Progress Report on Model Development for the Transport of Aerosol through Microchannels

Spent Fuel and Waste Disposition

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SUMMARY

This report summarizes the progress in the development of a phenomenological model of aerosol transport, deposition, and plugging through microchannels. The purpose of this effort is to introduce to a user community—involving researchers, regulators, and industry—a generic, reliable numerical model for the prediction of aerosol transport while accounting for potential deposition and plugging of the leak paths. In that regard, a graphical user interface (GUI) was generated by integrating the individual MATLAB scripts that make up the model and adding additional features to aid the user's general understanding of the problem. This report focuses on the model development and features included. Furthermore, predictions from the GUI are compared with experimental data (Sandia, 2018) for validation. The strength of the GUI is the ability of a user to plug in basic parameters, such as the initial pressure conditions and canister model specifications, to obtain a first-principles approximation of vital information such as the blowdown pressure differential, aerosol penetration, and deposition as a function of time. The user can obtain this without the know-how of the underlying MATLAB scripts, enabling the model to be readily applied by regulators, industry, and shareholders to reduce the uncertainty in off-site radiological consequence evaluations.

The report also lists future tasks under this work scope. This includes ongoing efforts to include additional crack geometries (divergent and divergent–convergent slots) to the model; GUI development and improvement based on feedback from users; and integration of aerosol source term data from sibling pin tests into the model to simulate more realistic canister stress corrosion cracking–induced aerosol release scenarios.

This report documents work performed under the Spent Fuel and Waste Science and Technology program for the US Department of Energy (DOE) Office of Nuclear Energy (NE). This work was performed to fulfill Level 3 Milestone M3SF-22OR010207012, "FY2022 Progress Report," within work package SF-22OR01020701.

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This report was developed with significant contributions, expert input, and guidance from Sam Durbin at Sandia National Laboratories and Rose Montgomery at Oak Ridge National Laboratory.

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ABBREVIATIONS

DOE	US Department of Energy
GDE	general dynamic equation
GUI	Graphical User Interface
ORNL	Oak Ridge National Laboratory
SCC	stress corrosion cracking
SNL	Sandia National Laboratories

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PROGRESS REPORT ON MODEL DEVELOPMENT FOR THE TRANSPORT OF AEROSOL THROUGH MICROCHANNELS

1. INTRODUCTION

Understanding the aerosol transport and depressurization process of a postulated initiating event in a spent fuel canister is important for determining potential consequences as a result of stress corrosion cracking (SCC) formations (Durbin 2021; Lanza 2021; Chatzidakis 2021; Philips 2021; Chu 2017). Currently, estimations on aerosol penetration and plugging through a crack in a spent fuel canister are based mostly on experimental correlations or empirical models based on a limited dataset (Chatzidakis 2018). To expand the understanding of the processes ongoing during the event and to extend the ability to predict released quantities, a phenomenological model is under development. The model includes aerosol transport and retention in leak paths as well as accounts for plugging formation (Chatzidakis 2019; Chatzidakis and Scaglione 2019). This model could be used to improve the accuracy of consequence assessments, reducing the uncertainty of radiological consequence estimations by accounting for the filtering effect of leak path aerosol deposition and plugging in the source term.

The model assumes a one-dimensional flow through a hydraulically equivalent leak path. The description is dynamic, with changing duct geometry due to plugging, and is also mechanistic. An extensive validation exercise (particle diameters: 0.01–10 μm and pressure difference up to 700 kPa) is in progress based on comparisons with experimental measurements and empirical correlations. This model could be used to improve the accuracy of consequence assessments, reducing the uncertainty of radiological consequence estimations by accounting for the filtering effect of leak path aerosol deposition and plugging of the source term.

