.

Based on the discussion above, in order to gain confidence in the simulation, model validation with experiments is preferable and necessary. The validation should not only include the mass transfer rate, but also other flow quantities. Regarding this issue, experiments on bubble columns are helpful. Bubble columns refer to the kind of apparatus where gas is sparged into a liquid-filled vessel in form of bubbles. Various multiphase phenomenon could occur during the operation of a bubble column, including bubble deformation, bubble induced turbulence, bubble coalescence, bubble breakup, and mass transfer across the interface. The large interfacial area between bubbles and the liquid phase makes bubble column a great multiphase reactor and is therefore widely used in chemical, metallurgical and pharmaceutical industries [39]. With its simplicity, different shapes of bubble columns are also widely used in research on two-phase mass transfer and general two-phase flow dynamics. Early experiments of bubble columns generally targeted at global characteristic like total gas hold up and volumetric mass transfer coefficient [40, 41]. With the development of flow measurement technique, localized data of the flow field and
phase distribution became available. Computer automated radioactive particle tracking (CARPT) was used by Devanathan et al. [42] and Degaleesan et al. [43] to obtain liquid phase velocity distribution. In the meantime, long existing methods of X-ray tomography and conductivity probe was adopted by Kumar et al. [44] and Buwa et al. [45] to measure void fraction distribution. With the help of particle image velocimetry, simultaneous measurement of liquid and gas phase velocity was carried out by Hassan et al. [46]. Later improvement of the laser technique and high speed camera allows for high time resolution measurement [47]. More sophisticated measurement arrangement with two cameras greatly improves the accuracy of phase discrimination, and more accurate measurement of the flow field could be obtained [48]. Another widely used method is flow visualization and image processing, sometimes coupled with PIV measurement. Two-phase flow visualization could supply information on phase distribution, bubble size distribution and information on gas phase velocity [37, 45, 49, 50]. As for mass transfer measurement, by virtue of the development of dissolved gas sensors, local concentrations can be measured with fair accuracy and time resolution, such as widely done for dissolved oxygen [51-53].
CHAPTER 3: PHYSICAL MODEL DESCRIPTION

In this chapter, the modeling of fission product removal with the Eulerian two-fluid model is discussed in detail. The derivation of the governing equation, the constitutive relation of phase interaction and the material properties of FLiNaK are included.

3.1. Modeling of Two-Phase Flow and Mass Transfer

The difficulty with two-phase flow modeling lies in the existence of one or several interfaces separating the immiscible fluid. These deformable interfaces cause strong coupling between boundary conditions and flow fields, along with sharp discontinuity of flow variables. The description of interface itself also brings challenges to computational fluid dynamics. The methods developed for tracking the interface can be divided into three categories [54]: Volume of Fluid (VOF) method, front tracking method, and level set method.

The Volume of Fluid method (VOF) method is popular as it can treat bubble coalescence with ease and be readily implemented in 3D simulation. The crucial part of this method is to derive the transport equation near the interface, which is not the ordinary conservation equation because of its discontinuity. Special methods, for example the donor-acceptor method, have been developed for calculation near the interface [55].

The front tracking method explicitly records the location of the interface. In 2D case, this could be done with x and y coordinate. However, it experiences significant difficulty for complex multi-phase flow and 3D case. Thus, the application of front tracking method is rather limited [56].

The level set method assumes the existence of a phase function, $\phi$, where $\phi(\text{liquid}) < 0$ and $\phi(\text{gas}) > 0$. In this sense, at the location of the interface, $\phi = 0$ and the meaning of this function can be interpreted as the distance to the interface. The transportation of this function is similar to ordinary conservation equation. However, special modifications need to be done to
maintain the distance interpretation of this function. It can be used in 3D simulation, but experiences numerical challenges when interfaces are close together [57].

Though the methods of explicitly tracking the interface are constantly being improved and could already be used in some complex flows, they are still extremely computationally expensive and suffer from numerical problems. The applications of these methods are largely reserved for low bubble numbers in small computational domains, which is not well suited for simulation of a reactor or reactor components. An alternative method of time averaging of the flow field is proposed and developed by several researchers and is now known as the Eulerian-Eulerian two-fluid model [58].

3.1.1. Eulerian-Eulerian Model

In this section, the basics of Eulerian two-fluid model are introduced. The void fraction of each Eulerian phase is first derived, followed with the governing equations based on void fraction. Phase in the Eulerian-Eulerian framework could be a single material, a mixture of species, or a collection of similar particles. Each phase possesses its own velocity, void fraction and properties. This model is widely used in different fields of engineering because of its flexibility and simplicity compared with other explicit models.

3.1.1.1. Volume Fraction and Averaging

Physically, interface between phases results in a boundary condition between the two continuum materials. However, the location of this boundary and the jump condition are also related to the phases it separates. The strong coupling between boundary condition and flow variable causes significant difficulty for analyzing. Naturally, implicit treatment of the interfacial condition is preferable if we want to study complicated flow conditions. This is achieved by introduction of void fraction. The phase density function $\xi_k$ is defined as
\[ \xi_k(\vec{r}, t) = \begin{cases} 1, & \text{if phase } k \text{ is present at } \vec{r} \text{ at time } t \\ 0, & \text{if otherwise} \end{cases}. \]  

(3.1)

The void fraction can be then defined as [58]

\[ \alpha_k = \frac{\int_{\Delta t} \xi_k \, dt}{\Delta t}, \]  

(3.2)

where \( \Delta t \) is chosen so that the discontinuity introduced by the interface is smoothed but still retaining the time domain characteristics. From the definition of \( \xi_k \), the following identities are obtained as

\[ \sum \xi_k = 1, k = 1,2, \ldots \]

\[ \sum \alpha_k = 1, k = 1,2, \ldots \]  

(3.3)

This averaging method is similar to that used in defining continuum materials. The void fraction defined by the time average of the phase density function represents the probability of finding phase \( k \) at a given point, which is used in deriving time averaged field equations.

For clarity, the notion of averaging introduced by Ishii et al. [58] is adopted here. Considering a physical quantity \( F \) that depends on space and time, a series of notions is defined as

\[ F_k = \xi_k F \]

\[ \overline{F_k} = \frac{\int_{\Delta t} F \xi_k \, dt}{\Delta t} \]

\[ \overline{\overline{F_k}} = \frac{\overline{F_k}}{\alpha_k}, \]  

(3.4)

where \( \overline{F_k} \) represents a time averaged quantity, and \( \overline{\overline{F_k}} \) is often called the phase averaged quantity. Furthermore, most quantities are weighted by mass in fluid mechanics. It is natural to write

\[ F = \rho \phi \]

\[ \overline{F_k} = \overline{\rho_k \phi_k}. \]  

(3.5)
Though for simplicity, the phase density $\rho_k$ is written as $\rho_k$ in the following section, since it is the density of the associated phase without averaging. With these definitions, the time averaged field equation for a phase weighted flow variable $\phi_k$ can be derived as

$$\frac{\partial \alpha_k \rho_k \phi_k}{\partial t} + \nabla \cdot \left( \alpha_k \rho_k \phi_k \bar{u_k} \right) = -\nabla \cdot \left( \alpha_k (\bar{f}_k + f^T_k) \right) + \bar{l}_{k,\phi},$$

(3.6)

where $\bar{f}_k$ is the flux for $\phi_k$, $\bar{l}_{k,\phi}$ is the source term and $f^T_k$ is the turbulence. For a more rigorous derivation, the source term could be distinguished as transfer between phases and pure source from the environment. The transfer between phases term could be related to the local velocity difference between the fluid and the interface [58]. However, in CFD modeling, the details near the interface could not be fully resolved even for explicit interface tracking method, and sub-grid modeling is usually needed [59].

Only time average of the phase density function is discussed here. A volume average is also possible, which leads to the Volume of Fluid (VOF) model [55]. In the following section, a brief introduction to the Eulerian-Eulerian two fluid model derived based on this time averaging method is presented.

### 3.1.1.2. Continuity Equation

Applying the averaging method introduced in the previous section by replacing $\phi$ with a constant 1, the continuity equation in Eulerian-Eulerian two-fluid model could be derived as

$$\frac{\partial \alpha_l \rho_l}{\partial t} + \nabla \cdot \left( \alpha_l \rho_l \bar{u_l} \right) = \Gamma_l$$

$$\frac{\partial \alpha_g \rho_g}{\partial t} + \nabla \cdot \left( \alpha_g \rho_g \bar{u_g} \right) = \Gamma_g$$

$$\Gamma_g + \Gamma_l = 0.$$  

(3.7)
The source term $I'$ here is related to the mass transfer at the interface, which could either be related to phase change or chemical equilibrium. The third equation expresses the conservation of mass at the interface. It is assumed that the interface cannot store mass.

3.1.1.3. Momentum Equation

Set the function $\phi$ to be the mass-weighted mean velocity, $\overline{u_k}$, the momentum conservation equation is derived as

$$
\frac{\partial \alpha_k \rho_k \overline{u_k}}{\partial t} + \nabla \cdot (\alpha_k \rho_k \overline{u_k} \overline{u_k}) = -\nabla (\alpha_k P_k) + \nabla \cdot \left( \alpha_k \left( \overline{\sigma_{k}} + \overline{\sigma_{k}^T} \right) \right) + \alpha_k \rho_k \mathbf{g} + \overline{M_k}
$$

$$
\overline{M_g} + \overline{M_I} = \overline{M_m}.
$$

(3.8)

The term $\overline{\sigma_{k}}$ and $\overline{\sigma_{k}^T}$ are the viscous and turbulent stress tensors, as a result of time averaging. The time averaged nature of the Eulerian model requires the turbulence of both phases to be modeled in the same way as RANS equation in the case of single-phase flow. The second equation describes the relationship between the interfacial source terms. Here, the summation of the source terms does not equal to zero, indicating that the interface can store momentum. The term $\overline{M_m}$ represents the contribution from surface tension. When external forces such as magnetic force are present, extra components should be added to the source terms.

In the present study, the phases are assumed to be isothermal and the energy equation is not included in the calculation. This is essentially the case for the xenon removal process, since the injected gas should be preheated and the temperature difference between the gas and salt is expected to be small. This is because the process is quite sensitive to temperature near the melting point, and plug could happen if the salt solidifies.
3.1.1.4. Turbulence Model

In the momentum equation, a turbulent stress tensor $\mathbf{\tilde{G}}_k^T$ is present. Similar to the single-phase RANS equation, this term is expressed as

$$
\mathbf{\tilde{G}}_k^T = -\rho_k \overline{u_k^i u_k^i}
$$

$$
\overline{u_k} = \overline{u_k} + u_k'.
$$

(3.9)

However, a major difference exists compared with the single-phase Reynold stress tensor. The time average of the unsteady term alone is not zero. It only becomes zero with phase weighted averaging. This leads to an additional bubble induced turbulence term along with the single-phase shear induced term. Though extensive research has been devoted to this topic [60-62], the turbulence formulation for Eulerian-Eulerian two-fluid model is still under development [63]. One reason behind this slow progress is the difficulty of model validation for the turbulent effect, where the presence of bubbles causes significant challenges for flow measurement and data interpretation. The model used in this study is proposed by Sato et al. [64], which could be viewed as a zero-equation model. This model first assumes that the Reynold stress could be decomposed into the shear induced term and the bubble induced term as

$$
\mathbf{\tilde{G}}_k^T = (\mathbf{\tilde{G}}_k^T)_{SI} + (\mathbf{\tilde{G}}_k^T)_{BI}.
$$

(3.10)

Then, as in the single-phase turbulence, Boussinesq closure hypothesis is used again for both terms, and thus Reynolds stress is again related to the derivatives of the phase weighted mean values and eddy viscosity. The eddy viscosity, or turbulent viscosity, is assumed to be a superposition of two individual terms. Sato et al. proposed a way to relate the bubble induced eddy viscosity with the relative motion between the dispersed phase and the continuous phase. Also, the $k - \epsilon$ formulation of turbulence is used in their derivation. This relation is based on the scaling of
turbulence mixing length and time scale related to bubble motion. The resulting constitutive relationship for the continuous phase is

\[
\mathbf{e}_k^T = -\frac{2}{3}(\rho_k k_k + \rho_k v_k \nabla \cdot \mathbf{u}_k)I + \rho_k v_k^T (\mathbf{r} \mathbf{u}_k + (\mathbf{r} \mathbf{u}_k)^T)
\]

\[
v_k^T = (v_k^T)_{SI} + (v_k^T)_{BI}
\]

\[
(v_k^T)_{BI} = C_v (1 - \alpha_k) d|\mathbf{u}_r|, \quad C_v = 0.6. \quad (3.11)
\]

In this formulation, no modification is made to the original \( k - \epsilon \) model except for the universal phase average. As a result, each term in the equation is multiplied with the continuous-phase void fraction [65]. In our modeling and validation setup, the RNG \( k - \epsilon \) model and realizable \( k - \epsilon \) model are investigated. As expected, the difference between these two models only slightly changes the simulation result, since the turbulence in this case is dominated by bubble induced term rather than the shear induced term. For the dispersed phase, the turbulence response model developed by Tchen et al. is adopted [66]. However, just like the model for shear induced turbulence, the model for dispersed phase turbulence only have a small effect on the result because of the large density ratio between phases. It should be pointed out that the argument made here is only applicable to bubble-liquid flow, which no longer holds true when heavy particles are included. For details on the topic of multiphase turbulence, one could refer to the review by Rzhuhak et al. [65] and Ziegenhein et al. [67].

3.1.2. Multiphase Species Transport Model

The Eulerian-Eulerian two-fluid model described in the previous section alone is not enough for simulating the xenon removal process, since the concentration of species is not addressed. The solution is to add the species mass fraction into the model.
3.1.2.1. Multiphase Species Transport Equation

The transport equation for the mass fraction of species \( i \) in phase \( k \) is

\[
\frac{\partial \alpha_k \rho_k Y_{k,i}}{\partial t} + \nabla \cdot (\alpha_k \rho_k Y_{k,i} u_k) = \nabla \cdot (\alpha_k \rho_k \left( D_k^t + D_{k,i} \right) \nabla Y_{k,i}) + \Gamma_{k,i}, \tag{3.12}
\]

where \( Y_{k,i} \) is the mass fraction of species in phase \( k \), \( D_{k,t} \) is turbulent diffusivity, and \( D_{k,i} \) is the molecular diffusivity. \( \Gamma_{k,i} \) is the mass of species \( i \) transferred to phase \( k \). This source term also appears in the continuity equation for phase \( k \). The turbulent diffusivity is included to account for the additional diffusion effect introduced by the turbulence, which is modeled as

\[
D_k^t = \frac{\mu_t}{\rho_k S_{c_t}}. \tag{3.13}
\]

The turbulent Schmidt number, \( S_{c_t} \), in current study is set at 0.7 based on literature [68]. Normally, the turbulent diffusion overwhelms the molecular diffusion, and thus a precise value for molecular diffusivity is less important when a strong turbulent effect is present for the diffusion within the phase. However, the mass transfer between phases is still controlled by molecular diffusion, and a precise value of diffusivity is still needed. Usually, the density of each phase will change based on the composition of species with a nonlinear form. Fortunately, considering the low solubility of inert gas in molten salt, this choice of density modeling does not have much influence on the result. Thus, the density of the mixture material is modeled to be a constant, same as the liquid density.
Figure 3.1: Illustration of Two-Resistance Model

Figure 3.2: Illustration of One-Resistance Model
3.1.2.2. Henry’s Law

As discussed before, interfaces between phases introduce discontinuity in flow variables. After time averaging, the discontinuity disappears along with the information near the boundary. Thus, a constitutive model for the mass transfer source term is needed to describe the mass transfer between phases. Modeling of this source term requires information of species at phase boundaries, which is described by Henry’s law [37]. The general form of Henry’s Law is

\[ P_g^i = H X^e_{li}, \]  

where \( X^e_{li} \) is the molar concentration of species \( i \) in liquid phase at equilibrium, with unit of \( mol/m^3 \). \( H \) is the Henry’s coefficient, and \( P_g^i \) is the partial pressure for species \( i \) in gas phase. This form of Henry’s law can be rewritten as

\[ \rho_l Y_{li} = K^p_{li}(T) Y_{gi} \rho_g, \]  

where \( K^p_{li} \) is the mass concentration equilibrium ratio. This means that at the interface the concentration ratio of species \( i \) in different phases is a function of temperature, \( T \). Experiments of inert gas equilibrium ratio in molten salt were carried out by Blander et al. [69] and their results will be used in our study until more data become available from the xenon removal experiment.

