

Remote Research Internship

Hydrogen Absorption/Desorption in Aqueous Medium

(Sep '24 - Mar'25)

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1 Nomenclature

Nomenclature

List of symbols

T	Absolute temperature (K)
P	Total pressure (bar): $P = P_{H_2} + P_{H_2O}$
R	Universal gas constant (83.14 bar·cm ³ /mol/K)
y_i	Mole fraction of component i in the vapor phase
x_i	Mole fraction of component i in the liquid phase
m_i	Molality of component i in the liquid phase (mol/kg)
\overline{V}	The partial molar volume(cm ³ / mol)
Greek letters	
α	Activity
φ	Fugacity coefficient
γ	Activity coefficient
μ	Chemical potential
λ	Interaction parameter
ξ	Interaction parameter
Subscripts	
a	Anion
c	Cation
Superscripts	
v	Vapor
l	Liquid
(0)	Standard state

Figure 1: Nomenclature

2 Introduction

Nuclear Power Plants are integral towards electricity generation with minimal green-house emissions. They facilitate the aim towards achieving our target of net-zero carbon emissions. In recent decades, nuclear energy has contributed significantly to the global electricity mix, supplying about 10 per cent of the world's total electricity demand [1]. Nuclear reactors operate primarily through controlled fission reactions, where the splitting of heavy atomic nuclei, such as uranium-235 or plutonium-239, releases a substantial amount of thermal energy. Subsequently, this heat is utilized to produce steam, which drives turbines connected to electric generators. However, the result of the nuclear reaction process are several by-products which need to be managed effectively to ensure safety and proper storage of the harmful products. One of the harmful byproducts of this process is the emission of H_2 gas.

Hydrogen emission in nuclear plants primarily arises from radiolysis of water, a process where ionizing radiation breaks down water molecules into hydrogen (H) and oxygen (O) [2]. Additionally, corrosion of zirconium-based fuel cladding in water-cooled reactors, such as pressurized water reactors (PWRs) and boiling water reactors (BWRs), leads to the production of hydrogen as a byproduct through the reaction.

The accumulation of hydrogen within the reactor containment system poses safety concerns, particularly due to the risk of hydrogen explosions, as witnessed in past nuclear incidents like the Fukushima Daiichi accident [3]. Thus, we need to devise methodologies and techniques through which we can store and transport the emitted Hydrogen gas safely. One such method is to transport the gas through pipelines by absorbing them into Aqueous H_2O .

Through this computational study, we seek to understand the dissolution of Hydrogen gas in Aqueous medium under a fixed volume container space. Understanding the absorption and desorption dynamics of hydrogen under varying pressure conditions is crucial for optimizing storage techniques and mitigating hydrogen losses. This study employs ANSYS Fluent software to study the phenomenon using Computational Fluid Dynamics with the help of Multiphase Models and User-Defined mass transfer rates.

By systematically investigating the interaction between hydrogen and aqueous media, this research aims to contribute to the broader discourse on hydrogen management in nuclear energy systems. The findings may also have implications for hydrogen utilization in clean energy technologies, particularly in the context of hydrogen economy development and nuclear-assisted hydrogen production [4].

2.1 Progress Actions

The project was broken down into various segments and sub-tasks to build the model for simulating the absorption process for the H_2 gas.

- One of the first step of the project was to review the current research literature for understanding about cavitation theories and Bubble Nucleation through gas cavities.
- The next step involved creating a comprehensive Gantt Chart to plan the complete research-study flow of work.

- Understanding the phases and species being used to model the problem and learning about CFD models and Multiphase models which are appropriate for the objective study.
 - Learning relevant software tools for implementing the planned models.
 - Compilation of results for conclusive remarks.
-

3 Literature Review

The literature review was based on compiling the experimental data about the hydrogen solubility rates in water at various temperatures and pressures. The goal of the compilation was to understand the trends of solubility with respect to changes in Pressure and Temperature. Along with the experimental data accumulation, papers were reviewed to find out about appropriate modelling software and specific models to be used for the task.

3.1 Experimental Solubility Data

Following are the results obtained for the Solubility trends:

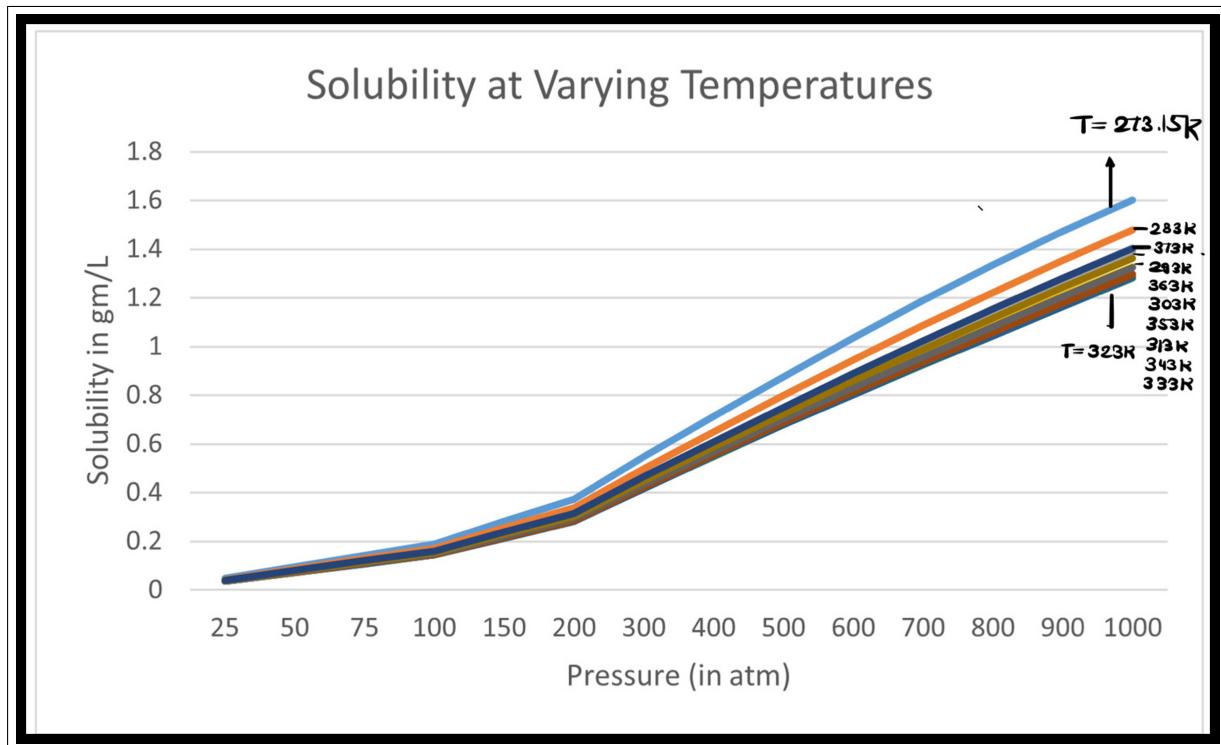


Figure 2: Solubility of H_2 at varying temperatures [8]