This report focuses on a graphical user interface (GUI) based on the model described above, which will allow stakeholders to quickly perform calculations related to aerosol transport and depressurization of spent fuel canisters. By expediting these calculations, we can potentially accelerate experimental design and understand important driving mechanisms without the need to access the source code. Even though the original body of code will be accessible and editable by all users, the proposed GUI will allow those without a strong knowledge of the specifics of the internal code to make temporary changes to inputs and perform estimations easily, without needing to search through the code and edit variable values directly. Simplifying access and making the process of inputting parameters much faster and more user-friendly will allow stakeholders to make informed decisions and better understand a multidimensional problem.

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2. BACKGROUND

The physical situation being modeled in this project involves a stainless-steel spent fuel canister with an SCC with a size on the order of 1–100 microns that has formed in the wall. Because spent fuel canisters are eventually pressurized, the gas inside of the canister will slowly leak out of the newly formed opening as a result of the pressure difference. As the gas travels through the crack, it may carry some solid aerosolized particles of spent fuel along with it. The primary goal is to determine how much radioactive material would leak out of a canister as it depressurizes if this kind of crack were to form.

Modeling aerosol transport, retention, and plugging involves the thermal hydraulic behavior of the canister and the microchannel/crack, as well as the geometric characteristics of the microchannel/crack and the aerosol features. The minimum set of parameters that must be identified includes the following:

- **Thermal hydraulics:** Within this group, the most important parameters are the pressure inside the canister, gas composition, pressure drop, and the wall temperature along the crack.
- **Crack geometry:** The parameters to be considered are mainly the crack path and its hydraulic diameter; information on the crack section shape and curvature is also very important.
- **Aerosol features:** Aerosol average composition, concentration, and size distribution are the reference parameters; others, regarding aerosol morphology, can be taken only with large uncertainties.

A phenomenological model based on the aerosol general dynamic equation (GDE) is currently under development with the goal of accurately simulating aerosol transport and deposition through microchannels. The model can simulate rough or smooth surfaces, irregular geometries, and unsteady flow. Four main deposition mechanisms—gravitational, Brownian diffusion, turbulent diffusion, and eddy impaction—have been included. Laminar, transition, and turbulent gas flow regimes have also been included in the model. The proposed model is being tested and compared with experimental and theoretical work to evaluate its validity and identify its range of applicability.

A compilation of available data in support of model development, including (1) canister characteristics, such as pressure, temperature, heat load, and environmental conditions; (2) particle size distribution within the canister for various scenarios; (3) crack characteristics, such as size, opening, and roughness; and (4) past models and experiments involving aerosol transport, which can be found in the work by Chatzidakis (2018a).

Because of the many parameters and features involved, high-fidelity modeling of aerosol is a slow and cumbersome process. In such scenarios, a simplified mechanistic model can provide reasonable estimates with less computational cost. A wide range of particle size, distribution, pressure differential, flow regimes, and microchannel dimensions can be simulated using the current model. The model can predict pressure change over time due to depressurization (transient state) and particle deposition within a large vessel, such as a canister, before, during, and following depressurization. A summary of the model characteristics is presented in Table 1.

Table 1. Summary of aerosol model characteristics.

Particle diameter	0.01 μm–10 μm
Particle distribution	Monodisperse / Polydisperse
Pressure	4 Pa–700 kPa
Microchannel dimensions	5 μm–1 mm
Coagulation	Yes
Deposition mechanisms	Brownian, Gravitational, Turbulent, Inertial, Thermophoresis
Plugging	Yes
Steady-state	Yes
Transient state	Yes
Flow regimes	Laminar, Transition, Turbulent, Choked
Depressurization	Yes
Fluid	Air, He

3. OVERVIEW OF THE GUI

The model has been described in previous progress reports (Chatzidakis 2018b; Chatzidakis 2019). Overall, the model can predict important quantities such as plug mass, gas passed, plugging time, plug profile, and aerosol penetration or retained fraction.

The GUI is based on a series of MATLAB scripts containing the aerosol transport code. Figure 1 shows a flowchart of the code and the different underlying steps that are involved in predicting the penetration, retention, and modifications to the crack as a result of depressurization. The scripts currently perform these calculations for two specific crack geometries—cylindrical and rectangular—to give specific outputs.