3.1.2.3. Source Term of Mass Transfer

Based on Henry’s law, the source term for the interfacial species transport can be modelled. The process for mass transfer at the interface is divided into three steps. First, the molecule or particles approach the interface from primary phase, by means of diffusion. The second step is the transfer of molecules across the interface. The last step is the transfer from the interface to the secondary phase by diffusion. Assuming the interface is sufficiently thin and cannot hold mass, the second step can be ignored, and only the first step and third step cause resistance to species
transfer. The resulting concentration profile is illustrated qualitatively in Figure 3.1. From the concentration profile, the species mass transferred between phases can be calculated from Fick’s law as

\[
\Gamma = k_\text{i} a_\text{i} \left( K^\rho_{\text{i},\text{g}} Y_{\text{g},\text{i}} - \rho_{\text{l}} Y_{\text{l},\text{i}} \right)
\]

\[
= k_\text{i} a_\text{i} \rho_{\text{l}} (Y_{\text{l},\text{i}} - Y_{\text{i},\text{interface}})
\]

\[
= k_\text{g} a_\text{i} \rho_{\text{g}} (Y_{\text{g},\text{i},\text{interface}} - Y_{\text{g},\text{i}}).
\]

(3.16)

Considering the jump condition by Henry’s Law, these equations could be simplified as [37]

\[
\frac{1}{k} = \frac{1}{k_\text{i}} + \frac{K^\rho_{\text{g}}}{k_\text{g}} \approx \frac{1}{k_\text{i}}
\]

\[
\Gamma_{\text{i},\text{g}} = k_\text{i} a_\text{i} \left( K^\rho_{\text{i},\text{g}} Y_{\text{g},\text{i}} - \rho_{\text{l}} Y_{\text{l},\text{i}} \right).
\]

(3.17)

The species concentration profile shown in Figure 3.1 might be slightly counterintuitive, considering that the transport could happen from low concentration to high concentration under certain circumstances, which appears to be a violation of Fick’s law. The reason behind this is, at the interface, the transport of species is no longer controlled by diffusion or species concentration, and what is balanced at the interface is the Gibbs free energy for the considered species. This complicated physics of interface is simplified by Henry’s law with a zero-thickness interface model.

Normally, the diffusion in gas phase is much stronger than that in liquid phase, and the mass transfer coefficient could be approximated by the liquid phase coefficient only. This approximated version is often referred as one-resistance model and is widely used in bubble-liquid flow, as depicted in Figure 3.2.
3.1.2.4. Species Transport at Free Surface

The time averaging method used in the previous section could smooth out the discontinuity introduced by the interface for dispersed flow. However, this method is not applicable for a free surface. For this type of situation, the VOF model is often adopted to explicitly track the interface. The two-fluid model, on the other hand, could also be modified to track this large interface based on certain regime transition criteria, though the physical meanings of void fraction (time averaging) and volume fraction (volume averaging) is mixed with this method. Nevertheless, for both methods, the convenience brought by time averaging disappears. To model the mass transfer in this case, we need to address the discontinuity of species from the Henry’s law,

$$\rho_l Y^i_l = K^h_{l,i}(T) Y^i_g \rho_g.$$  \hspace{1cm} (3.15)

An analytical solution exists for this distribution if a zero-thickness stationary interface is prescribed. However, in CFD, a finite thickness interface is present. If this discontinuity is included into the simulation, the result will depend on how we prescribe the interface and the grid size near this interface. The conservation of mass and species is hardly guaranteed. To overcome this difficulty, a different approach is adopted [70, 71], which is often called as Continuous-Species-Transfer (CST) method. A combined field variable $C_{m,i}$ for species in both phases could be defined based on volume mixing as

$$C_{k,i} = \rho_l Y_{k,i}$$

$$C_{m,i} = \alpha_g C_{g,i} + \alpha_l C_{l,i}.$$  \hspace{1cm} (3.18)

$C_{m,i}$ is not a phase level variable, rather, it is defined at the mixture level, and the discontinuity caused by the chemical equilibrium is considered in the source term of the transport equation for $C_{m,i}$. To derive the source term $S$, Henry’s law and Fick’s law are used and a mixture diffusivity $D_{m,i}$ is defined as
\[ K^\rho C_{g,i} = C_{l,i} \]
\[ D_{g,i} \nabla C_{g,i} = D_{l,i} \nabla C_{l,i} \]
\[ D_{m,i} = \alpha_g D_{g,i} + \alpha_l D_{l,i} \]  
(3.19)

Assume that the flux of \( C_{m,i} \) is expressed in the phases weighted form and share the same mixture diffusivity in the interface region, then the flux and source term is [70]

\[
J_{m,i} = \alpha_g J_{g,i} + \alpha_l J_{l,i}
\]
\[
= - (\alpha_g D_{m,i} \nabla C_{g,i} + \alpha_l D_{m,i} \nabla C_{l,i})
\]
\[
= - D_{m,i} \nabla (\alpha_g C_{g,i} + \alpha_l C_{l,i}) + D_{m,i} (C_{g,i} - C_{l,i}) \nabla \alpha_g
\]
\[
C_{g,i} - C_{l,i} = (1 - K_{l,i}^\rho) C_{g,i} = \frac{C_{m,i}(1 - K_{l,i}^\rho)}{\alpha_g + K_{l,i}^\rho(1 - \alpha_g)}
\]

\[
\nabla \cdot J_{m,i} = \nabla \cdot \left( D_{m,i} \nabla C_{m,i} - D_{m,i} \left( \frac{C_{m,i}(1 - K_{l,i}^\rho)}{\alpha_g + K_{l,i}^\rho(1 - \alpha_g)} \right) \nabla \alpha_g \right). \tag{3.20}
\]

Thus, the final transport equation for \( C_{m,i} \) becomes

\[
\frac{\partial C_{m,i}}{\partial t} + \nabla \cdot (C_{m,i} \mathbf{u}) = \nabla \cdot (D_{m,i} \nabla C_{m,i}) + S
\]
\[
S \equiv - \nabla \cdot \left( \frac{D_{m,i} C_{m,i}(1 - K_{l,i}^\rho)}{\alpha_g + K_{l,i}^\rho(1 - \alpha_g)} \nabla \alpha_g \right). \tag{3.21}
\]

3.2. Modeling of Constitutive Law

With time averaging, the discontinuity introduced by the interface is smoothed, resulting in an additional phase interaction term in the transport equation. Since the interface is now treated implicitly, the new phase interaction term could not be resolved solely by simulation. Constitutive law from experiment or analysis is required to complete the modeling. In this section, the modeling of these constitutive law is introduced.
3.2.1. Phase Interaction

In the momentum equation, an extra phase interaction term $\bar{M}_k$ appears after performing the time averaging. This term is related to the momentum transfer between different phases. Based on theoretical and experimental studies, various phase interaction terms have been identified and modelled. Drag force, lift force, virtual mass force, turbulent dispersion force, wall lubrication force and Basset force are considered in various situations. These interaction terms are assumed to be addible as

$$M_k = M_{\text{drag}} + M_{\text{turb}} + M_{\text{lift}} + M_{\text{vm}}, \quad (3.22)$$

where, in the current study, the wall lubrication and Basset force are not considered based on analysis of the flow condition and geometry.

3.2.1.1. Drag Force

Drag is the force exerted on the dispersed object against relative motion. It includes the pressure and viscous force around the object. The drag for a deformable interface is much more complicated than that for a hard sphere. Based on sensitivity study of interfacial closure laws [72], it is concluded that the drag force is sufficient to capture the global flow patterns of buoyancy driven bubbly flow, and other forces only have a secondary effect to the flow field. However, it is also noted that the other forces are necessary to obtain a physical void fraction distribution. Generally, the drag force is expressed as

$$M_{\text{drag},d} = -\frac{3C_D}{4}\rho_c a_i |\mathbf{u}_r| (\mathbf{u}_r), \quad \mathbf{u}_r = \mathbf{u}_d - \mathbf{u}_c, \quad (3.23)$$

where $a_i$ is the interfacial area concentration for interaction between phases, the subscript $c$ represents the properties of continuous phase and $d$ represents the properties of the dispersed phase. The modeling of drag force then reduces to modeling of the drag coefficient $C_D$. 
The drag force on a hard sphere in Stokes flow is well known by engineers, since an analytical solution exists for this special case. The drag coefficient in this case is given by

\[
C_{D,\infty} = \frac{24}{Re}, \quad Re = \frac{\rho d |u_r|}{\mu},
\]

where \(d\) is the diameter of the bubble or droplet. From this expression, the drag coefficient only depends on Reynold number. This is generally true for a hard sphere. However, for fluid particles such as bubbles and drops, the particle shape could be distorted, and the drag coefficient is no longer solely dependent on Reynold number. For these kind of particles, the drag coefficient could be calculated from the mean diameter as [73]

\[
C_{D,\infty} = \frac{2}{3} d \sqrt{\frac{g \Delta \rho}{\sigma}}, \quad \text{for } N_\mu \geq \frac{36 \sqrt{2}(1 + 0.1 Re^{0.75})}{Re^2}
\]

\[
N_\mu = \frac{\mu_c}{\left(\rho_c \sigma \frac{\sigma}{g \Delta \rho}\right)^{1/2}}.
\]

For a detailed discussion of particle morphology and drag coefficient, one could refer to the books by Clift et al. [74] and Ishii et al. [58].

The previous discussion is about the drag coefficient on a single particle. When multiple particles are present in the system, the interaction between them leads to a modified drag coefficient based on the characteristic of particles. For distorted particles, the drag coefficient increases with increasing void fraction. On the other hand, in churn turbulent regime and slug bubble regime, the drag coefficient would decrease with increasing void fraction. When high void fraction flow is expected, this modification is necessary for a reasonable prediction. For our modeling, three models by Schiller and Naumann [75], Tomiyama [76], Ishii and Zuber [58] are evaluated. The Schiller-Naumann model [75] is based on experimental result for a single hard
sphere. The Tomiyama model [76] considers the morphology of a fluid particle. The Ishii and Zuber model [58] further takes the modification in a multiparticle system into account. The details about these models are given in Table 3.1.

Table 3.1: Models of drag coefficient $C_D$

<table>
<thead>
<tr>
<th>Model Name</th>
<th>$C_D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Schiller and Naumann [75]</td>
<td>$\begin{cases} \frac{24(1 + 0.15Re^{0.687})}{Re}, &amp; Re \leq 1000 \ \frac{0.44}{Re}, &amp; Re &gt; 1000 \end{cases}$</td>
</tr>
<tr>
<td>Tomiyama [76]</td>
<td>$\max \left( \min \left( \frac{24}{Re} (1 + 0.15Re^{0.687}), \frac{72}{Re}, \frac{8}{3} \left( \frac{Eo}{Eo + 4} \right) \right), \right.$ $Eo = \frac{g\Delta \rho d^2}{\sigma}$</td>
</tr>
<tr>
<td>Ishii and Zuber [58]</td>
<td>$C_{D,\text{sphere}} = \frac{24}{Re_m} (1 + 0.15Re_m^{0.687})$, $C_{D,\text{cap}} = \frac{8}{3} (1 - \alpha)^2$</td>
</tr>
<tr>
<td></td>
<td>$C_{D,\text{ellipse}} = \frac{2}{3} E_a Eo^{1/2}, E_a = \left( \frac{1 + 17.67 f_\alpha^6}{18.67 f_\alpha} \right)^2, f_\alpha = \frac{\mu_c}{\mu_m} (1 - \alpha)^{1/2}$</td>
</tr>
<tr>
<td></td>
<td>$Re_m = \frac{\rho_s u_r d}{\mu_m}, \frac{\mu_m}{\mu_c} = (1 - \alpha)^\frac{2.5(\mu_d + 0.4\mu_c)}{\mu_c + \mu_d}$</td>
</tr>
</tbody>
</table>

3.2.1.2. **Lift**

Lift force is the additional lateral force exerted on a fluid particle in a viscous shear flow, which is not included in the steady state drag. Lift force and other lateral forces are important in predicting the phase distribution. In the literature, some constitutive relationships have been
proposed based on experiment and numerical simulation, though the applicable range is hard to evaluate.

From the experimental work by Tomiyama et al. [77], the lift force could be modelled in the following form,

\[ \mathbf{M}_{\text{lift,d}} = -C_L \rho l \alpha_d (\mathbf{u}_r) \times (\nabla \times \mathbf{u}_c). \] (3.26)

The lift coefficient is a function of bubble diameter, surface tension and density of the fluid. Various models for determining \( C_L \) have been proposed. The model given by Tomiyama et al. is

\[
C_L = \begin{cases} 
0.288 \tanh(0.121 \max(Re, 7.374)), & Eo_d < 4 \\
0.00105 Eo_d^3 - 0.0159 Eo_d^2 - 0.0204 Eo_d + 0.474, & 4 \leq Eo_d \leq 10 \\
-0.27, & 10 < Eo_d
\end{cases}
\] (3.27)

The Tomiyama lift model, along with the analytical result for potential flow [78] (\( C_L = 0.5 \)) and the model proposed by Hibiki and Ishii [79] are evaluated under certain flow condition in this document. The Hibiki and Ishii model [79] is based on the work by Legendre et al. [80], where a modification factor \( \xi \) is multiplied to the original correlation as

\[
C_L = \xi \sqrt{C_{L,\text{low}}^2 + C_{L,\text{high}}^2}, \quad \xi = 2 - e^{2.92 D_d^{2.21}}, \quad D_d^* = \frac{d}{4} \frac{\sigma}{g \Delta \rho}
\] (3.28)

\[
C_{L,\text{low}} = \frac{6}{\pi^2 \sqrt{2 Re G_s}} \left( \frac{2.255}{1 + \frac{0.1 Re}{G_s}} \right)^{1.5}, \quad C_{L,\text{high}} = 0.5 \left( 1 + \frac{16 \frac{Re}{29}}{1 + \frac{29}{Re}} \right), \quad G_s = \frac{d}{2 |\mathbf{u}_r| |\nabla \times \mathbf{u}_c|}.
\]

Since the modeling of lift force has not been well-developed, a sensitivity study might be preferable when being applied to a specific flow condition.