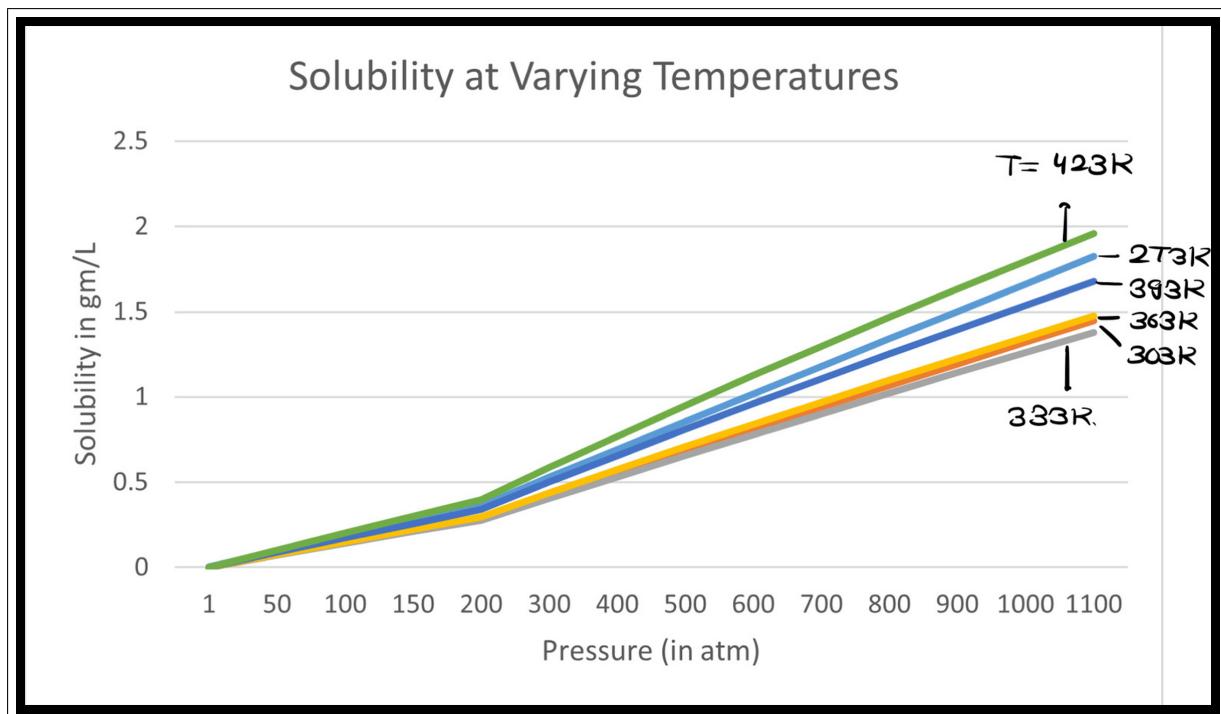


Figure 3: Solubility of H_2 at varying temperatures [9]

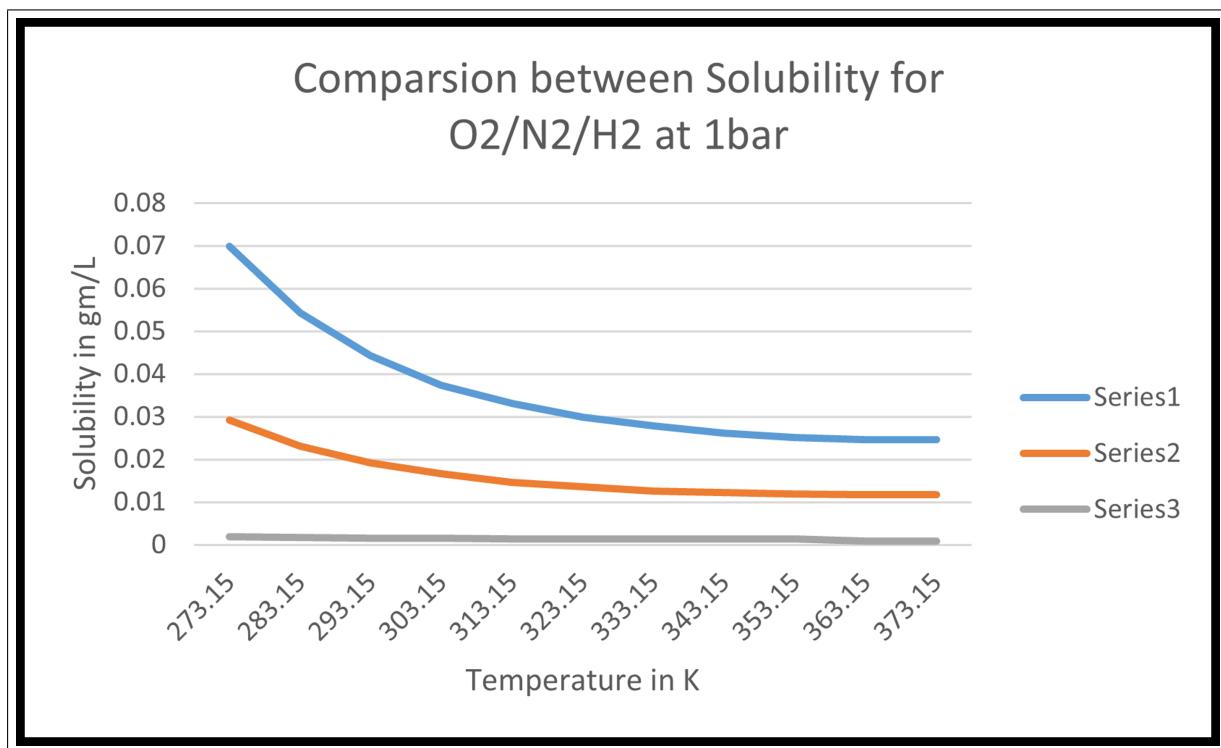


Figure 4: Solubility Comparison between various gases at fixed Pressure and Varying Temperature

3.2 Formula for Solubility Calculation

The literature was also used to understand the formula that could be used to create the mass transfer User-Defined Function for integrating with the CFD Model.