For the GUI, these scripts were converted to functions so that they could be called upon to perform their respective calculations. Then, the GUI was constructed in MATLAB App Developer. The code of the GUI served the role of an executive function that would call on the necessary premade functions based on the user's inputs. The executive function first chooses the correct function based on the chosen geometry, and then all of the necessary user inputs from the GUI are fed into the function, which calculates the desired outputs and passes these back to the GUI to be displayed. Additionally, separate functions were written for the geometry display tool, which accepts all of the parameters related to the crack geometry and outputs a 3D plot showing the crack.

Because crack formation is a complicated process, many assumptions had to be made for these calculations to be possible and fast. The assumption that introduces the most error is the crack geometry. In real-world scenarios, SCCs have complex geometries, but for this project, the crack was modeled using a cylindrical microchannel or a thin rectangular slot. Other crack geometries will be implemented in the future with the goal of more accurately modeling a naturally formed SCC. Another assumption made for this project was that the aerosolized spent fuel exists evenly dispersed inside the container with a uniform concentration and particle size at the start of the depressurization. A detailed description of the model and assumptions can be found in Chatzidakis (2018).

The code also performs blowdown calculations using a mass flow rate equation that was formed accounting for the conservation of mass and conservation of momentum to retrieve the mass flow rate, time, and pressure change from a leaked cannister (Chatzidakis 2018). The aerosol penetration calculations are performed using a one-dimensional aerosol transport equation, which is solved numerically using the finite difference method. This solution takes into account the following aerosol deposition mechanisms: Brownian diffusion, gravitational settling, inertial settling, and turbulent diffusion. Additionally, the model allows the option of including plugging in the leak path.

The GUI is able to perform calculations quickly, with runtimes that are typically no more than a few seconds for cases within the scope of the calculations, and this complements the GUI's inherent benefit of allowing users to quickly enter data and run the code. There is some instability in the mass flowrate outputs, especially at higher pressures, which has been partially remedied through modifications to the time steps however this instability still appears with some sets of geometry and pressure, and this is still being investigated.

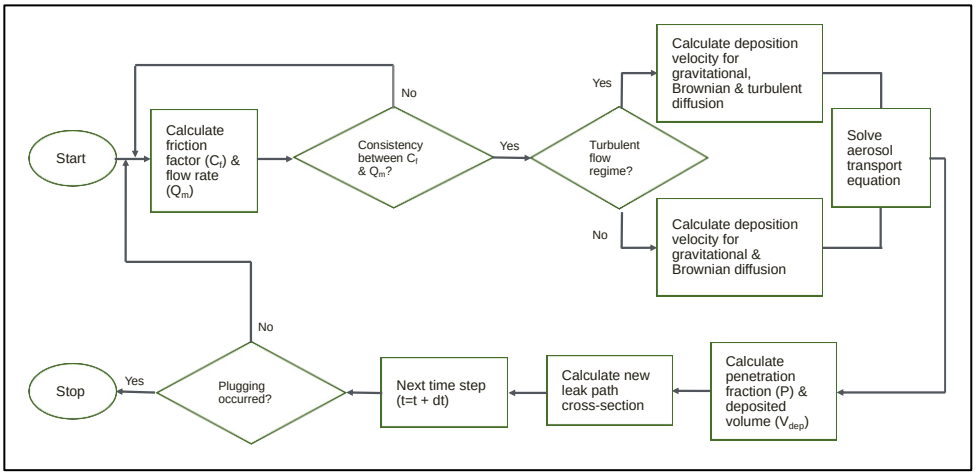

Figure 1. Block diagram for the numerical solution of the aerosol transport equation (Chatzidakis 2018b).