3.2.1.3. Turbulent Dispersion

The turbulent dispersion force describes the dispersion of fluid particles in a turbulent flow. It is likely from the fluctuation component of interfacial forces, especially drag, acting on the fluid
particles. Lopez de Bertodano [81] performed an ensemble average of the fluctuating component of the drag force on all the bubbles passing a fixed point, where the distribution of bubbles is based on the kinetic equation by Reeks [82, 83]. The resulting turbulent dispersion force is

\[ M_{TD,d} = -(C_{vm}\rho_c + \rho_d)\lambda^T \cdot \nabla \alpha, \]  

(3.29)

where \( \lambda^T \) is the phase space diffusion tensor [84]. This equation could be simplified and related to other flow variable under certain conditions as [84, 85]

\[ M_{TD,d} = -C_{TD}\rho_c k_c \nabla \alpha_d, \quad \text{for small bubbles} \]

\[ M_{TD} = -\frac{1}{\tau_d} (C_{vm}\rho_c + \rho_d)(\nu_c^t)_B \nabla \alpha, \quad \text{for large bubbles} \]

\[ \tau_d = \frac{4}{3} \frac{(C_{vm}\rho_c + \rho_d)}{\rho_c C_D} \frac{d}{\nabla |\bar{u}_r|}, \]  

(3.30)

where the term \( |\bar{u}_r| \) is the root mean square relative velocity between phases.

On the other hand, Simonin et al. [86] derived the turbulent dispersion force by taking the time average of the fluctuating part of \( M_{drag} \) and using the drift velocity between phases as

\[ M_{TD,d} = C_{TD}K_{dc} \nu_{dc}^t \left( \frac{\nabla \alpha_c}{\alpha_c} - \frac{\nabla \alpha_d}{\alpha_d} \right), K_{dc} = \frac{|M_{drag}|}{u_r}, \]  

(3.31)

where \( \nu_{dc}^t \) is a modified turbulent viscosity, \( S_{dc} \) is the dispersion Prandtl number and \( K_{dc} \) is sometime referred as the linearized drag. Burns et al. [87] also derived a similar equation based on Farve average, where the turbulent viscosity equals to the eddy viscosity of the continuous phase. This version of turbulent dispersion force may not be applicable when high local void fraction is present, since the term \( \frac{\nabla \alpha_c}{\alpha_c} \) could go to infinity and cause numerical problem.

In the current study, the simple model, \( M_{TD,d} = -C_{TD}\rho_c k_c \nabla \alpha_d \), proposed by Lopez de Bertodano [85] is used with \( C_{TD} = 1 \).
3.2.1.4. Virtual Mass Force

The virtual mass is the additional force when relative acceleration is present. The virtual mass force for a spherical particle using potential flow theory could be derived as [88]

\[ F_{vm,d} = -\frac{\rho_c V_d}{2} \frac{d u_r}{d t}, \]

\[ V_d = \frac{4}{3} \pi r_d^3. \quad (3.32) \]

Applying this to the time averaged equation, the source term could be derived as

\[ M_{vm,d} = -\frac{1}{2} \alpha \rho_c \frac{d u_r}{d t}. \quad (3.33) \]

3.2.1.5. Interfacial Area Concentration and Bubble Diameter

Transport between phases is controlled by interfacial area concentration \( a_i \). Thus, precise prediction of \( a_i \) is crucial in two-phase flow modeling. There are multiple ways of computing \( a_i \) in existing literature. One method is establishing a transport equation for interfacial area concentration, as described by Ishii et al. [89]. This method requires a proper boundary condition and initial condition for \( a_i \) and is still under development.

The other one is an algebraic method including a bubble diameter \( d \) as

\[ a_i = \frac{6 \alpha}{d} \quad (3.34) \]

where \( \alpha \) is the dispersed phase void fraction. Obviously, the bubble diameter \( d \) is important in determining \( a_i \). Regarding the condition of the validation experiment in the current work, a widely used equation for calculating \( d \) in buoyancy driven flow introduced by Gaddis et. al. [33, 78, 90] is used,

\[ d = \left[ \left( \frac{6 L_c \sigma}{\rho_c g} \right)^{\frac{4}{3}} + \left( \frac{81 \nu_c \dot{V}}{\pi g} \right) + \left( \frac{135 \dot{V}^2}{4 \pi^2 g} \right)^{\frac{4}{5}} \right]^{\frac{1}{4}}, \quad (3.35) \]
where $\dot{V}$ is the volumetric flow rate, and $L_c$ is the hydraulic diameter of the inlet orifice. For pipe flow, the bubble diameter could be estimated as

$$d = C \sqrt[\frac{\sigma}{g\Delta \rho}}, \quad C \text{ is an empirical constant.} \quad (3.36)$$

3.2.2. Mass Transfer Coefficient Model

The source term for species mass transfer $I_{l,i}$ is

$$I_{l,i} = k_l a_i \left( K \rho_{l, i} \rho_{g, l} Y_{g, l} - \rho_l Y_{l, i} \right). \quad (3.15)$$

The mass transfer coefficient $k$ needs to be modelled in order to calculate the source term. Regarding the mass transfer with soluble gases, three types of models could be found in the literature. These include the analogy between mass and heat transfer, the penetration model, and the surface renewal model.

From the view of Fick’s law, the species transfer and heat transfer in each phase share the same functional form. Using this analogy, the mass transfer coefficient $k$ could be related to the heat transfer coefficient $h$, and the Sherwood number $Sh$ could be related to the Nusselt number $Nu$. In this way, the experimental correlation for heat transfer, which is easier to measure, could be used for the species mass transfer as [91]

$$k = \frac{Sh_d \left( Re, Sc_i \right) D_l^i}{d_g}$$

$$Nu = 2 + 0.6Re^{\frac{1}{2}}Pr^{\frac{1}{3}} \rightarrow Sh_d = 2 + 0.6Re^{\frac{1}{2}}Sc^{\frac{1}{3}}. \quad (3.37)$$

However, considering the large difference between mass diffusivity and thermal diffusivity, this analogy may not always be applicable.

The penetration model is based on the simple film theory. The mass transfer is assumed to be controlled by a diffusion layer of certain thickness. By proposing a way to calculate this film
thickness, the mass transfer coefficient could be calculated [92]. The slip penetration model proposed by Higbie et al. [93] is

\[ k = \frac{D}{\delta} = C \frac{D}{d \sqrt{u_r}}. \] (3.38)

Another class of model is the surface renewal model, where the species mass transfer is assumed to be controlled by renewal of the liquid near the interface by turbulent eddies. Lamont et al. derived an eddy cell model as [94]

\[ k = C \sqrt{D \epsilon / \nu}, \quad C = 0.4. \] (3.39)

Jajuee et al. [95] performed a more rigorous derivation and arrived at a similar surface renewal stretch model as

\[ k = C \sqrt{D \epsilon / \nu}, \quad C = \frac{4}{\sqrt{\pi}}. \] (3.40)

In their original work, they further evaluate the turbulent dissipation rate \( \epsilon \) based on existing correlation and arrived at the final correlation, which is not used in the present simulations.

3.2.3. Properties of Molten Salt

To simulate the xenon removal process in the molten salt reactor, the properties of molten salt and solute xenon are necessary. A short literature review is included here on the properties of FLiNaK, which is the candidate salt for Transatomic Power MSR, which is also the salt used in the ongoing xenon removal experiment. FLiNaK has a composition of 46.5% LiF, 11.5% NaF and 42% KF. This composition is chosen around the eutectic point of the mixture to minimize its melting point.
3.2.3.1. Density

The density of FLiNaK is shown in Figure 3.3, where the dashed line indicates unknown effective range. The red bold line is used in the simulation, which is the correlation given by Janz [96]. The error bar is plotted to show the uncertainty of the experiment, if reported. The style for other figures is the same. The uncertainty about density measurement is small compared with other properties, and the difference between different measurements could be attributed to the impurities in the salt. Also, different measurements basically fall in the uncertainty region of other correlation. As can be seen from the figure, the density of FLiNaK drops with increasing temperature, as one would expect. In the simulation, the density at 923K is used, which is the operation temperature in the MSRE. The corresponding equations in the figure are given in Table 3.2.

Table 3.2: Density Correlation of FLiNaK

<table>
<thead>
<tr>
<th>Correlation ((kg/m^3))</th>
<th>Effective Range (K)</th>
<th>Uncertainty</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>2555 (-0.6T)</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Hoffman et al. [97]</td>
</tr>
<tr>
<td>2729.3 (-0.73T)</td>
<td>Unknown</td>
<td>(\pm 5%)</td>
<td>Powers et al. [3, 98]</td>
</tr>
<tr>
<td>2579.3 (-0.624T)</td>
<td>940 – 1170</td>
<td>(\pm 2%)</td>
<td>Janz [96]</td>
</tr>
<tr>
<td>2655.64 (-0.68T)</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Cohen et al. [3, 99]</td>
</tr>
<tr>
<td>2603 (-0.669T)</td>
<td>750 – 1100</td>
<td>Unknown</td>
<td>Salanne et al. [100]</td>
</tr>
</tbody>
</table>
Figure 3.3: Density of FLiNaK

Figure 3.4: Viscosity of FLiNaK
3.2.3.2. Viscosity

The viscosity of FLiNaK is quite sensitive to the temperature of the salt as shown in Figure 3.4. Especially, the viscosity changes significantly near the melting point. This sensitivity is not present in water-based heat transfer system, which might require special care when performing a safety analysis. As for the temperature used in the simulation, at 923K, the viscosity is around five times of that of water at room temperature. Considering the density of the salt, the kinematic viscosity is around 2.5 times of water at room temperature. For a scaling experiment, 30% CaCl₂ solution has been used in literature to match the kinematic viscosity of the liquid. The correlation given by Kubíková et al. [101] is selected in this document. The difference between measurements is larger than that for density, which is likely from the impurities in the salt. The well-known Stokes–Einstein equation indicates that the viscosity of a mixture will increase with impurities. Alternatively, the different could be caused by the slight difference in the salt composition used in different experiments.

Table 3.3: Viscosity Correlation of FLiNaK

<table>
<thead>
<tr>
<th>Correlation ((kg/m \cdot s))</th>
<th>Effective Range</th>
<th>Uncertainty</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2.5 \times 10^{-5} e^{\frac{4790}{T}})</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Hoffman et al. [97]</td>
</tr>
<tr>
<td>(4 \times 10^{-5} e^{\frac{4170}{T}})</td>
<td>773 – 1073</td>
<td>10%</td>
<td>Cohen et al. [102]</td>
</tr>
<tr>
<td>(3.877 \times 10^{-5} e^{\frac{4327}{T}})</td>
<td>770 – 1025</td>
<td>2.4 \times 10^{-5}</td>
<td>Kubíková et al. [101]</td>
</tr>
<tr>
<td>(1.1 \times 10^{-4} e^{\frac{3379}{T}})</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Vriesema [103]</td>
</tr>
<tr>
<td>(2.487 \times 10^{-5} e^{\frac{4478.62}{T}})</td>
<td>770 – 1173</td>
<td>±2%</td>
<td>Janz [96]</td>
</tr>
<tr>
<td>(6.23 \times 10^{-5} e^{\frac{3921.4}{T}})</td>
<td>727 – 1144</td>
<td>Unknown</td>
<td>Merzlyakov et al. [104]</td>
</tr>
<tr>
<td>(e^{-3.049 \times 10^{-3}} e^{\frac{3847}{T}})</td>
<td>993 – 1163</td>
<td>±2.5%</td>
<td>Cibulková et al. [105]</td>
</tr>
<tr>
<td>(1.633 \times 10^{-3} e^{\left(\frac{2762.9}{T}+\frac{3.115E6}{T^2}\right)})</td>
<td>773 – 993</td>
<td>±2%</td>
<td>Tørklep et al. [106]</td>
</tr>
</tbody>
</table>
Figure 3.5: Thermal Conductivity of FLiNaK

Figure 3.6: Specific Heat Capacity of FLiNaK
3.2.3.3. Thermal Conductivity

The correlations for thermal conductivity and specific heat capacity are also included here, though in the current CFD model the liquid is assumed to be isothermal. From Figure 3.5, the dispute in literature is obvious. Ewing et al. [107] argued that the difference with the measured thermal conductivity is likely from the radiation and natural convection, which is not considered in some of the early experiments [108]. Without the radiation, the thermal conductivity would be overestimated. This could explain the relatively large value given in the figure. In this document, the recent correlation given by Khokhlov et al. [109] is recommended, which basically predicts the same thermal conductivity given by Smirnov et al. [110].

Table 3.4: Thermal Conductivity Correlation of FLiNaK

<table>
<thead>
<tr>
<th>Correlation ((J/kg \cdot K))</th>
<th>Effective Range</th>
<th>Uncertainty</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.5</td>
<td>810.93 – 963.71</td>
<td>Unknown</td>
<td>Hoffman et al. [97, 111]</td>
</tr>
<tr>
<td>2.6</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Cohen et al. [102]</td>
</tr>
<tr>
<td>0.6</td>
<td>763 – 1125</td>
<td>Unknown</td>
<td>Ewing et al. [107]</td>
</tr>
<tr>
<td>1.3</td>
<td>Unknown</td>
<td>Unknown</td>
<td>Vriesema [103]</td>
</tr>
<tr>
<td>(0.36 + 5.6 \times 10^{-4}T)</td>
<td>790 – 1080</td>
<td>±0.012</td>
<td>Smirnov et al. [110]</td>
</tr>
<tr>
<td>(24.288 – 6.042 \times 10^{-2}T + 4.222 \times 10^{-5}T^2)</td>
<td>800 – 920</td>
<td>±25%</td>
<td>Janz et al. [96]</td>
</tr>
<tr>
<td>(-0.34 + 0.5 \times 10^{-3} + 32/M)</td>
<td>790 – 1080</td>
<td>±4%</td>
<td>Khokhlov et al. [109]</td>
</tr>
</tbody>
</table>

3.2.3.4. Specific Heat Capacity

The specific heat capacity of FLiNaK is shown in Figure 3.6. In the review by Williams [112], it is argued that the dependency of heat capacity with temperature is small. Salanne et al. [100] reported a constant heat capacity of 1769\(J/kg \cdot K\), and similarly Hoffman et al. [97, 111] reported a constant value of 1884.06\(J/kg \cdot K\). On the other hand, the data taken by Rogers et al.
[113] suggests that the heat capacity increases with temperature in the operation range of MSR. Nevertheless, the existing measurements roughly fall within each other’s uncertainty range. In this document, the correlation by Rogers et al. [113] is suggested.