Following is the formula that was obtained through the literature:

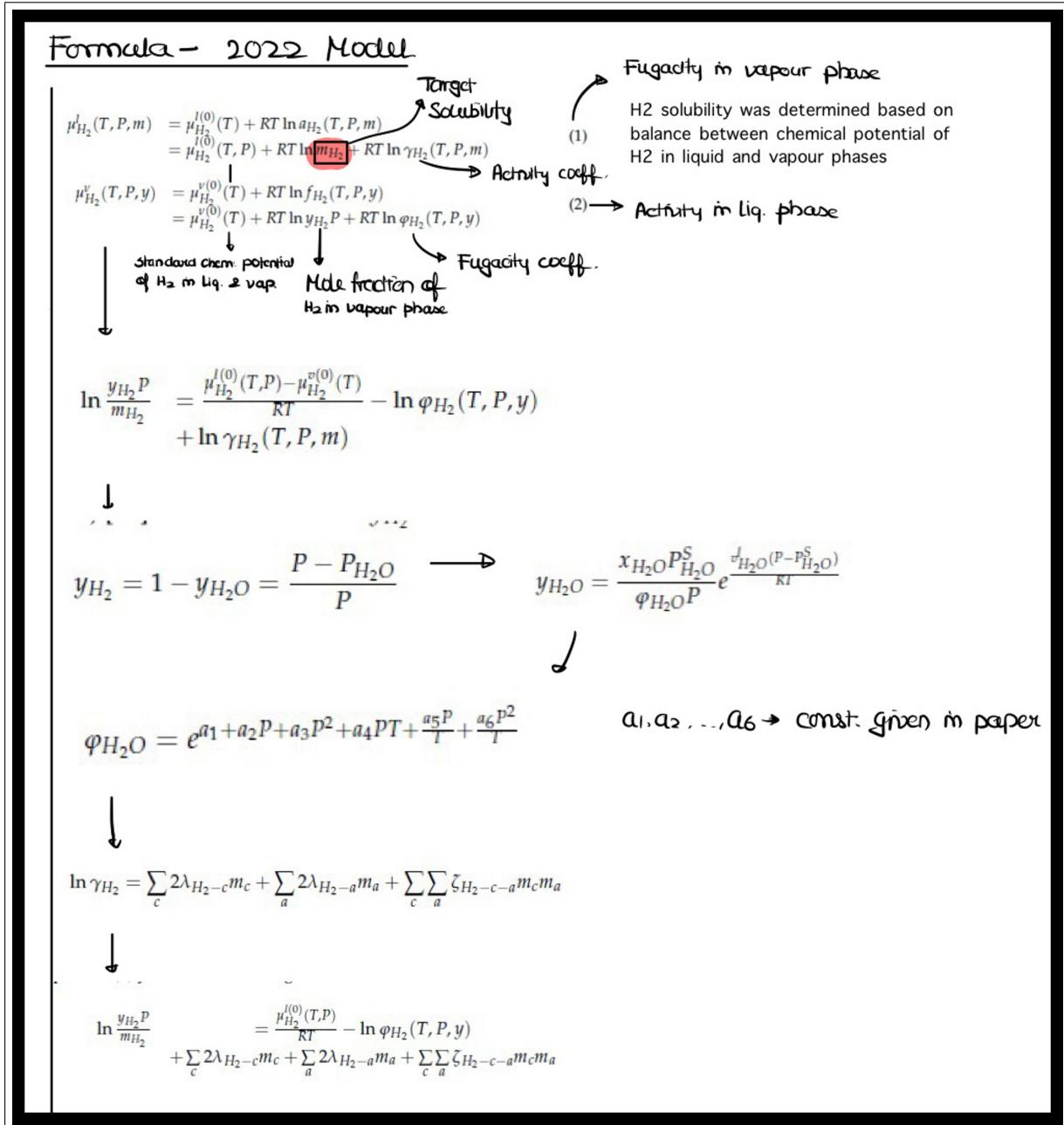


Figure 5: Formula for deriving solubility

3.3 Models for CFD Modelling

The following information was obtained about various models that could be used for setting up the models in the ANSYS Fluent Software

Our objective: Gas-Liquid mass transfer

- VoF Model: Used in case of 2 immiscible liquid flow. Solve momentum equation and track the volume fraction of each fluid throughout the domain.
- Mixture Model: Homogeneous Multiphase flows with very strong coupling and phases moving at the same velocity. Primarily, used to find properties such as viscosity. But concentration of phases can be determined through Volume fraction and mass fraction.
- Eulerian Model: Modelling of multiple separate, interacting phases. Pressure is same for all phases at a given point in the domain. Momentum and continuity equations are solved for each phase.
- Wet Steam Model: Used for rapid expansion of wet steam. The flow mixture is modeled using the compressible Navier-Stokes equations and additional transport equations.
- Multiphase Mass Transfer Model: UDF Model: We can input models for different types of processes In UDF, Fluent automatically adds source contribution to all relevant momentum and scalar equations. UDFs for user-defined sources: When inputting source terms (Mass Source/Energy Source) directly into momentum, energy, and scalar equations.

Through the study about various models, it was suggested that Multiphase Mass Transfer and the Volume of Fluid Model was suitable to be used for the problem modelling.

Following literature was read to gain a brief overview about different CFD approaches to setting up models and solving CFD problems[5], [6], [7].

4 ANSYS Fluent CFD Model

Through the course of the research project, knowledge was gained about:

- Learning the CFD Solver Software ANSYS Fluent
- Applying mass transfer model to basic mass transfer problems
- Understanding the MACROS involved in User-Defined Function

The following are the details about the ANSYS Fluent Model that has been used for the project.

Model Used: Volume of Fluid Model Number of Eulerian Phases: 2 Species Model: Species Transport Phase 1: H₂(gas) + H₂O(liquid) Phase 2: H₂(gas) Initial Pressure = 101,325 Pa Initial Temperature = 300K

The model was applied to a square geometry in such a way that Initially, Phase 1 was patched to the lower half and Phase 2 was patched to the upper half.

Now, Mass Transfer mechanism was used for mass transfer between the 2 phases. The UDF that was used for the mass transfer was based on the Fortran Code for Equations of State. The Peng-Robinson Equation of State theory was used to construct the UDF. The Linearized Mass Transfer Model has been used while constructing the UDF.