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4. FEATURES PRESENT IN THE GUI AND BENCHMARKING

The GUI allows for a wide range of parameters related to the properties of the canister, the crack geometry, and the properties of the gas and aerosol inside the canister. The various input options (as seen in Figure 2) include:

1. Cannister Parameters
    - Canister Model – This dropdown menu automatically populates the free volume, length, and pressure fields with values taken from one of a few standard spent fuel canisters
    - Free Volume – The volume inside of the canister that can be occupied by gas (canister inner volume – spent fuel volume)
    - Pressure – Pressure inside the canister in Pascals
    - Temperature – Temperature inside the canister in Kelvin
2. Channel Parameters
    - Select type dropdown menu – This dropdown menu switches between a circular capillary geometry and a rectangular channel geometry
    - Length – The wall thickness of the canister in meters; it is the depth of the opening normal to the surface of the canister
    - Opening – This value is either the diameter of the circular opening or the height of the rectangular opening in meters
    - Width – This is the width of the rectangular opening and has no effect on the circular geometry
    - Area – The cross-sectional area of either opening in square meters; this field is automatically populated when new dimensions are entered
    - Hydraulic Diameter – Automatically populated field with the hydraulic diameter; either the diameter value for circular geometry or 4 times the area divided by the perimeter for the rectangular geometry
3. Gas properties
    - Select dropdown menu – This dropdown menu chooses between air and helium, which then prepopulates the gas property entries
    - Rg – Specific gas constant of the chosen gas in J/kgK, this can either be manually entered or prepopulated from the dropdown menu
    - Viscosity – Viscosity of the gas in PaS
    - Gamma – The isentropic expansion factor of the gas (Cp/Cv)
4. Aerosol properties
    - Concentration – Concentration of the aerosol in the canister in kilograms per cubic meter
    - Geometric Particle Diameter – Physical diameter of the aerosol particles, enter in microns
    - Particle density – Density of the material that composes the aerosol particles

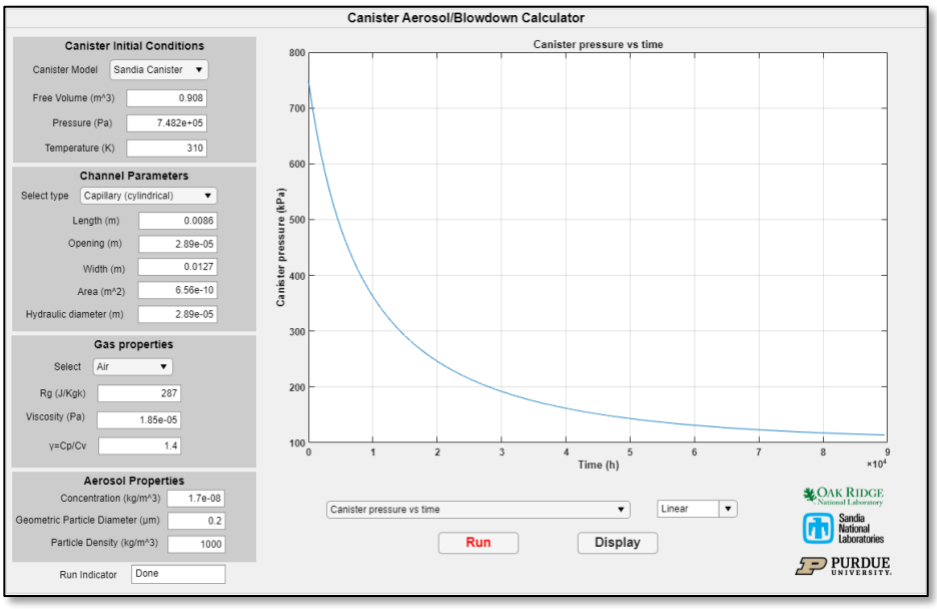

Figure 2. Screenshot of the GUI with all available input options. The GUI is displaying the canister pressure over time as a blowdown occurs.

Thus, the user has the option to modify the crack geometry by changing the canister wall thickness, opening radius, or opening width and length depending on the chosen geometry. The user can also change parameters related to the spent fuel canister, such as starting pressure, temperature, and volume. These fuel canister options come with a dropdown menu of pre-set values matching up with a few common industry canisters (Figure 2). Finally, the properties of the gas and aerosol inside the canister can be changed, with a dropdown menu that populates the inputs with the correct values for helium or air, as well as options for changing the size, concentration, and density of the aerosol particles.