Table 3.5: Specific Heat Capacity Correlation of FLiNaK

<table>
<thead>
<tr>
<th>Correlation ((J/kg \cdot K))</th>
<th>Effective Range</th>
<th>Uncertainty</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>1884.06</td>
<td>755.37 – 1144.26</td>
<td>±10%</td>
<td>Hoffman et al. [97, 111]</td>
</tr>
<tr>
<td>979.22 + 1.0657T</td>
<td>750 – 860</td>
<td>±2%</td>
<td>Rogers et al. [113]</td>
</tr>
<tr>
<td>1906.3</td>
<td>750 – 830</td>
<td>±4%</td>
<td>Janz et al. [96]</td>
</tr>
<tr>
<td>660 + 1.37T</td>
<td>1063 – 1233</td>
<td>±13</td>
<td>Khokhlov et al. [109]</td>
</tr>
<tr>
<td>1769</td>
<td>750 – 1100</td>
<td>Unknown</td>
<td>Salanne et al. [100]</td>
</tr>
</tbody>
</table>

3.2.3.5. Surface Tension

Table 3.6: Surface Tension Correlation of FLiNaK

<table>
<thead>
<tr>
<th>Correlation ((10^{-4}T))</th>
<th>Effective Range</th>
<th>Uncertainty</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2726 – 1.014 \times 10^{-4}T</td>
<td>770 – 1040</td>
<td>±2%</td>
<td>[96]</td>
</tr>
</tbody>
</table>

The surface tension of the salt is important in the calculation of drag for distorted bubbles and the bubble diameter. Only limited data could be found for this quantity. The correlation reported by Janz et al. [96] is used in this document.
Figure 3.7: Solubility of Helium in FLiNaK with Pressure

Figure 3.8: Solubility of Noble Gases in FLiNaK
3.2.3.6. Xenon Solubility and Diffusivity

The solubility data of noble gas in molten salt is quite limited. In terms of solubility of noble gas in FLiNaK, only one experiment could be found in the literature, which is conducted by Blander et al. [69] at ORNL. It is found in the experiment that the addition of 4% UF4 to the salt has basically no impact on the solubility. The solubility of gases in liquid could generally be expressed by Henry’s law, that the equilibrium molar fraction of the dissolved gas is proportional to the gas’ partial pressure in the gas phase as

$$p_g^i = HX_{i,d}^e.$$  \hspace{1cm} (3.14)

This relation is usually valid in low pressure and dilute mixture cases. Blander et al. [69] measured the solubility of Helium in FLiNaK at different partial pressure, as shown in Figure 3.7. The linear relationship between the solubility and partial pressure is a confirmation of Henry’s law for the inert gas-FLiNaK system.

Then, the solubility of He, Ne, Ar in the fluoride at different temperatures are experimentally measured, as shown in Figure 3.8. The solubility data are expressed in terms of $K_{k,i}^e$, which is the equilibrium ratio in Henry’s law in different formats, as

$$K_{k,i}^\rho = \frac{\rho_k Y_{k,i}^e}{\rho_j Y_{j,i}^e} = K_{k,i}^e.$$  \hspace{1cm} (3.41)

The equilibrium ratio will increase with temperature and decrease with molecule mass or diameter, as shown in the figure.

Xenon solubility data for FLiNaK are not available, but the theoretical values are listed, as shown in Table 3.7. For none reacting gas-liquid system, the solubility of gases could be calculated based on classic chemical equilibrium theory as

$$RT \ln K_{k,i}^\rho = T \Delta S - \Delta H$$
\[ \ln K^p_{k,i} = -\frac{\Delta \mu}{k_B T}. \]  

(3.42)

According to the second equation, the solubility could be calculated with the chemical potential \( \Delta \mu \) of the dissolution process. An early theory assumes the dissolution is governed by the “surface energy” to form cavities in the salt, which is used by Blander et al. [69, 114] A more sophisticated model would consider the effect of volume exclusion, dispersion interaction and polarization of gas and salt [115]. Nevertheless, the theoretic value given here is likely within the right magnitude according to the comparison of theoretic value and measurement for other gases. Therefore, the theoretical solubility of Xenon would be used in our simulation, until experimental becomes available.

Table 3.7: Solubility of Noble Gases in FLiNaK [116]

<table>
<thead>
<tr>
<th>Gas</th>
<th>Temp. (°C)</th>
<th>Measured ( K^p_{k,i} \times 10^3 )</th>
<th>Calculated ( K^p_{k,i} \times 10^3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>600</td>
<td>8.09</td>
<td>28.3</td>
</tr>
<tr>
<td></td>
<td>700</td>
<td>14.0</td>
<td>46.8</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>20.3</td>
<td>70.7</td>
</tr>
<tr>
<td>Ne</td>
<td>600</td>
<td>3.12</td>
<td>3.94</td>
</tr>
<tr>
<td></td>
<td>700</td>
<td>6.00</td>
<td>8.63</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>9.84</td>
<td>16.4</td>
</tr>
<tr>
<td>Ar</td>
<td>600</td>
<td>0.645</td>
<td>0.146</td>
</tr>
<tr>
<td></td>
<td>700</td>
<td>1.43</td>
<td>0.509</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>2.99</td>
<td>1.41</td>
</tr>
<tr>
<td>Xe</td>
<td>600</td>
<td>-</td>
<td>0.011</td>
</tr>
<tr>
<td></td>
<td>700</td>
<td>-</td>
<td>0.057</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>-</td>
<td>0.212</td>
</tr>
</tbody>
</table>
CHAPTER 4: VALIDATION EXPERIMENTS

Limited data for validation of CFD is available in literature, and what is available only includes measurement of one or two of the three important quantities related to liquid-gas mass transfer. For experiments focused on hydrodynamics, void fraction and liquid velocity profiles might be available. For mass transfer experiments, void fraction and volumetric mass transfer coefficient are usually measured. Though some simulation works validated their model in these three aspects [34], the fact that different geometries and conditions were used in those experiments would inevitably reduce the fidelity of the validation. In the attempt to fill this gap and provide confidence for our CFD model, a series of experiments on the same apparatus have been carried out with flow visualization, PIV and dissolved oxygen measurement. The apparatus used in these experiments is a simplified version of an ongoing two-phase mass transfer experiment in molten salt, as a first step towards the design of a gaseous fission product removal system. The experimental data obtained here will be used to validate our CFD model, which will be further applied to the molten salt experiment for comparison. The validated model with information supplied by the molten salt experiment will be finally used in a design analysis of the fission product removal system in a subsequent study.

The experiments are carried out in a small-scale cylindrical bubble column made of acrylic, as shown in Figure 4.1. The bubble column is 2.5 inch in diameter and 6.55 inch in height. This aspect ratio is slightly smaller compared with industrial cylindrical bubble columns but is adopted anyway in accordance with the ongoing molten salt experiment. Different pieces of acrylic are cut and glued together. A 0.12-inch hole at the center of the bottom serves as the gas inlet. A corresponding acrylic flange could be attached at the top of the bubble column, where sparging tube and other instruments could be installed. This design is to replicate the other ongoing molten
salt experiment with air-water, as shown in Figure 4.2. For the model validation experiments, this setup introduces unnecessary complexity to the geometry modeling. Therefore, in most of the experiments, the gas is injected from the bottom. In the experiment, the bubble column is filled with deionized water. The gas used in this experiment is air, from the laboratory compressed air supply. The flowrate is measured with Dwyer RMB-49-SSV panel mounted gas flow meter, which has a measurement range from 0.5SCFH to 15SCFH and 5% accuracy. The cylinder is surrounded by a square water box filled with water to limit the refraction caused by the column curvature. With the water box and thin wall thickness, the influence of refraction becomes negligible. This is confirmed by pictures of an immersed standard ruler at different location. The spacing of the ticks is practically uniform. The actual physical length for pixels could be easily derived based on the known dimensions of the bubble column. However, for the part that is not covered by the water box, the refraction is not negligible, and the corresponding area in the obtained videos is cropped out. In the following section, procedures and results of each experiment will be discussed in detail.

Figure 4.1: Bubble Column with Water Box Used in Our Experiment.
4.1.1. Visualization Experiment

Flow visualization with high speed camera is widely used in two-phase flow experiments. Visualization can provide qualitative information of bubble movement, morphology and phase distribution in a wide region, which are often useful for flow regime identification and could infer more accurate measurement. Research is also made for quantitative measurement of bubble velocity and size distribution with visualization, though the accuracy is often limited. In this section, the technique for time averaged void fraction measurement with flow visualization is introduced. The results are compared with existing experimental correlation as a validation.

4.1.1.1. Experiment Procedure

The flow visualization experiments were carried out by sparging in air bubbles at different flowrates from the bottom. Photron’s high speed camera MINI AX100 is used in our experiments to generate the high-speed videos. A frame rate of 250 fps is found to be enough for bubble
identification, tracking, and accurate measurement of void fraction. The maximum measurement time of the camera is limited by its internal memory. Therefore, a higher frame rate is not favorable since the total averaging time will be reduced.

Proper illumination of the flow region is important to obtain a high-quality video for post processing. First is the selection of light source. At high frame rate, the change in luminosity of the incandescent light bulb caused by the AC power cycle becomes noticeable, which is troublesome for post processing. One solution is to use DC power with the incandescent light bulb, which would be quite expensive. The other choices are halogen photography lamps [117] and LED light [49]. The frequency of LED light is controlled by the electronic circuits driving them, which can create a vast array of refresh rates. Some high-power LED light is free of this flickering, and SANSI’s 5500lm flood light is used in our experiment. Apart from the selection of light, the illumination method is also important. To obtain a uniform background, the light source is usually placed behind a light screen and the flow region. The uniform backlight would be reflected at the bubble interface, making the boundary of the recorded bubbles appears darker than the center and the background. This distinction in brightness would allow for bubble identification. The intensity of backlight should be such that this contrast is maximized.

Prior to the bubble injection, a short video of the background was taken for contrast. The background image is calculated by averaging the pre-recorded high-speed video, which could minimize the disturbance of the light source. This background image is important for bubble identification, which will be discussed in the next section.
4.1.1.2. Image Processing

The videos are processed in MATLAB to extract the information of bubbles. These videos are taken in 8bit grayscale model. The brighter pixel possesses a higher intensity, and the overall level scale is from 0 to 255.

First, the videos are cropped to the region where the water box exists, since the refraction through the wall could be corrected. Then, the pre-recorded background is subtracted from each frame. Since the background is brighter than the bubbles, this subtraction is not directly the subtraction of the grayscale value. The easiest way to perform this subtraction is to first inverse the images and then make the subtraction. The resulting figure is inversed again to restore the normal grayscale. This process produces a figure with white background and black bubbles. The inverse of the grayscale is given as

$$I_{\text{inv}} = |255 - I|. \quad (4.1)$$
After the background extraction, the images are enhanced by adaptive histogram equalization. This algorithm performs histogram equalization on small areas of the figure to minimize the noise processed. The target histogram is a flat distribution, and the equalization would distribute the local intensity value more evenly. After the enhancing, the figure is subjected to a Gaussian filtering to remove the noise in the original figure and those created in the enhancing. At last, the obtained grayscale image is reversed and converted to binary image, where morphology algorithm becomes available. The objects in the binarized figures are identified by continuity of the white regions. Since the bubble is only darker at its boundaries, these objects often form a ring, rather than a solid region after binarization. It is then necessary to fill these hollow regions. The final filled bubbles and other images are shown in Figure 4.3.

In the binarized image, bubbles are identified by continuity of the white regions. With this identification, the equivalent diameter, estimated volume and the averaged void fraction in this region could be calculated with the following equations,

\[
\begin{align*}
\text{\( r_{eq} \)} & = \sqrt{\frac{A}{\pi}} \\
\text{\( V_{est} \)} & = \frac{4}{3} \pi r_{eq}^3 \\
\text{\( \alpha_g \)} & = \frac{\sum V_{est}}{V_{volume}}.
\end{align*}
\] (4.2)

The existence of cap, spherical and elliptical bubbles at the same time poses difficulties for the identification, since distinguishing them in 2D images are not easy, as shown in Figure 4.3. Therefore, instead of specifically sorting the bubbles into groups, they are all treated equally in the processing. This is compensated by the usage of equivalent diameter. The total area is used to derive the equivalent diameter, which preserved the cross-sectional area. The error associated with this method is analyzed in the Appendix. The measurement results for volume averaged void
fraction or gas hold up at different gas flowrates and the convergence behavior of this method are shown in Figure 4.4 and Figure 4.5.

Figure 4.4: The Convergence of Void Fraction with Increasing Frames.

Figure 4.5: The Average Void Fraction at Different Superficial Gas Velocity.
The most favorable feature of flow visualization experiment is that it provides information of the flow over the whole region. In addition to the measurement of volume averaged void fraction, we could obtain a 2D void fraction profile with the visualization method. From the identified bubble diameter and estimated volume, we could project the instant bubble distribution onto a 2D plane and average it in time. This is exactly the definition of void fraction used in Eulerian-Eulerian two-fluid model [58]. The projection is done by weighing the bubble by its relative dimension in the projection direction. This will produce a 2D line averaged void fraction profile that could be compared with 3D simulation data after line averaging, as shown in Figure 4.6. The procedure is described by the equations below, the bubble diameter is multiplied with a distribution function \( \eta \) considering the shapes of individual bubbles,

\[
\alpha_{2D}(x, y) = \left( \phi(x, y, t) \frac{\eta(x, y, t)d_{eq}}{L_{proj}} \right)
\]

\( \phi = 1 \) when bubble is present, \( \phi = 0 \) when liquid is present.

\[
\eta(x, y, t) = \frac{\sqrt{r_m^2 - r^2}}{r_m}, \quad r_m = \max(r), \quad r \text{ is the distance to the centroid.}
\]

The choice of \( \eta \) is important to produce the correct projected void fraction profile. The form of \( \eta \) is taken from the simple geometric relationship of a sphere, as shown in Figure 4.7. However, the bubbles in the experiment are not perfect spheres, and how to choose the scaling factor becomes troublesome. If the equivalent diameter is chosen to be the scaling factor, the off-center value will drop very fast, causing a significant underestimation of the void fraction. Therefore, the farthest point from the center \( r_m \) is chosen as \( R \). The 2D void fraction profile and the volume averaged void fraction obtained with the method described in this section will serve as an important way to validate the CFD model.
The Geometric Meaning of $\eta$

Using $d_{eq}$

Using $r_m$

Figure 4.6: Process of Obtaining the 2D Void Fraction Profile

Figure 4.7: The Distribution Function $\eta$ and its Effect
4.1.2. Particle Image Velocimetry (PIV) Experiment

In this section, the widely used particle image velocimetry for velocity measurement is discussed. PIV is extensively used in single phase flow experiment to measure the flow velocity. The velocity in a wide region could be measured simultaneously with acceptable error for most engineering purposes. However, when being used in two-phase flow, additional challenges are introduced by the existence of interfaces. The problem and the corresponding solution are discussed in the following section.