Following are the details for the complete CFD Setup

4.1 Meshing

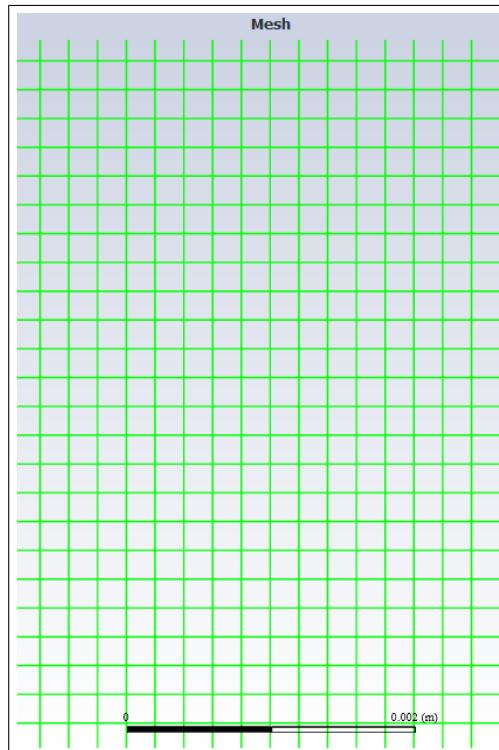


Figure 6: Magnified Mesh for cell visualization

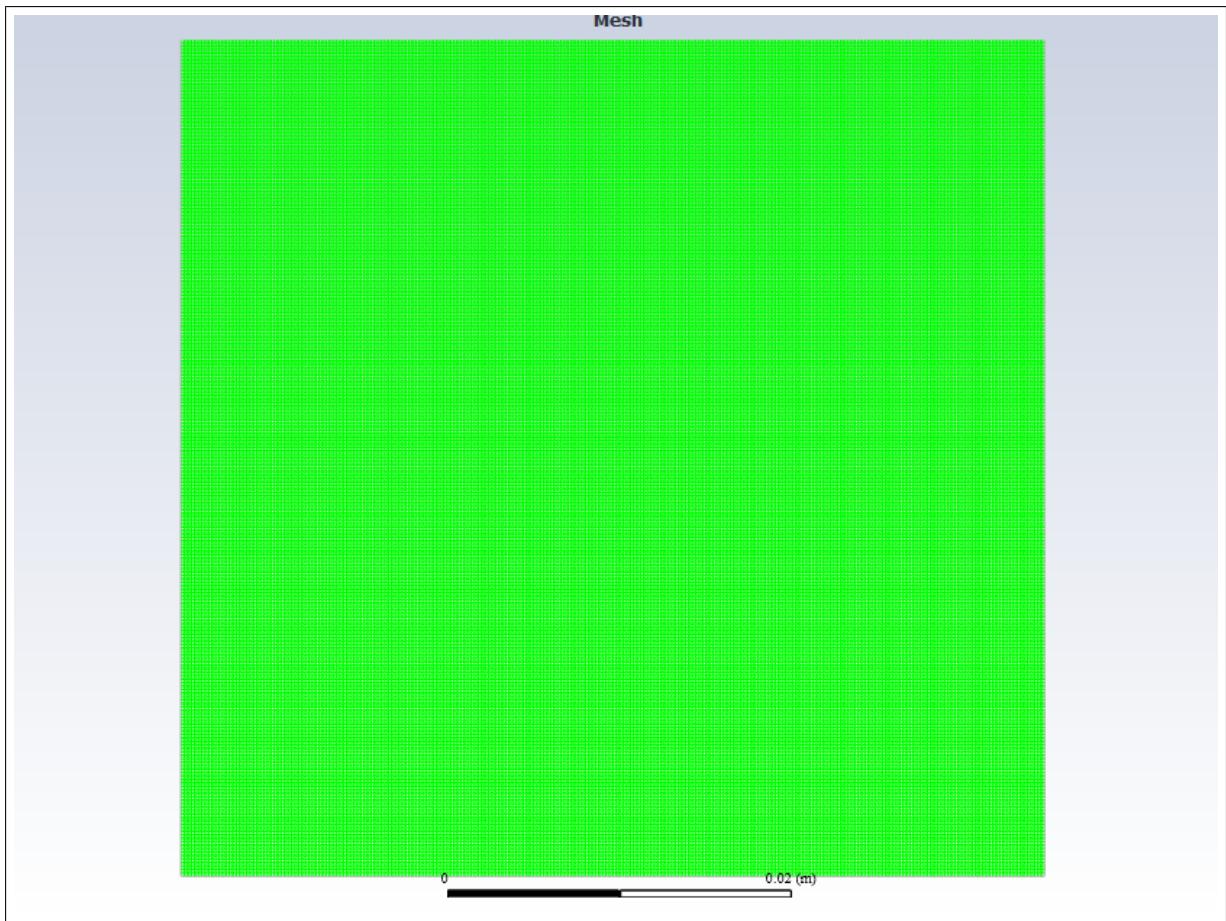


Figure 7: Mesh for the Model

4.2 Multiphase Model

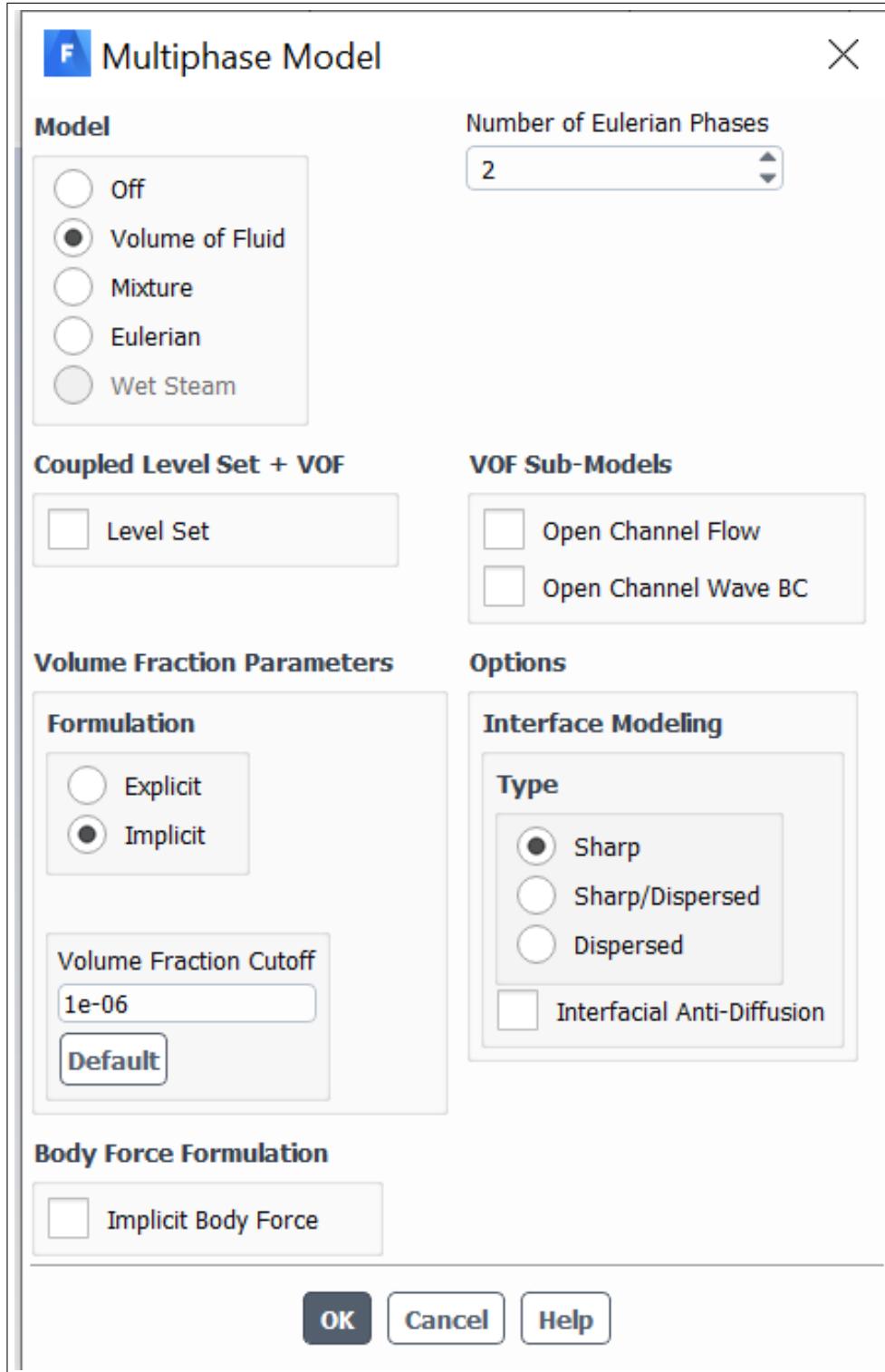


Figure 8: Multiphase Model used for the Setup

4.3 Species Transport Model

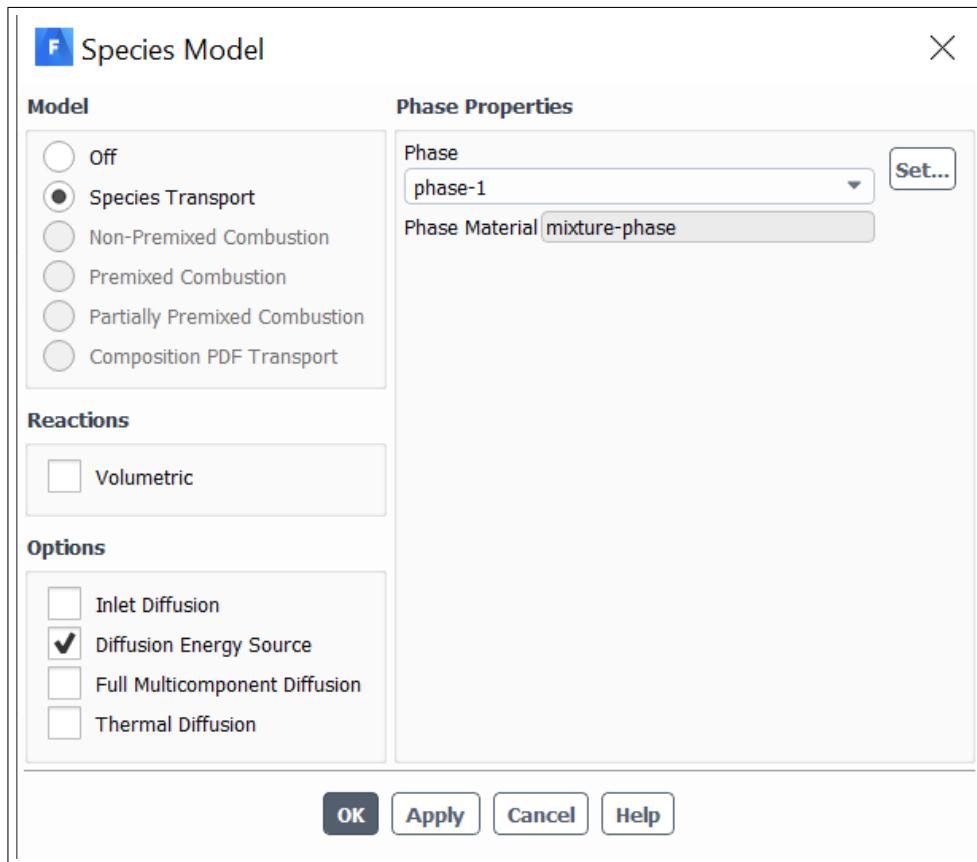


Figure 9: Species Transport Model

4.4 Materials Used

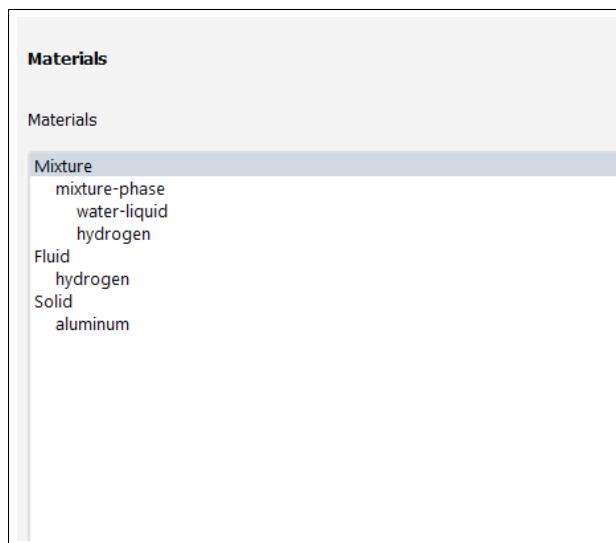


Figure 10: Materials Used

4.5 Initialization

Solution Initialization

Initialization Methods

Hybrid Initialization
 Standard Initialization

Compute from

▼

Reference Frame

Relative to Cell Zone
 Absolute

Initial Values

Gauge Pressure (pascal)
0

X Velocity (m/s)
0

Y Velocity (m/s)
0

Temperature (k)
300

phase-1 h2
0

phase-2 Volume Fraction
0.5

Initialize Reset Patch...
Reset DPM Sources Reset Statistics

4.6 User-Defined Function

The UDF used for the model is given below:

```
#include "udf.h"

#define R_GAS 8314.4621      // J/kmol·K
#define TC_H2 33.15          // K
#define PC_H2 13.0e5          // Pa
#define OMEGA_H2 -0.215
#define HENRY_H2 7.12e6        // Pa·m³/mol
#define M_PI 3.14159265358979323846

// Cubic equation solver (C89-compliant)
int solve_cubic(double a3, double a2, double a1, double a0, double roots[3]) {
    double Q, R, Q3, R2, theta, A, B;
    int i;