4.1.2.1. Experiment Procedure

The Particle Image Velocimetry (PIV) experiment is carried out basically under the same condition as the flow visualization experiment. The geometry, flow condition and camera setup are the same. The LED illumination is now replaced with a thin laser sheet across the center of the
flow region, which is perpendicular to the high-speed camera. The laser sheet is created by Dantec Dynamics’ RayPower system. The laser is synchronized with the high-speed camera so that the shutter and the laser pulse is triggered at the same time, which also stabilizes the laser intensity for longtime operation and therefore increase the accuracy. Each pair of frames is taken in 0.002s, and the frame-pairs are taken at 20Hz. An example of the frame-pair (double frame) is shown in Figure 4.8. The short interval between frames ensures the accuracy of the PIV measurement. On the other hand, the low frequency of recorded frame-pairs or double-frames allows averaging over a long period, which is important to obtain a time averaged profile.

Before sparging starts, proper amount of polyamide seeding particles of 50μm are added to the water. These particles will reflect the laser and appear to be a white dot in the obtained video. In order to increase the contrast of the image and therefore the accuracy, the laser should be the only light source in the experiment, unless other light sources could be filtered out by special optical fiber. Too many or too few particles could increase the uncertainty of the measurement.

4.1.2.2. Data Processing

The video is first processed to prepare the PIV data, then analyzed with adaptive PIV algorithm using Dynamic Studio by DANTEC. The images are divided into different interrogation areas, which could be overlapping or not overlapping. Each particle in the image is best to occupy more than two pixels for best performance of the algorithm. This requires a proper choice of particle diameter. The number of particles is best above 10 in each interrogation area. Limited number of particles could increase the uncertainty of the measurement. On the other hand, if the particles are too dense in the flow, the uncertainty is also increased. In addition to the number of particles, the interrogation area should also be smaller or at least comparable to the characteristic length of the flow. The liquid velocity is obtained by analyzing the cross-correlation between the
frames. The resulting vector could be viewed as the average velocity within each interrogation area.

The difficulty with two-phase PIV experiment is the existence of phase boundary, which also causes reflection in addition to the seeding particles. To minimize the error introduced by the reflected laser at bubble interface, the large bright area in the obtained video is masked, as shown in Figure 4.9. The details of this method are discussed in the work by Sathe et al. [48].

The velocity field and contour plot at $U_g = 2.5 \text{ mm/s}$ after time averaging are shown in Figure 4.10. The constant presence of bubbles near the inlet region introduces considerable error to the final vector map. For this reason, a transparent box is put in the figure. Another interesting measurement is the turbulence kinetic energy of the liquid phase, which could be estimated as

$$k^t = \frac{1}{2} (2u'{}^2 + v'{}^2).$$

(4.4)

Here, the fluctuation is assumed to be axially symmetric to arrive at the turbulence energy in Figure 4.11. However, since the interrogation area is larger than the smallest turbulence scale, we could lose a part of the energy spectrum, therefore underestimating $k^t$. On the other hand, the uncertainty of the measurement will inevitably be included in the real fluctuation, which would likely cause a small overestimation in the result.
Figure 4.9: The Masked Image and Instant Velocity Field.

Figure 4.10: Velocity Field and Axial Velocity from PIV Experiment at \( U_g = 2.5 \text{mm/s} \)
4.1.3. Dissolved Oxygen Charging Experiment

The mass transfer between the liquid and the gas is directly related to the removal of gaseous fission product. Various models are proposed for the mass transfer rate. In this section, the mass transfer coefficient is measured by monitoring the concentration of dissolved oxygen in the sparging process. The results from this experiment would be used to inspect and calibrate the mass transfer model in our simulation.

4.1.3.1. Experiment Procedure

The mass transfer coefficient of the bubble column is measured by monitoring the oxygen concentration in water. The dissolved oxygen (DO) concentration is measured by YSI’s optical DO probe, FDO 4410 IDS. It has an accuracy of 1.5% and measurement range of 0 to 20 mg/L and a 95% response time of 45 second. The probe has a semi-permeable membrane where dissolved oxygen could enter. The presence of oxygen in this membrane with embedded
luminescent dye would change the characteristic of the luminescence when blue excitation light is applied. This change is proportional to the partial pressure of oxygen in the membrane, which could be measured by the optical sensor in the probe. Since oxygen is not consumed in this process, there is no requirement for minimum flow rate as required by galvanic dissolved oxygen probe, meaning the disturbance of the flow could be minimized in the sparging process by placing it in the low velocity region.

In the experiment, the deionized water is first boiled for around a minute to remove the dissolved oxygen. Then the low dissolved oxygen water is sealed and stored in the bubble column for several hours for it to cool down to room temperature, therefore the solubility change due to temperature variation during experiment is negligible. When the temperature reaches the room temperature, the DO probe is inserted into the water from above, and air is sparged into the liquid from the bottom to charge it with oxygen, as shown in Figure 4.3. The injection is from a compressed air supply line, which is controlled by an in-line pressure regulator and followed by a panel-mounted air flowmeter. The concentration of the dissolved oxygen is measured every second. Considering the response time of the probe, certain lag would exist in the measurement result. The result taken at each second should be viewed as a time averaged value over the characteristic response time. However, this would not change the time constant of the process if the process is much longer than the response time.

Based on Henry’s law and two-resistance model [22], the absorbed oxygen per unit volume per unit time could be expressed as:

\[ \Gamma_{l,i} = k a_i \left( K_{l,i}^0 \rho_g Y_{g,i} - \rho_l Y_{l,i} \right). \]  

(3.17)

By assuming a homogenous oxygen concentration over the volume, the average oxygen concentration could be calculated by
\[ C_{l,O^2} - C_{l,O^2}^* = (C_{l,O^2}^0 - C_{l,O^2}^*)e^{-ka_it}. \]  (4.5)

The \( ka_i \) term in the exponential function is the volumetric mass transfer coefficient and could be calculated based on our dissolved oxygen concentration data. Variables \( C_{l,O^2}^* \) and \( C_{l,O^2}^0 \) are the equilibrium and initial concentration of the dissolved oxygen. The inverse of the volumetric mass transfer coefficient has the dimension of time, which is the characteristic time for this process. The equilibrium concentration is determined by running the sparging process at high flow rate long enough until the concentration of dissolved oxygen becomes stable.

### 4.1.3.2. Results and Discussion

Figure 4.12 shows the result obtained at a flowrate of 1 SCFH, corresponding to \( U_g \) of 2.5 mm/s. In this figure, the concentration of oxygen changes with time in an exponential manner, as we expected for a homogeneous solution. Thus, this concentration could be properly described by the expression that is derived above. By fitting the curve with least square method, the important volumetric mass transfer coefficient \( ka_i \) could be extracted. It is possible to decouple the mass transfer coefficient \( k \) from the interfacial area \( a_i \). Though, without accurate and direct measurement of \( a_i \), this decoupling is not so beneficial with the huge uncertainty caused by it. Thus, it is more justified to keep the parameter directly measured from the experiment, without any empiricism introduced.

A series of experiments are carried out at different gas flowrates where three experiments at the same condition were done to check the repeatability of our method. To validate the experiment procedure, the experiment data are also compared with the existing experiment correlation given by Hikita et al. [118]. The results are shown in Figure 4.13. From the comparison, the agreement between the correlation and our result is acceptable relative to the uncertainty of the measurement. The minor difference between our measurement and previous experiment could
result from the different geometries used in the experiments, which could not be reflected by superficial velocity alone. The repeatability of the experiment results is an indication of the small uncertainties of our method. A detailed uncertainty analysis is included in the Appendix.

From Figure 4.13, it is shown that the mass transfer coefficient increases with the superficial velocity of the gas. The slope, on the other hand, is decreased with increasing gas flowrate.

\[ (C - C_{sat}) = (C_0 - C_{sat}) \exp(-ka_t t) \]

Coefficients (with 95% confidence bounds):

\[ (C - C_{sat}) = 5.396(5.39, 5.402) \]

\[ ka_t = -0.003148(-0.003153, -0.003143) \]

Figure 4.12: Dissolved Oxygen Concentration during Air Bubble Sparging
Figure 4.13: Relation between $ka_i$ and $U_g$
CHAPTER 5: SIMULATION RESULTS AND DISCUSSION

In order to choose the proper constitutive relation, grid size and boundary condition, a series of sensitivity studies are carried out. The simulation results for different cases are compared with each other. Some experiment results are also used here for reference. The base case we arrived at from these sensitivities study is summarized in Table 5.1. The simulation is carried out in ANSYS FLUENT 19.2 with user defined functions. The UDFs are included in the Appendix B.

Table 5.1: Summary of Models and Setups for Base Case

<table>
<thead>
<tr>
<th>Setups</th>
<th>Turbulence</th>
<th>Bubble Induced Turbulence</th>
<th>Mesh</th>
<th>Outlet</th>
<th>Bubble Diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>Forces</td>
<td>Drag</td>
<td>Lift</td>
<td>Turbulent Dispersion</td>
<td>Virtual Mass</td>
<td>Wall Lubrication</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RNG $k - \varepsilon$ [119] Tchen’s Theory [66]</td>
<td>Sato [64]</td>
<td>Medium</td>
<td>Free Surface</td>
<td>Gaddis [90]</td>
<td></td>
</tr>
<tr>
<td>Forces</td>
<td>Ishii and Zuber [58]</td>
<td>Tomiyama [77]</td>
<td>Lopez de Bertodano [85]</td>
<td>Not Included</td>
<td>Not Included</td>
</tr>
</tbody>
</table>

5.1. Sensitivity Study

In this section, the results of various sensitivity studies are presented. The sensitivity studies of different constitutive relation, model parameters, boundary conditions and mesh are carried out and reported. The results from these studies would help us make reasonable simplification and assumption for our simulation.

5.1.1. Grid Independence

To determine a proper grid size, a grid independence study was performed on three sets of mesh. The resulting void fraction profiles at the axial cross section are illustrated in Figure 5.1.
The profiles for different grid sizes are quite similar. Examining the volume averaged void fraction, as shown in Table 5.2, the medium mesh is good enough for our purpose, since the variation by using a much finer mesh is only 0.5%. The averaged void fraction is chosen to represent the convergence because it is the controlling factor of interfacial mass transfer. Thus, in the remainder of this document, unless specifically mentioned, the medium mesh is used in the simulation.

![Figure 5.1: Grid Independence Study – Void Fraction](image)

Table 5.2: Grid Independence Study with Averaged Void Fraction

<table>
<thead>
<tr>
<th>Cell Number</th>
<th>24026</th>
<th>81969</th>
<th>240324</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha^v$</td>
<td>$7.22 \times 10^{-3}$</td>
<td>$7.51 \times 10^{-3}$</td>
<td>$7.55 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

5.1.2. Influence of Interfacial Forces

As discussed in Section 3.2.1, various interfacial forces have been proposed in the literature. The lateral forces such as lift force and turbulent dispersion force are not fully understood. In this sense, a sensitivity study of these forces could show their relevance to our flow
condition. For sake of computational power and numerical stability, it may not be necessary to include all these forces. The data from simulation is projected to the 2D plane by line average,

\[ \alpha_{2D} = \frac{\sum \alpha_i V_i \kappa_i}{\sum V_i \kappa_i}, \]  

(5.1)

where \( V_i \) is the cell volume intersected with the projection line. The factor \( \kappa_i \) is multiplied considering the different importance of mesh cells, which is a function of characteristic cell length and distance to the projection line.

From the result shown in Figure 5.2, the lift force in this case will reduce the dispersion of gas phase, indicating the lift coefficient is reversed in sign. The model used here is the Tomiyama model. Comparing the simulation result with and without the lift force, it could be concluded that lift force only has a secondary effect to the void fraction distribution and volume averaged void fraction. The gray box on the top of experimental figure accounts the blocked area by the flange. The volume averaging for simulation is only carried out in the region that is not blocked. Turbulent dispersion force, on the other hand, plays a significant role in the simulation. When it is not included, the dispersion of gas phase is limited and far from the experiment result. However, this does not mean the turbulent dispersion force would always have a large impact regardless of the flow condition. In our experiment, the gas injection into the liquid introduces significant amount of shear induced and bubble induced turbulence, which is different from ordinary two-phase pipe flow. This turbulence is the root of the strong influence of turbulent dispersion force. Lastly, the virtual mass force only slightly changes the void fraction distribution near the inlet. The relative velocity soon reaches its maximum, and the influence of virtual mass force becomes negligible. On the other hand, the virtual mass force model could cause some numeric instability and increase the computational cost. Therefore, unless strong acceleration is found, the virtual mass force will not be included in the simulation.
Experiment
\[ \alpha^v = 0.752\% \]

Base-Case
\[ \alpha^v = 0.751\% \]

No Lift Force
\[ \alpha^v = 0.782\% \]

No Turbulent Dispersion
\[ \alpha^v = 0.340\% \]

With Virtual Mass Force
\[ \alpha^v = 0.798\% \]

Figure 5.2: Sensitivity of Interfacial Forces
5.1.3. Modeling of Drag

In Section 3.2.1.1, three drag models are introduced: Tomiyama [76], Schiller-Naumann [75] and Ishii-Zuber model [58]. The comparison of these models is shown in Figure 5.3. The void fraction profiles share a similar shape, though the void fraction with Schiller-Naumann model [75] is much smaller than the other two. The Schiller model does not consider the increase of drag when the bubbles are distorted. And as the result, significant underestimation of the void fraction is presented, which is clearly shown in the comparison. The difference between Ishii-Zuber model [58] and Tomiyama model [76], on the other hand, is small. The Tomiyama model does not consider the modification of drag when multiple bubbles are present, which leads to a smaller void fraction prediction. However, in the case of low void fraction flow, this modification seems to be limited, even when locally high void fraction is present. Nevertheless, since the result of Ishii-Zuber model is more close to the experiment, and is applicable to more general cases, it is selected in our modeling.
Figure 5.3: Comparison of Drag Models – Void Fraction
5.1.4. Modeling of Lift

In Section 5.1.2, it is shown that the lift force only has a secondary effect on the void fraction profile. Here, the modeling of lift force is examined in more detail. In Figure 5.4, simulation results with different lift models are compared with the experiment. \( C_L = 0.5 \) corresponds to the lift force from potential flow theory, which is often used for air-water bubble column with small bubbles. Since the bubble size in our experiment and simulation is larger than those cases, the overprediction of the dispersion is not surprising. The Tomiyama model [78] and Hibiki and Ishii model [79] take account of the inversion of lift force at large bubble diameter. Comparing with the case without lift force, it is clear that the lift force is inversed in our simulation. In fact, the Tomiyama model gives a uniform \( C_L = -0.27 \), and the Ishii-Hibiki model results in approximately a \( C_L \) of -0.5.

From the figure, it is shown that Tomiyama model gives better result compared with the Hibiki and Ishii model. Thus, Tomiyama model is selected in our modeling. However, as mentioned in Section 3.2.1, the lateral forces are difficult to be distinguished from each other in the experiment. The change of turbulent dispersion and turbulence model might change the conclusion that are being drawn here. The models selected could be accurate enough for engineering purpose, but the modeling of lateral forces certainly requires more experimental and theoretical research.
Figure 5.4: Comparison of Lift Models – Projected Void Fraction
5.1.5. Turbulence Modeling

The turbulence modeling for multiphase flow includes two parts. The first part is modeling of the shear induced turbulence for each phase, and the second part is the modeling of bubble induced turbulence. The modeling of shear induced turbulence is not so different from single phase flow, though in our simulation, the turbulence equation for gas phase is not solved, but predicted with Tchen’s theory [66]. This method could reduce the equations to be solved, and only has a limited influence on the result, since the density and viscosity of gas phase are much smaller than the liquid.