    Q = (3 * a1 - a2 * a2) / 9.0;
    R = (9 * a2 * a1 - 27 * a0 - 2 * a2 * a2 * a2) / 54.0;
    Q3 = Q * Q * Q;
    R2 = R * R;

    if (R2 < Q3) {
        theta = acos(R / sqrt(Q3));
        roots[0] = -2 * sqrt(Q) * cos(theta / 3.0) - a2 / 3.0;
        roots[1] = -2 * sqrt(Q) * cos((theta + 2 * M_PI) / 3.0) - a2 / 3.0;
        roots[2] = -2 * sqrt(Q) * cos((theta - 2 * M_PI) / 3.0) - a2 / 3.0;
        return 3;
    }
    else {
        double sign_R = (R >= 0) ? 1.0 : -1.0;
        A = -sign_R * pow(fabs(R) + sqrt(R2 - Q3), 1.0 / 3.0);
        B = (A != 0) ? Q / A : 0;
        roots[0] = (A + B) - a2 / 3.0;
        return 1;
    }
}
```

Figure 12: Cubic Equation Solver

```

real compute_phi(real P, real T, real y_H2) {
    real kappa, alpha, a, b, A, B, Z, sqrt2, term, ln_phi;
    double coeff[4];
    double roots[3];
    int num_roots, i;

    kappa = 0.37464 + 1.54226 * OMEGA_H2 - 0.26992 * OMEGA_H2 * OMEGA_H2;
    alpha = pow(1 + kappa * (1 - sqrt(T / TC_H2)), 2);
    a = 0.45724 * (R_GAS * R_GAS * TC_H2 * TC_H2) / PC_H2 * alpha;
    b = 0.07780 * R_GAS * TC_H2 / PC_H2;

    A = (a * P) / (R_GAS * R_GAS * T * T);
    B = (b * P) / (R_GAS * T);

    coeff[0] = -(A * B - B * B - B * B * B);
    coeff[1] = A - 3 * B * B - 2 * B;
    coeff[2] = -(1 - B);
    coeff[3] = 1.0;

    num_roots = solve_cubic(coeff[3], coeff[2], coeff[1], coeff[0], roots);

    Z = roots[0];