On the other hand, the liquid phase turbulence is limited to the category of $k-\epsilon$ two-equation models, because the mass transfer model we are using explicitly utilizes the turbulent dissipation rate. Within the category of $k-\epsilon$ models, a similar comparison is performed for the renormalization group model (RNG) and realizable model [119, 120]. Since the $k-\epsilon$ model modifies the effective viscosity of the liquid phase, which would change the liquid phase velocity, the velocity field at the cross section is compared with the result from PIV experiment in Figure 5.5. Apart from that, the void fraction profiles are also compared in Figure 5.6. From the velocity field, the realizable model predicts a higher liquid velocity, and thus a smaller void fraction. Apart from that, the location of maximum velocity is shifted downwards. The RNG model gives a more accurate velocity field and averaged void fraction and is thus selected for our simulation model.

The other part of the modeling, the bubble induced turbulence, is described in section 3.1.1.4. Since it arises from the time averaging of the governing equation, the absence of it is likely to cause the solution to be unstable in time. This is confirmed by the simulation result shown in Figure 5.7. The distribution of void fraction starts to fluctuate and constantly changes its shape.
without the bubble induced turbulence term. Thus, it should always be considered whenever large, deforming bubbles are expected to be present.

Figure 5.5: Comparison of Turbulence Model – Axial Velocity
Figure 5.6: Comparison of Turbulence Model (L: RNG, R: Realizable) – Void Fraction

Figure 5.7: Sensitivity of Bubble Induced Turbulence – Void Fraction
5.1.6. Modeling of Boundary Condition

The boundary condition for a bubble column is somewhat troublesome. Ideally, the simulation should include the free surface and the cover gas above it. The actual outlet should be a pressure outlet at the top of the cover gas. The void fraction in the cover gas should be one, and the outflow should be purely gas. However, as discussed by Jakobsen et al. [31], this approach may cause numerical difficulty for the solver. On the other hand, theoretically, even though the time averaging method at the free surface may still be able to smooth out the discontinuity of the interface, the constitutive relation derived for dispersed flow will be invalid. One possible solution is to create multiple flow regimes and detect the free surface based on the gradient of void fraction. Each regime has its own constitutive relation. However, this is even more numerically difficult to solve and much more expensive, and the conservation of mass at the interface is not well preserved, which makes it impractical for our goal.

Thus, a simplified boundary condition is chosen. The top of the computational domain is assumed to be the free surface of the liquid. Since the simulation is restricted in the liquid phase, the shape and the flow condition at the free surface are not known a priori. Thus, certain assumptions need to be made. A common assumption is to set the top surface as a free-slip wall, where the normal velocity of the liquid is zero and no friction exist on the wall. This is close if the gas flowrate is not too large. Another practice is using the regular pressure outlet at the top surface, and the volume fraction for gas in the backflow is set to be zero. In terms of hydrodynamics, these two assumptions are not so different if the water level is large enough compared to the column diameter. The comparison of these two boundary conditions is shown in Figure 5.8 and Figure 5.9. However, when it comes to mass transfer, the fact that the pressure outlet allows backflow becomes problematic. Since the condition at the boundary is not known a priori, the species concentration
in the backflow could not be determined, which causes significant difficulties in tracking the total mass of species in the liquid. Thus, the free-slip wall assumption is adopted in our model.

Figure 5.8: Sensitivity of Boundary Condition – Void Fraction

Figure 5.9: Sensitivity of Boundary Condition – Liquid Velocity
5.1.7. Sensitivity of Bubble Diameter

The bubble diameter used in the simulation is calculated based on the inlet condition and fluid properties according to the following equation by Gaddis et. al. [90]

\[ d = \left[ \left( \frac{6L\sigma}{\rho_c g} \right)^{\frac{4}{3}} + \left( \frac{81\nu_c V}{\pi g} \right) + \left( \frac{135V^2}{4\pi^2 g^2} \right)^{\frac{1}{5}} \right]^{\frac{1}{4}}. \] (3.35)

The bubble diameter at 2.5mm/s superficial velocity is 7.2mm. This equation is derived for the departure diameter of the bubble at the inlet based on force balance. As long as the breakup and coalescence are limited, this diameter should represent the characteristic length of the bubbles. To assess the influence of choosing the bubble diameter, a sensitivity study is performed for 3 different sizes that are observed in the experiment at 2.5mm/s. The result is shown in Figure 5.10. Bubble diameters larger and smaller than the calculated bubble diameter are selected. Bubble diameter larger than 7.2mm only slightly change the result. On the other hand, with a diameter much smaller than 7.2mm, the void fraction does increase. From experiment data [58], the terminal velocity of a single distorted bubble is independent of the particle size. Thus, the void fraction is rather independent of the bubble diameter for the current case. However, by reducing the bubble diameter, the drag is not completely in the distorted regime over the computational domain, and the terminal velocity is reduced, and thus the void fraction is increased. The conclusion from this study is that, if the bubbles stay in the distorted regime, the exact prediction of the bubble diameter is not necessary to obtain a velocity field and void fraction profile. However, for smaller bubbles, the simulation result will be more sensitive to the diameter, and experiment may be preferable to assist the simulation.
Figure 5.10: Sensitivity of Bubble Diameter – Void Fraction
5.1.8. Sensitivity of Material Properties

In some preliminary xenon removal design for molten salt reactors, the salt is diverted from the main flow to remove the xenon, where the temperature could be changed. Also, different MSR designs have different operating temperatures and salt composition, resulting in different salt properties. Thus, it is necessary to study the sensitivity of material properties on the key transport quantities in the sparging process. In the following section, the sensitivity of liquid phase properties is presented. The properties used in the simulation are taken at three different temperature points (750K, 922K and 1100K) based on the reported experiment data, corresponding to approximate upper and lower bound of FLiNaK operation temperature. Thus, the variation of density (2111kg/m$^3$ to 1893kg/m$^3$) is not as significant as that of viscosity (0.0124Pa · s to 0.002Pa · s). The sensitivity of viscosity is shown in Figure 5.11. From the result, the void fraction increases significantly with viscosity, because the viscous force becomes the limiting factor of the void fraction once the gas phase leaves the inlet region. On the contrary, the volumetric mass transfer coefficient decreases considerably. Still, this result is consistent with reported experiments, as summarized in [39]. The high viscosity reduces the velocity of the gas phase, and thus increases the residence time for the bubbles, which gives rise to the void fraction. On the other hand, decreasing $k_{ai}$ with increasing $\alpha_g$ clearly indicates a negative sensitivity of viscosity to $k$ or $A_i$. Li and Prakash pointed out that the high viscosity will increase the portion of large bubbles, which reduces the interfacial area and $k_{ai}$ [121]. However, in the current simulation, only one group of bubbles is considered, and the change of bubble diameter with viscosity change is insignificant to that of the mass transfer coefficient. Thus, the decrease of $k_{ai}$ here is caused by decrease of $k$, as indicated by the correlations in Section 3.2.2. From the analysis above, the interfacial mass transfer coefficient $k$ also has an important role in the sensitivity of viscosity.
Physically, the high viscosity increases the boundary layer thickness, which will have a negative effect on convection and thus the mass transfer coefficient.

The sensitivity of density, however, is not as significant as that of the viscosity. From Figure 5.12, the void fraction is practically unchanged with density. The magnitude of the void fraction change is comparable to the numerical uncertainty. From the experiment correlations reported in [39], the void fraction is a weak function of density, with a scaling factor around $-0.15$. Regarding mass transfer coefficient, $k_a$ slightly increases with liquid density. This also agrees with the trend reported in experimental study [122]. Looking at the governing equation in Chapter 3, nearly every term scales linearly with liquid density, except for the transport term of the gas phase. This means the change in liquid density will affect the gas phase movement, which in turn changes the liquid phase flow field. However, if the average void fraction is limited, a limited effect of density is expected.

The sensitivity study of density and viscosity is reported above. Since it is not possible to vary density or viscosity alone for FLiNaK, the combined thermal effect should be studied. Density, viscosity and surface tension at different temperatures are considered in this combined effect simulation. The result is shown in Figure 5.13. It appears that the temperature effect is dominated by variation in viscosity. The volumetric mass transfer coefficient increases with temperature, where the slope is slightly larger, compared with viscosity effect alone. The void fraction decreases with the temperature as expected, but with a smaller slope compared with viscosity alone. From the discussion above, we already know reducing the density and increasing the viscosity have contrary effect on void fraction, and thus we have a reduced slope here. The result for mass transfer coefficient is similar with the case reported for the air-water system. However, it is opposite to the result in an experiment with a CO$_2$ – Molten Carbonate system [21]. In that experiment, they
conclude that the opposite relation to air-water system could be a result of surface tension. They argue that higher surface tension could reduce the rising velocity for bubbles and thus reduce the mass transfer coefficient $k$, which leads to the reduction of $ka_i$. However, it should be noted that the reduction of rising velocity should also increase the void fraction, which would increase the interfacial area concentration $a_i$, leading to the increase of $ka_i$. Thus, further experiment and more inclusive modeling is needed to confirm the temperature effect on $ka_i$ in molten salt system.

Figure 5.11: Sensitivity of Viscosity
Figure 5.12: Sensitivity of Density

Figure 5.13: Sensitivity of Temperature
5.2. Benchmark Study

In Section 5.1, a sensitivity study is performed and compared with experiment data. The model is then validated against experiment data and some simple cases with analytical solutions.

5.2.1. Validation of Void Fraction Prediction

From the visualization experiment, the volume averaged void fraction could be measured. These volume averaged quantities play an important role in determining the proper CFD model. The sensitivity studies are performed at 2.5mm/s superficial gas velocity, and here we would like to see its performance at different gas flowrates. The result is considered acceptable, with a maximum error of 3%, as shown in Figure 5.14. The simulation agrees quite well with the measured void fraction. The result from the visualization experiment is compared with the plot given by Krepper et al. [123] to validate the experimental procedure. The difference between our measurements and reported data could be attributed to the difference in the geometries of the bubble columns, which couldn’t be reflected by superficial velocity alone.

In addition to the volume averaged measurement, the simulation is also compared with the localized void fraction data. The radial void fraction profile at three axial locations from experiment and simulation are compared in Figure 5.15, Figure 5.16 and Figure 5.17.

From the comparison, general agreement between the simulation and experiment for the radial profile is found. It appears that the void fraction near the inlet is underpredicted, yet overpredicted at the top. The underprediction near the inlet may be related to the absence of virtual mass force, without which the velocity of the gas phase could be overestimated. From axial location of 6.8 cm to 9 cm, the predicted void fraction profiles are essentially unchanged, yet the measured void fraction profile becomes more dispersed. This may indicate the deficiency of the models for lateral forces. Nevertheless, the agreement of averaged void fraction and radial profile
between experiment and simulation is considered acceptable, with a maximum local error less than 25%. This validation study puts confidence in the simulation result at low gas flowrate, which is the flowrate to be used in the molten salt experiment.

Figure 5.14: Comparison of Predicted Averaged Void Fraction with Experiment
Figure 5.15: Void Fraction Profile 6.8 cm Above the Inlet at 2.5mm/s $U_g$

Figure 5.16: Void Fraction Profile 3.8 cm Above the Inlet at 2.5mm/s $U_g$
Figure 5.17: Void Fraction Profile 9 cm above the Inlet at 2.5mm/s $U_g$

Figure 5.18: Comparison of Liquid Velocity Field at 2.5mm/s $U_g$
5.2.2. Validation of Liquid Velocity Prediction

The void fraction prediction of our CFD model has been validated against visualization experiments. The other important flow variable to be considered is the velocity field. The reliable measurement technique for gas velocity on a plane has not been well established, but the liquid velocity field could be measured quite accurately with the Particle Image Velocimetry (PIV) method introduced in Section 4.1.2. Since the diffusivity in gas phase is much larger than that in the liquid, the concentration of fission product in the gas phase should be more uniform, which makes the gas velocity profile less important in the fission product removal process. However, the concentration profile of the fission product is highly related to the circulation in the liquid phase. The velocity field at 2.5mm/s $U_g$ is shown in Figure 5.18. General agreement between the experiment and simulation is found, though the movement near the inlet is not well captured. To quantitively compare the velocity, contour plots of the axial velocity are shown in Figure 5.19.

From the contour plot, the velocity field near the inlet is not well captured. However, further from the inlet, the agreement between the experiment and simulation improves. This could be seen from Figure 5.20, Figure 5.21 and Figure 5.22. Above axial location of 6.8 cm from the inlet, the simulation and measurement closely match. The measured velocity profile appears to be asymmetric, which is caused by the reflection at the bubble interface. The laser illuminates from the left, and a bright area will be produced in the PIV image when the laser is directly onto the bubbles. Post processing of the images removes most of these highlighted areas, but still causes a small overprediction of the velocity field on the right. In conclusion, our CFD model could satisfactorily predict the velocity field when strong localized effect is not present. Even when these effects are present, general agreement is still achieved.
Figure 5.19: Comparison of Liquid Axial Velocity, $U_z$ [m/s], at 2.5mm/s $U_g$

Figure 5.20: Comparison of Liquid Axial Velocity at 3.8cm
Figure 5.21: Comparison of Liquid Axial Velocity at 6.8cm

Figure 5.22: Comparison of Liquid Axial Velocity at 9cm
5.2.3. Verification of Pure Diffusion

Before the mass transfer validation, the diffusion within the phase is tested first, which is necessary to capture the species concentration distribution within the liquid. Considering the simple 1D diffusion problem, the concentration on one side of the cylinder is prescribed and the flux on the other is zero, and the initial concentration in the liquid is zero. The solution could be solved analytically if axial symmetry is assumed.

\[
\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2}
\]

\[
C(z = 0) = C^0
\]

\[
\frac{\partial C}{\partial z} (z = H) = 0
\]

\[
C - C_0 = -C^0 \sum_{n=0} \sin[(n + 0.5)\left(\frac{\pi z}{H}\right)] \frac{e^{-\left(\frac{(n+0.5)^2\pi^2 D t}{H^2}\right)}}{(2n+1)\pi}.
\]

The simulation result is compared with the analytical solution in Figure 5.23. The concentration of xenon on one side is set to be the equilibrium concentration at 900 psi, which is the maximum xenon pressure in the molten salt experiment. From the result, the simulation and analytical result agree exactly with each other, confirming that species diffusion in the simulation is well captured.
5.2.4. Verification of Species Transport at Free Surface

Due to the chemical equilibrium at the interface, the species concentration has a sharp change at the thin liquid-gas interface. This sharp change becomes a discontinuity when the interface is modeled without thickness. This discontinuity, in the case of dispersed interface, could be smoothed, resulting in an extra source term in the species transport equation. The rate of species transfer could be related to other flow variables and the concentration gradient between phases. However, at the free surface, this is no longer available, as discussed in Section 3.1.2.4. Therefore, special treatment of mass transfer at the interface is needed. The Continuous-Species-Transfer (CST) is implemented and tested for a simple 1D problem [70, 71]. At the beginning, the liquid is charged with certain gas and in contact with another gas at the free surface. Assuming axial symmetry, the problem becomes 1D. The equilibrium ratio at the interface is arbitrarily set as 3,
and the diffusivity is different for gas and liquid. The concentration distribution could be easily derived by solving the 1D equation as

\[ 3C_{l,i} = C_{g,i} \]

\[ D_l \nabla C_{l,i} = D_g \nabla C_{g,i}, \ \text{at the interface} \]

\[ \frac{\partial C}{\partial t} = D \nabla^2 C. \] (5.3)

The comparison of the CST model and the dispersed flow model is shown in Figure 5.24. From the result, it is shown that since the dispersed model does not capture the jump of species concentration and thus the diffusion-driven species transfer process, yet the CST model gives a much better prediction. When the liquid is in motion and convection also contributes to the mass transfer, the dispersed model may result in less accuracy. However, if we want to study the capability of cover gas to extract xenon from the salt, the CST model or other method needs to be applied in order to correctly capture the physics.