    for (i = 1; i < num_roots; i++) {
        if (roots[i] > Z) Z = roots[i];
    }

    sqrt2 = sqrt(2.0);
    term = log((Z + (1 + sqrt2) * B) / (Z + (1 - sqrt2) * B));
    ln_phi = (Z - 1) - log(Z - B) - (A / (2 * sqrt2 * B)) * term;
    return exp(ln_phi);
}

```

Figure 13: Fugacity Coefficient Calculator

```

DEFINE_LINEARIZED_MASS_TRANSFER(h2_mass_transfer, cell, thread,
    from_index, from_species_index,
    to_index, to_species_index,
    lin_from, lin_to)
[ real m_transfer = 0.0, K = 1e-5, area_vol = 1e3;
Thread* gas_th = THREAD_SUB_THREAD(thread, to_index);
Thread* liq_th = THREAD_SUB_THREAD(thread, from_index);
real T = C_T(cell, gas_th);
real P_gas = C_P(cell, gas_th);
real y_H2 = C_YI(cell, gas_th, to_species_index);
real phi = compute_phi(P_gas, T, y_H2);
real ff_gas = phi * y_H2 * P_gas;
real mw_H2 = 2.016e-3, mw_H2O = 18.015e-3;
real mass_frac_H2 = C_YI(cell, liq_th, from_species_index);
real x_H2 = (mass_frac_H2 / mw_H2) / (mass_frac_H2 / mw_H2 + (1 - mass_frac_H2) / mw_H2O);
real f_liq = HENRY_H2 * x_H2;

m_transfer = K * area_vol * (ff_gas - f_liq);
*lin_from = -K * area_vol * HENRY_H2 * (1.0 / mw_H2) / pow((mass_frac_H2 / mw_H2 + (1 - mass_frac_H2) / mw_H2O), 2);
*lin_to = K * area_vol * phi * P_gas;

return m_transfer;
]

```

Figure 14: Linearized Mass Transfer Model

5 Results and Analysis

5.1 Qualitative Analysis

The results that have been obtained for the model are as follows:

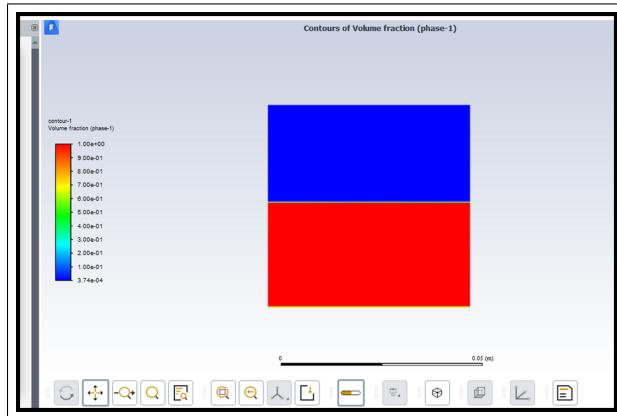


Figure 15: Initialization

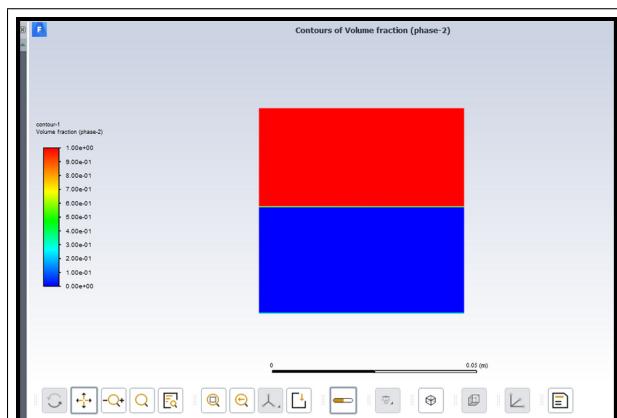


Figure 16: Initialization

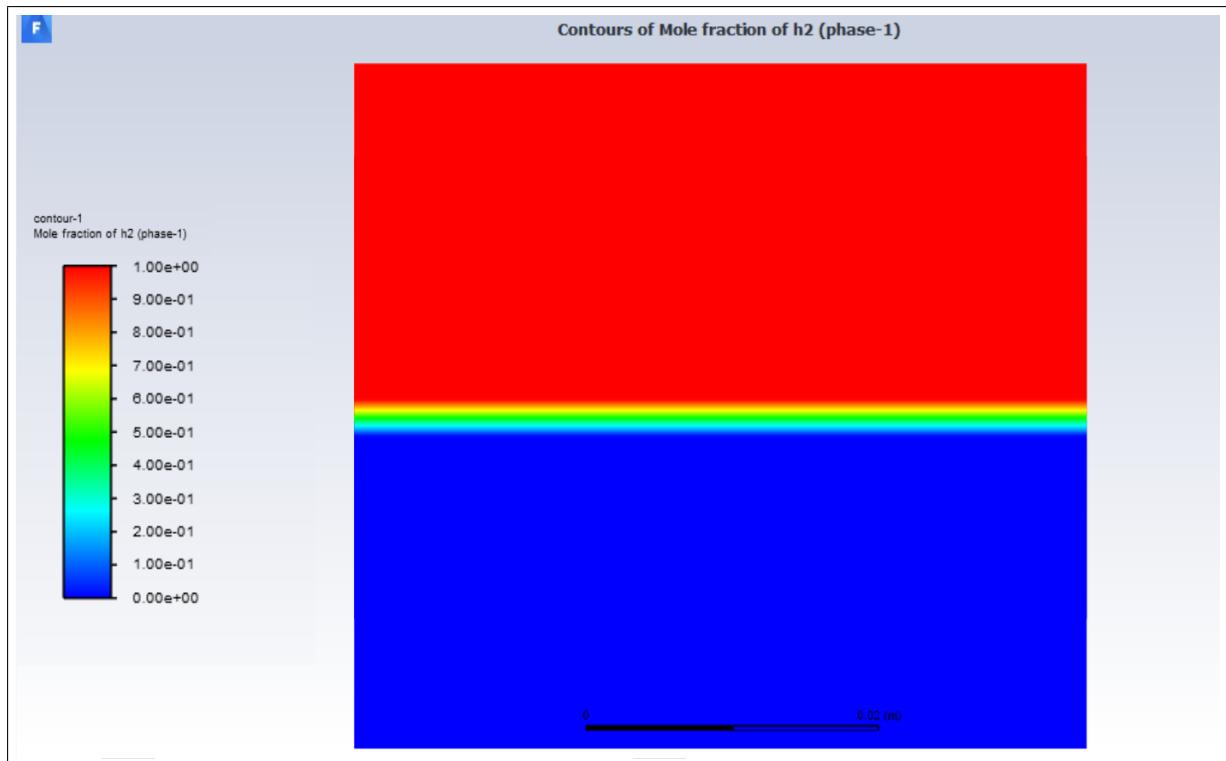


Figure 17: Contour for Mole Fraction of H2

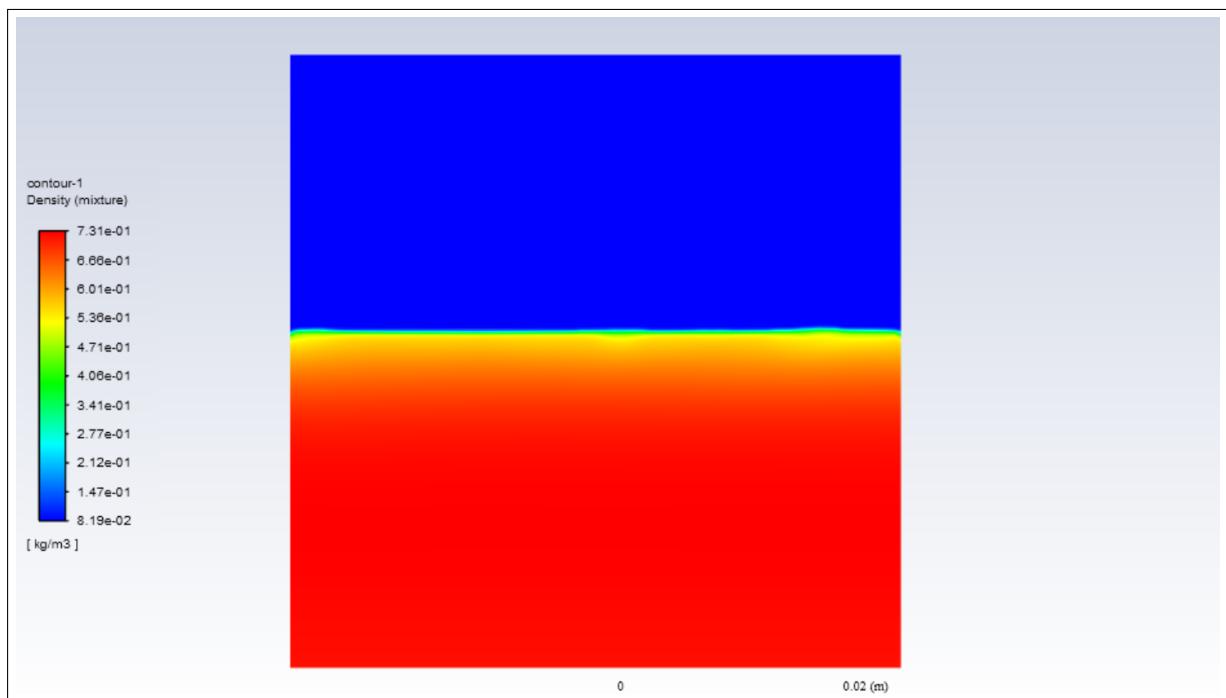


Figure 18: Contour for Density

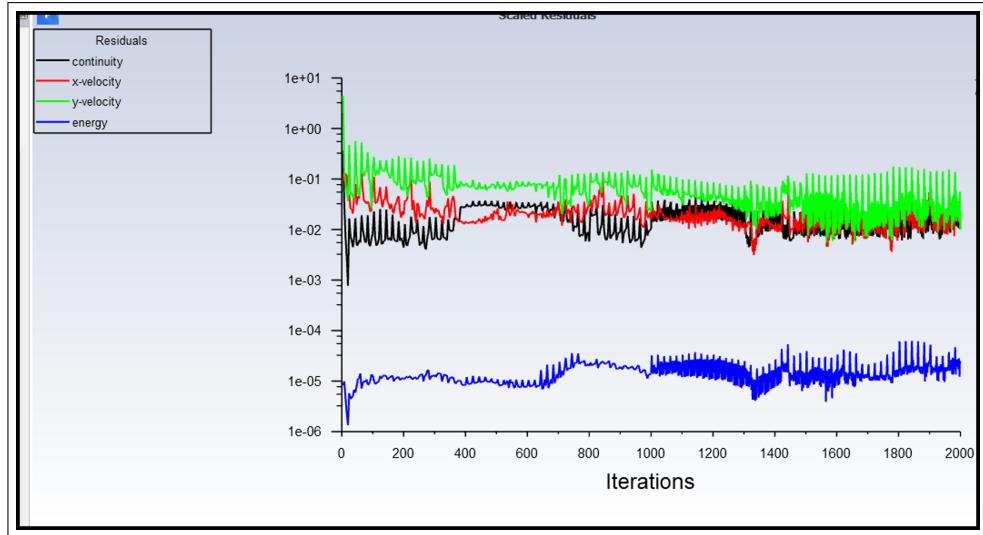


Figure 19: Residual Plot for the Results

5.2 Analysis and Conclusion

Through the contour plots, we can interpret that the H₂ was absorbed into the Aqueous patched part and thus resulted in the increase in mass fraction of H₂ in this lower section of the container. Moreover, it was visibly indicated using the Density contour. The density was higher for the lower section of the geometry and gradually decreased till the middle line indicating the presence of lighter molecules in the geometry. Moreover, it remained constant for the upper half, indicating that it contained a purely gaseous phase of H₂ gas. The value for the density of the upper half was considerably lower than the bottom half.

We can obtain the result for the mass fraction of H₂ with respect to change in the vertical direction as follows:

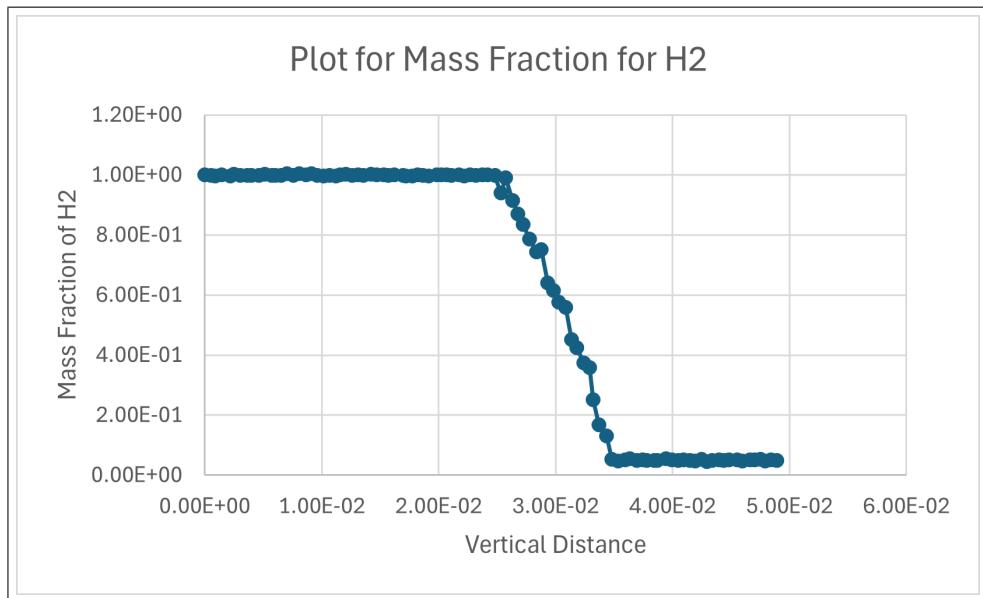


Figure 20: Mass Fraction of H₂ along vertical

6 Proposed Gantt Chart

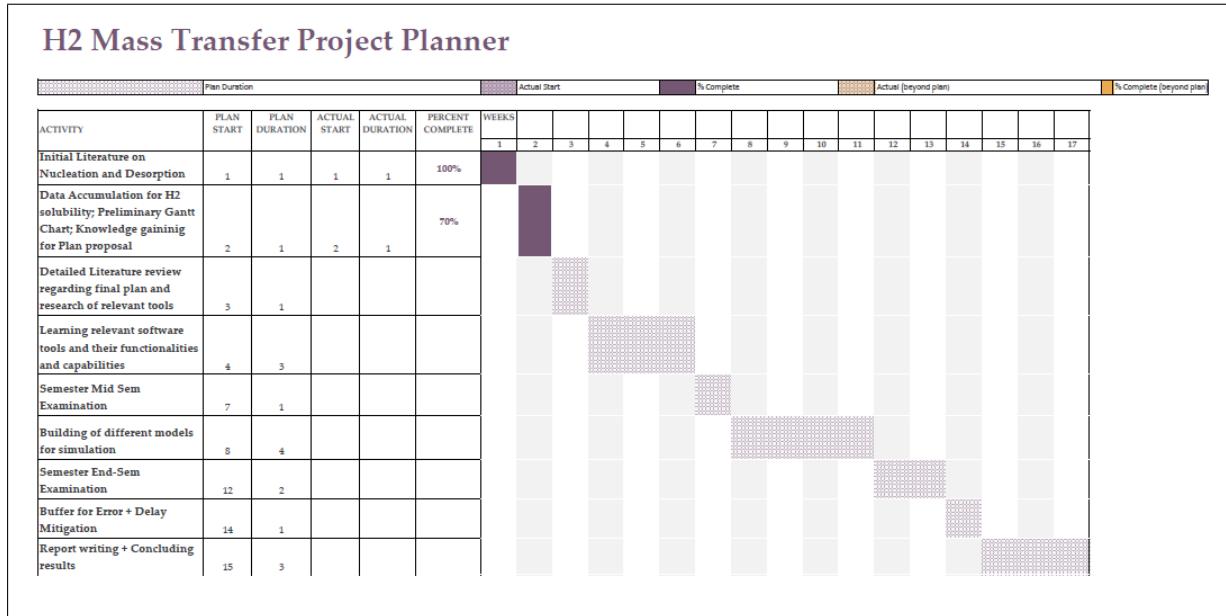


Figure 21: Gantt Chart proposed for the project

7 Acknowledgement

I would like to express my heartfelt gratitude to Dr. Junfeng Yang for his continued support and guidance through the project. I would also like to thank the Dr. Yang and University of Leeds for providing the opportunity to learn and contribute to research in Computational Fluid Dynamics.

8 References

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