![Graph showing species transport at free surface](image)

Figure 5.24: Comparison of Different Species Transport Model at Free Surface
5.2.5. Validation of Mass Transfer Prediction

Now that the velocity and void fraction prediction have been validated, the last step is to validate the mass transfer model. In Section 3.1.2, the modeling of species transport is discussed in detail. The constitutive model for mass transfer coefficient needs to be related to local flow variables. The Ranz-Marshall model is based on analogy between mass transfer and heat transfer, utilizing the widely used Nusselt number correlation developed by Ranz et al. [124]. The surface renewal model and surface renewal stretch model are based on the similar assumption that the mass transfer near the bubble surface was controlled by renewal rate of the liquid at the bubble surface [94, 95], which is then related to liquid turbulent dissipation rate. The simulation result obtained with different models is listed in Table 5.3. The prediction of the Surface Renewal Stretch model is quite close to the experiment result. However, the prediction given by Ranz-Marshall model [124] is far from the experiment, which is likely to be related to the difference of phase interface, since liquid drops were used in the original work by Ranz et al. [124] and we shouldn’t expect identical behavior to a gas bubble. The mobility of the interface and the turbulence near it might have a big impact on the result, considering the good performance of the Surface Renewal Stretch model. Moreover, the analogy between heat and mass transfer itself may not hold, with the large difference between thermal diffusivity and mass diffusivity. The two surface renewal models, on the other hand, yield much better result. The performance of the surface renewal model is greatly improved, and the surface renewal model almost yield the exact experimental result without any calibration, indicating the physics related to this phenomenon is captured.

Table 5.3: Simulation and experimental volumetric mass transfer at \( U_g = 2.5mm/s \)

<table>
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<tr>
<td>( ka_l (1/s) )</td>
<td>2.07 \times 10^{-4}</td>
<td>1.2 \times 10^{-3}</td>
<td>3.2 \times 10^{-3}</td>
<td>3.1 \times 10^{-3}</td>
</tr>
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</table>
The concentration profiles of the dissolved oxygen during the simulation for the surface renewal stretch model at $U_g = 2.5 \text{mm/s}$ are shown in Figure 5.25. At the beginning of the simulation, the mass fraction of oxygen in the liquid phase is set as $4.2 \times 10^{-6}$. The colormap is fixed to show the comparison between different profiles. The concentration of the dissolved oxygen becomes basically uniform after 30s. Though local variation of the concentration still exists, it is negligible compared with the concentration change. This observation justifies the volume averaged method described in Equation (4.5), where the liquid is treated as a homogeneous solution. The direct comparison of the dissolved oxygen concentration is shown in Figure 5.26. Good agreement is found between the simulation and experiment in the 114.7s simulation time. From the result, the CFD model could predict the mass transfer process within the uncertainty of the experiment measurement.

Figure 5.25: Concentration Profile During the Simulation
Figure 5.26: Comparison of the Dissolved Oxygen Concentration
CHAPTER 6: SUMMARY AND FUTURE WORK

A CFD model is proposed for simulation of the xenon removal process in molten salt with inert gas sparging. In order to predict the mass transfer rate, the velocity and phase distribution of each phase are needed. Therefore, the Eulerian two-fluid model coupled with species transfer is used in the simulation to capture the local velocity, phase distribution and species concentration at the same time. For accurate prediction of the process, a careful evaluation of the constitutive relations and material properties is required. Therefore, the interfacial forces, turbulence model, material properties, mesh size, bubble diameter and boundary conditions are first evaluated through sensitivity study in a cylindrical bubble column. The geometry is chosen according to an ongoing molten salt xenon experiment. The results obtained in the current study could serve as a reference for the corresponding experiment and be compared with the future experiment result.

In the sensitivity study, the drag force and turbulent dispersion force are identified as important in the process of gas injection. The drag force is the most important force for bubbly flow. It controls the relative velocity between phases and therefore the residence time of the bubble in the column, which determines the total void fraction of the gas phase. When the bubble size becomes relatively large, its shape would significantly deviate from a spherical particle. The corresponding drag coefficient would change significantly compared with a perfect sphere. This phenomenon should be considered by the drag model. The turbulent dispersion force accounts for the dispersion of dispersed particles in a turbulent fluid. It would flatten the distribution of void fraction and push the bubbles into the bulk liquid. When bubbles are injected into the liquid, strong shear induced turbulence is present, making the turbulent dispersion force relatively large compared to other lateral forces. On the other hand, the lift and virtual mass force only have secondary effect on the result. The lift force describes the movement of bubble in a shear flow. In
the current geometry, its magnitude is small compared with the turbulent dispersion force. However, in flow with limited turbulence, this force would be quite important for correct prediction of the void fraction profile. The virtual mass force is only presented when particles are accelerated. In the region slightly away from the injection, its influence could be ignored. Apart from the interfacial forces, it is shown that the bubble induced turbulence also plays an important role in predicting the right void fraction profile. As for bubble diameter, the simulation is not quite sensitive to it if the flow is in the distorted bubble regime. This result from the fact that for large distorted bubble, its rising velocity in a stagnation liquid is basically constant relative to its size. However, if smaller bubbles are present, the modeling of bubble diameter becomes very important for the simulation.

In order to validate the CFD model, three sets of experiment are done in an air-water bubble column with the same geometry used in the simulation. Since the specific interaction model is scaled, the model validated with air-water experiment is applicable to molten salt in principle. The void fraction profile is measured with a visualization experiment, the velocity profile is obtained with Particle Image Velocimetry and the mass transfer coefficient is measured by tracking the dissolved oxygen concentration. Through the validation, it is concluded that the CFD model could satisfactorily predict the volume averaged void fraction. The local void fraction and velocity profile a few centimeters away from the inlet could be predicted, though the agreement immediately near the inlet is limited. The mass transfer coefficient from the simulation is compared with the experimental data. It is concluded that with calibration from the experiment, the mass transfer could be predicted. However, further evaluation of the mass transfer model is needed.

With the CFD model established and validated against various cases, it is concluded that the model can reasonably capture the underlining physics of the two-phase mass transfer process,
and therefore as an engineering tool for designing a commercial scale xenon removal system. However, the simulation is only validated against the air-water experiments, with known properties of oxygen solubility and diffusivity. Though theoretical estimation for xenon solubility and empirical value for diffusivity in molten salt are reported and used in the current work, the accuracy of these values is still questionable. Moreover, the models validated with air-water experiments may still need to be evaluated with experiment in molten salt. Though the models are written in dimensionless form, its dependence on the material properties may not be sufficiently validated, since most of the validations are carried out in air-water or steam-water flow. Another potential issue is that the flow in a commercial scale xenon removal system could be much more complicated than the flow presented in the validation experiment. The model reported here may not be completely applicable in various conditions of the removal system.

With the discussion above, various experiments in molten salt are favorable. A carefully designed xenon experiment is needed to obtain the solubility and diffusivity of xenon in molten salt, which are directly related to the mass transfer process. Without concrete measurement of these properties, the uncertainty of simulation would be relatively large. The CFD model developed in the thesis could be applied to such experiments for validation, and the result from the simulation could in return infer the experiment procedure.

Other than the properties of xenon, the two-phase flow phenomenon in molten salt is basically an unvisited research area. It would be helpful to infer from such experiments that whether the drag law, the bubble morphology map and the bubble diameter prediction obtained from air-water experiment is still applicable to the molten salt system. Similar visualization experiment presented in the thesis could be performed for molten salt. However, the procedure and material used in such experiment is expected to be quite different.
For a natural application of the model developed in this thesis, it could be used to simulate the prototypical xenon removal system designed for the MSBR [7]. The performance of the different components could be simulated and compared with the reported experimental data. Once satisfactory result is obtained, the sensitivity of various design parameters could be evaluated. Moreover, an extended validation in co-current pipe flow could be helpful to the design analysis.
REFERENCES

12. Kasten, P.R., E.S. Bettis, and R.C. Robertson, *Design studies of 1000-Mw(e) molten salt breeder reactors.* ORNL-3996, Oak Ridge National Lab., Tenn.


APPENDIX A: UNCERTAINTY ANALYSIS

A.1. Uncertainty for Visualization Experiment

The void fraction in the experiment is derived by estimating the volume of each bubble. The possible uncertainty sources are: 1) uncertainty from the gas flow meter, 2) the error in the bubble identification process, and 3) the error from the volume estimation method.

1) Error from the gas flow meter

   The quantity being measured is the void fraction at a point, and the averaged void fraction. Since the void fraction is relatively small, a linear dependence of gas flow rate could be assumed. This assumption is reasonable, judging from the experiment result. Therefore, the void fraction could be written as the flowrate, $q$, times some other function. The uncertainty is therefore additive.

   This is also true for the other three uncertainty sources.

   \[
   \alpha = qf
   \]

   \[
   \frac{d\alpha}{\alpha} = \sqrt{\left(\frac{dq}{q}\right)^2 + \left(\frac{df}{f}\right)^2}.
   \]

   The uncertainty of the gas flow meter is 5%.

2) Error in the bubble identification process

   In obtaining the individual bubbles, the images are processed. Some bubbles are not fully recognized and lose a part of their shape. Sometimes, two bubbles are recognized as one, since they are connected. By looking at each frame, roughly speaking, around 10% bubbles lost a small fraction, and around 10% bubbles become connected. Since this error is highly related to the error in the volume estimation, a rigorous evaluation is not easily performed. Instead, this error is considered an error to the correct bubble volume. In this way, the error for bubble identification could be expressed as
\[
\frac{V_{ob}}{V_{ext}} = \left(\frac{V_{ob}}{V_{ap}}\right)\left(\frac{V_{ap}}{V_{ext}}\right)
\]

\[
d\frac{V_{ob}}{V_{ext}} = \sqrt{\left(\frac{dV_{ob}}{V_{ap}}\right)^2 + \left(\frac{dV_{ap}}{V_{ext}}\right)^2}.
\]

The relative error shown here, after multiplied with its probability of occurrence, equals to \(\frac{df}{f}\). For most cases, the bubbles lost 20% of its cross-sectional area. Assuming the bubble is spherical, this means

\[
r_{ap} = \sqrt{0.8A/\pi}
\]

\[
V_{est} = \frac{4}{3}\pi r_{ext}^3
\]

\[
V_{ap} = \frac{4}{3}\pi r_{ext}^3 \times (0.8)^3
\]

\[
\frac{V_{ap}}{V_{est}} = 0.72.
\]

Since only 10% of bubble has this problem, the final uncertainty should be 2.8%. Considering the contribution from worst cases, this uncertainty is increased to 5%.

One the other hand, for the bubbles in contact with each other, the overlapped cross section is around 40% of their cross-sectional area. For these bubbles, assuming they are identical, we have

\[
r_{ap} = \sqrt{1.6A/\pi}
\]

\[
V_{est} = \frac{8}{3}\pi r_{ext}^3
\]

\[
V_{ap} = \frac{4}{3}\pi r_{ext}^3 \times (1.6)^3
\]

\[
\frac{V_{ap}}{V_{est}} = \frac{(1.6)^3}{2} \approx 1.
\]
This means, for most cases, the overlapping does result in significant uncertainty. For the extreme cases, where there is no overlapping or completed overlapped, the error becomes

$$\frac{V_{ap}}{V_{est}} = \frac{(2)^3}{2} = 1.41$$

$$\frac{V_{ap}}{V_{est}} = 0.5.$$

In a way, the two errors are balancing each other. Since we are only concerned with time averaged measurement, this error will be much smaller than the error from losing a part of the bubble shape.

3) Error in estimating the bubble volume.

This error comes from the fact that bubbles are not perfect spheres, but we are still assuming them to be sphere in the volume estimation method. The error related with this error source could be estimated by considering the typical distorted bubble shape. The cap and elliptical bubbles with prescribed dimensions are analyzed with this method.

To begin with, for a random oriented surface element in space, the projected area onto a plane is proportional to its surface area as

$$< A_{proj} > = \frac{1}{4} < A_{surf} >.$$  

Considering an ellipsoid with $a = 1, b = 2, c = 4$, which is already a highly distorted bubble, the derivation goes as

$$A_{surf} \approx \left[ \frac{(ab)^{1.6} + (bc)^{1.6} + (ac)^{1.6}}{3} \right]^{\frac{1}{1.6}} 4\pi$$

$$< V_{est} > = \frac{4}{3} \pi \left[ \frac{< A_{surf} >}{4\pi} \right]^{\frac{3}{2}}.$$
\[ V_{el} = \frac{4}{3} \pi abc \]

\[ \frac{V}{<V_{est}>} = 0.704. \]

which means even for highly distorted bubbles, the overestimation of our measurement is still within an acceptable range. The error associated with cap bubbles could be analyzed with a similar method, which could lead to an underestimation of the bubble volume around the same level of uncertainty. The different errors for cap bubble and ellipse bubble compensate with each other, which would reduce the overall error of our measurement. Since we are averaging the measurement in time, and the occurrence of cap shape and ellipsoid bubbles are similar, the error should be greatly reduced. To be conservative, assuming a gaussian distribution for the error, and using the 3\( \sigma \) principle to reduce the maximum error by 3. This is not rigorously defined but meant to give an upper limit for this error source.

Combining the error from the three process, a conservative estimation of uncertainty in measured void fraction is given as,

\[ \frac{\Delta \alpha}{\alpha} = \sqrt{5\%^2 + 5\%^2 + 10\%^2} = 12.2\%. \]

A.2. Uncertainty of the Mass Transfer Coefficient

The uncertainty of mass transfer coefficient measurement could come from the 1) gas flowrate, 2) the oxygen probe and 3) the curve fitting uncertainty.

1) Error from the gas flow meter is described previously and results in 5% uncertainty.

2) Error from the oxygen probe: The oxygen probe has an accuracy of 1.5% based on manufacturer specifications.
3) The uncertainty from curve fitting: The uncertainty from curve fitting is negligible compared with the other two error sources, on the order of 0.1%.

The combined uncertainty for mass transfer coefficient measurement would be

\[ \frac{dk}{k} = \sqrt{5\%^2 + 1.5\%^2} = 5.2\%. \]

A.3. Uncertainty of the PIV Experiment

The uncertainty of PIV measurements, particularly in two-phase flow, remains an open research area. Reliable method for determining the uncertainty for instantaneous measurement is still under development. The result of a PIV experiment is obtained by calculating the cross correlation of the two frames at each interrogation area, which is hard to be directly related to other quantities [125]. Instead, an easier way of estimating the error is to relate the uncertainty of measurement to the cross-correlation plane. Using the Peak Height Ratio method introduced by Charonko et al. [126], instantaneous errors at each interrogation area is obtained. This uncertainty should be interpreted as the uncertainty of measurement, rather than systematic or instrumental error, since it is commonly accepted that the PIV technique itself is bias free [125]. However, the result used in this document is the time averaged velocity, therefore these errors are averaged with the volume weighted root mean square as

\[ \bar{\epsilon} = \sqrt{\frac{\sum (u_i \epsilon_i)^2}{\sum u_i^2}}. \]

The distribution of uncertainty at different points is shown in Figure A.1. From the result, an uncertainty of 0.1 is reasonable for the instantaneous measurement. The uncertainty above 0.1 is mostly located in the region close to the inlet, which was already reported as low confidence. With repeating measurement of the same quantity, the uncertainty will be reduced by a factor of
\( \sqrt{n} \), where \( n \) is the number of measurements we made. The problem here is the time averaged profile is not obtained with one frame pair, but thousands of frames, and the meaning of the number of measurements \( n \) is obscured. In Figure A.2, the time averaged axial velocity at 6.9 cm above the inlet is shown. It appears 2000 frame pairs are enough to obtain the time averaged velocity, which is 400s in time. Since over 4000 frame pairs is recorded in the experiment, we could argue that two measurements were carried out in the process. Assuming the uncertainty of 0.1 is carried along from the instantaneous measurement to the time averaged measurement, we arrived at an uncertainty of \( \frac{10}{\sqrt{2}}\% \approx 7\% \). From the local maximum and minimum of the measurement, with a variation of 5\%, the uncertainty of 7\% seems to be a good estimation.

Figure A.1: Averaged Uncertainty
Figure A.2: Convergence of Averaged Axial Velocity at axial location of 6.9 cm
APPENDIX B: UDFS

B.1. UDF for Mass Transfer Coefficient for Surface Renewal Stretch Model

surface_renewal_stretch.cpp

#include "udf.h"
//UDF written in c++ for use in FLUENT 19.2 for the surface renewal stretch model

DEFINE_LINEARIZED_MASS_TRANSFER(surface_renewal, c, mixture_thread, from_phase_index, from_species_index, to_phase_index, to_species_index, lin_from, lin_to) {
  real diameter = 0.01;
  real kRatio = 1.0/33.6163;
  real diffusivity = 2.4E-9;
  real kViscosity = 1.0048E-6;
  real K = sqrt(4*diffusivity/3.1415927)*6.0/sqrt(sqrt(kViscosity))/diameter;
  real gDensity = 1.225;
  real lDensity = 998.2;

  Thread *liq = THREAD_SUB_THREAD(mixture_thread, from_phase_index);
  Thread *gas = THREAD_SUB_THREAD(mixture_thread, to_phase_index);

  real Tdr = C_D(c,liq);
  real yGas = C_YI(c,gas,to_species_index)*gDensity;
  real yLiq = C_YI(c,liq,from_species_index)*lDensity;
  real gVof = C_VOF(c,gas);

  *lin_from = K*sqrt(sqrt(Tdr))*(yLiq-kRatio*yGas);
  *lin_to = -K*sqrt(sqrt(Tdr))*(yLiq-kRatio*yGas);

  return K*sqrt(sqrt(Tdr))*(yLiq-kRatio*yGas)*gVof;
}

114
B.2. UDF for CST Model for Free Surface Mass Transfer

`SourceTerm.cpp`

```cpp
#include "udf.h"
#include "sup_source.h"

DEFINE_SOURCE(c_free_source,c,t,dS,eqn)
{
    /* Calculate the divergence of a term using GS theorem to get the source term
     * real flux = 0.0, vol, source = 0;
     * int n;
     * face_t f;
     * Thread *tf, *tf2;
     * // This is for pointing to the secondary phase to get the VOF value
     * int phase_domain_index = 1;
     * // 1 is the phase domain index for secondary phase. And we should set the secondary phase as liquid, or salt, to be consistent with the other definitions.
     * real rc[ND_ND];
     * C_CENTROID(rc,c,t);
     * 
     * vol = C_VOLUME(c,t);
     * 
     * There is no way to make S = A phi + B */
    dS[eqn] = 0;

    /* loop through all the faces to get the sum of flux */
    c_face_loop(c,t,n)
    {
        real NV_VEC(c_current), NV_VEC(A), NV_VEC(G_Vof_f);
        real NV_VEC(Gf1), NV_VEC(Gf2);
        real K = 3;
        real C_f, Vof_f = 0, D;
        real rf[ND_ND], cf[ND_ND];
        /* The VOF here should be that of the liquid */
        cell_t c0, c1 = -1;
        Thread *t0, *t1 = NULL;
        /* get the id for the face and its thread */
        f = C_FACE(c,t,n);
        tf = C_FACE_THREAD(c,t,n);
        /* Vector for the face element A and reset its direction outward*/
        F_AREA(A,f,tf);
        F_CENTROID(rf,f,tf);
        NV_VV(cf,=,rf,-,rc);
    }
}
```
if (NV_DOT(cf,A)<0)
{
    NV_VS(A,=,A,*(-1));
}

/* Now we need to get the C at the face.
A shame is that we can't get it directly*/
c0 = F_C0(f,tf);
c1 = F_C1(f,tf);
t0 = THREAD_T0(tf);
t1 = THREAD_T1(tf);

if (BOUNDARY_FACE_THREAD_P(tf))
{
    /* if the face is at the boundary, we just use the cell value.
   Though C might be set at the boundary but what we get is a zero flux
   I imagine C will not be stored this way
   Just use the cell center value*/
    C_f = C_UDSI(c0,t0,0);
    D = C_UDSI_DIFF(c,t,0);
}
else
{
    C_f = (C_UDSI(c0,t0,0)+C_UDSI(c1,t1,0))/2;
    D = (C_UDSI_DIFF(c0,t0,0)+C_UDSI_DIFF(c1,t1,0))/2;
}

//'f' is the id for the current face in its face thread 'tf'.
//we need to change this thread to a deeper level with THREAD_SUB_THREAD
//we should define tf2 as the phase level thread, as above

//switch to sub-thread at 2ed phase
//re-define the temporary pointers
tf2 = THREAD_SUB_THREAD(tf,phase_domain_index);

c0 = F_C0(f,tf2);
c1 = F_C1(f,tf2);
t0 = THREAD_T0(tf2);
t1 = THREAD_T1(tf2);
if (BOUNDARY_FACE_THREAD_P(tf2))
{
    Vof_f = C_VOF(c0,t0);
    vec_vof(Gf1,c0,t0);
\[ NV_V(G_{\text{Vof}_f},=,Gf1); \]

else
{
\[ V_{\text{Vof}_f} = (C_{\text{VOF}}(c0,t0)+C_{\text{VOF}}(c1,t1))/2; \]
vec_vof(Gf1,c0,t0);  
vec_vof(Gf2,c1,t1);  
NV_V_VS(G_{\text{Vof}_f},=,G_{\text{Vof}_f},+,Gf1,*,0.5);  
NV_V_VS(G_{\text{Vof}_f},=,G_{\text{Vof}_f},+,Gf2,*,0.5);  
}

//NV_D(G_{\text{Vof}_f},=,1,1,1);
NV_VS(c\_current, =, G_{\text{Vof}_f}, *, C_f*(K - 1)/(Vof_f+K*(1-Vof_f))*D);
flux += NV_DOT(c\_current,A);
}
source = flux/vol;
return source;
}

sup_source.h

int vec_vof(real ga[], cell_t c, Thread *t)
{
    //ga is the gradient to be created, c is the cell id, t is the thread for th is cell.
    //Write the second level loop for creating vof gradient
    //Create the vector, at the given cell center.

    //using g-s theorem to get the gradient
real ga_temp[3];
real vol = 0;
face_t f;
Thread *tf, *tf2;
in n;
real rc[ND_ND];
C\_CENTROID(rc,c,t);
// initialize the gradient, in case it is not given properly.
vol = C\_VOLUME(c,t);
NV_D(ga,=,0,0,0);

c\_face\_loop(c,t,n)
{
    real NV\_VEC(A);
    real Vof_f = 0;
    real rf[ND_ND], cf[ND_ND];
/* The VOF here should be that of the liquid */
cell_t c0, c1 = -1;
Thread *t0, *t1 = NULL;
/* get the id for the face and its thread */
f = C_FACE(c,t,n);
tf = C_FACE_THREAD(c,t,n);
/* Vector for the face element A*/
F_AREA(A,f,tf);
F_CENTROID(rf,f,tf);
NV_VV(cf,=,rf,-,rc);
if (NV_DOT(cf,A)<0)
{
    NV_VS(A,=,A,*(-1));
}

tf2 = THREAD_SUB_THREAD(tf,1);
c0 = F_C0(f,tf2);
c1 = F_C1(f,tf2);
t0 = THREAD_T0(tf2);
t1 = THREAD_T1(tf2);
if (BOUNDARY_FACE_THREAD_P(tf2))
{
    Vof_f = C_VOF(c0,t0);
}
else
{
    Vof_f = (C_VOF(c0,t0)+C_VOF(c1,t1))/2;
}
Vof_f = 1;
NV_VVS(ga,=,ga,+,A,*,Vof_f);
}

ga[0]=ga[0]/vol;
ga[1]=ga[1]/vol;

return 0;
}

TransientTerm.cpp
#include "udf.h"

DEFINE_UDS_UNSTEADY(c_unsteady,c,t,i,apu,su)
{
    /* Remove density in the transient term */
    real physical_dt, vol, phi_old;
    physical_dt = CURRENT_TIMESTEP;
}
vol = C_VOLUME(c,t);
*apu = -vol / physical_dt;/* implicit part*/
phi_old = C_STORAGE_R(c,t,SV_UDSI_M1(i));
*su = vol*phi_old/physical_dt;/* explicit part*/
}

VolumeFlux.cpp
# include "udf.h"
DEFINE_UDS_FLUX(volumeflux, f, t, i)
{
    /* rho is the average density at the face, as used by FLUENT
    rho1 is the density at this cell center
    rho2 is then density for the adjacent center
    */
    real rho,rho1,rho2;
    if (BOUNDARY_FACE_THREAD_P(t))
    {
        rho = C_R(F_C0(f,t),THREAD_T0(t));
    }
    else
    {
        rho1 = C_R(F_C0(f,t),THREAD_T0(t));
        rho2 = C_R(F_C1(f,t),THREAD_T1(t));
        rho = (rho1+rho2)/2;
    }
    return F_FLUX(f,t)/rho;
}
NOMENCLATURE

\( k_B \) Boltzmann constant

\( d \) Bubble diameter

\((v^T_k)_BI\) Bubble induced eddy viscosity

\((\theta^T_k)_BI\) Bubble induced turbulence stress

\( L_c \) Characteristic length of the flow

\( \Delta \mu \) Chemical potential of the dissolution process

\( \bar{\rho}_k = \rho_k \) Density of phase k

\( C_D \) Drag coefficient

\( \mu \) Dynamic viscosity

\( (v^T_k)_SI \) Eddy viscosity

\( \Delta H \) Enthalpy change

\( \Delta S \) Entropy change

\( E_o = \frac{g \Delta \rho d^2}{\sigma} \) Eötvös number

\( d_{eq} \) Equivalent diameter of bubble

\( r_{eq} \) Equivalent radius of bubble

\( u'_k \) Fluctuation component of \( u_k \)

\( g \) Gravitational acceleration

\( l \) Grayscale value

\( H \) Henry’s coefficient

\( R \) Ideal gas constant

\( a_i \) Interfacial area concentration
\( \nu \)  
Kinematic viscosity

\( K_{k,i} \)  
Mass concentration equilibrium ratio for species \( i \) in phase \( k \)

\( C_{k,i} \)  
Mass concentration of species \( i \) in phase \( k \)

\( C_{g,i} \)  
Mass concentration of species \( i \) in the gas phase

\( C_{l,i} \)  
Mass concentration of species \( i \) in the liquid phase

\( C_{m,i} \)  
Mass concentration of species \( i \) in the mixture level

\( Y_{k,i} \)  
Mass fraction of species \( i \) in phase \( k \)

\( k_k \)  
Mass transfer coefficient on phase \( k \) side

\( \Gamma_{l,i} \)  
Mass transfer of species \( i \) source term to phase \( k \)

\( \Gamma_k \)  
Mass transfer source term to phase \( k \)

\( X_{k,i}^e \)  
Molar concentration of species \( i \) in phase \( k \) at equilibrium

\( D_{k,i} \)  
Molecular diffusivity for species \( i \) in phase \( k \)

\( D_{k,i} \)  
Molecular diffusivity of species \( i \) in phase \( k \)

\( p_g^i \)  
Partial pressure of species \( i \) in the gas phase

\( \phi_k \)  
Phase and density weighted time averaged quantity \( \phi \) of phase \( k \)

\( \xi_k \)  
Phase density function

\( \mu_m \)  
Phase mixture viscosity

\( \bar{F}_k \)  
Phase weighted time averaged quantity \( F \) of phase \( k \)

\( P \)  
Pressure

\( \mathbf{u}_r \)  
Relative velocity between dispersed phase and continuous phase

\( Re_L = \frac{U_l L_c}{\nu} \)  
Reynold number for channel flow

\( Re = \frac{\rho d |\mathbf{u}_r|}{\mu} \)  
Reynold number for phase interaction
\[ Sc = \frac{\nu}{D} \] Schmidt number

\[ (\mathbf{6}_k^T)_{SI} \] Shear induced turbulence stress

\[ Sh_L = \frac{kL_c}{D} \] Sherwood number for channel flow

\[ Sh_d = \frac{kd}{D} \] Sherwood number for phase interaction

\[ \overline{M}_m \] Source term for interfacial forces for mixture

\[ \overline{M}_k \] Source term for interfacial forces on phase \( k \)

\( U_g \) Superficial gas velocity

\( U_l \) Superficial liquid velocity

\( \sigma \) Surface tension

\( T \) Temperature

\( \bar{F} \) Time averaged quantity \( F \)

\( D^k \) Turbulent diffusivity for species transport

\( \epsilon \) Turbulent dissipation rate

\( k^t \) Turbulent kinetic energy

\[ Sc_t = \frac{\nu^t}{D^t} \] Turbulent Schmidt number

\[ \mathbf{\omega}_k^T \] Turbulent stress tensor of phase \( k \)

\( \mathbf{u}_k \) Velocity of phase \( k \)

\[ N_\mu = \frac{\mu_c}{\left( \rho_c \sigma \sqrt{\frac{\sigma}{gD\rho}} \right)^{\frac{1}{2}}} \] Viscosity number

\[ \mathbf{\omega}_k \] Viscous stress tensor

\( \alpha_k \) Void fraction of \( k \) phase

\( \alpha^v \) Volume averaged void fraction
\dot{V} \quad \text{Volumetric flow rate}