The range of output options available in the GUI can be seen in Figure 3. This includes:

- Mass flowrate vs pressure difference – Plots the mass flowrate in kg/s against the difference in pressure between the canister interior and atmospheric pressure
- Total mass leaked vs time – Plots the cumulative mass leakage against time
- Reynolds number vs time – Plots the Reynolds number of the flow exiting the canister against time
- Mach number vs time – Plots the Mach number of the flow exiting the canister against time
- Penetration fraction vs velocity – Plots the percent of aerosol particles that escape the canister as a function of the velocity
- Deposition fraction vs mass flowrate – Plots the percent of aerosol particles that do not escape the canister as a function of the mass flowrate

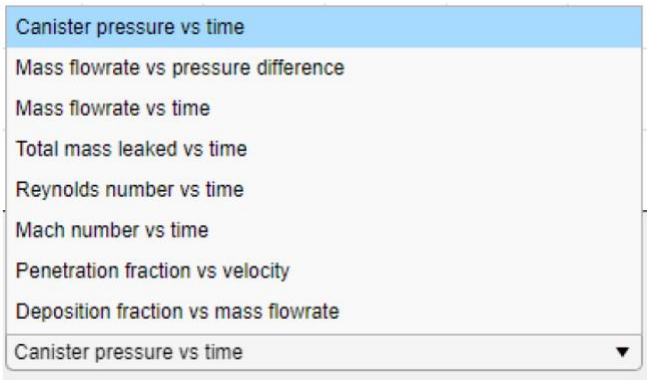

Figure 3. Drop-down menu showing output options.

A second dropdown menu to the right of the output dropdown menu (Figure 2) allows the user to change between linear, semi-log, and log-log axes for the output plots.

The GUI also includes a feature to display the crack geometry. If the user presses the display button shown in Figure 2, then a second window will appear with an interactable 3D graph of the opening geometry with the entered dimensions. An example of the geometry display showing the cylindrical opening is shown in Figure 4.

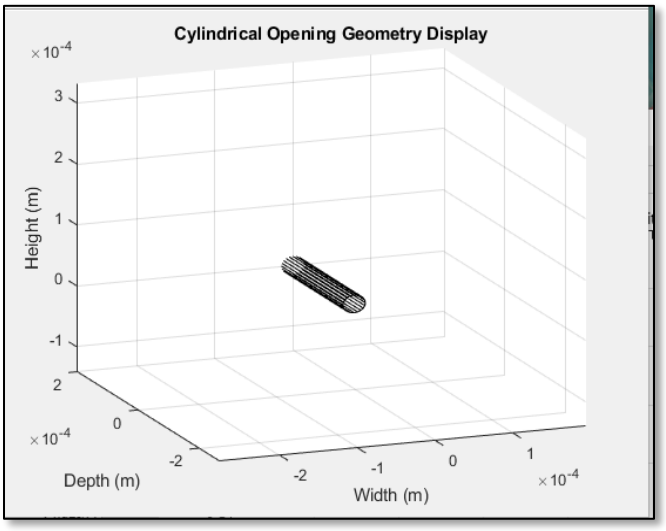

Figure 4. 3D plot of an example cylindrical crack geometry.

Lastly, a "Run" indicator was also added to display the text "Processing..." if the code is still running. Once the code is finished running, the indicator displays the text "Done."

The models used by the GUI were benchmarked against a large set of publicly available experimental data (Chatzidakis and Scaglione 2019). Because the GUI directly calls upon the models developed by ORNL, the GUI itself needed to be validated only for run time and debugging. The estimations from those models were in good agreement with experimental measurements and were able to calculate outputs related to gas blowdown reasonably well. An example of the outputs from the GUI is shown below in Figure 5. The results displayed by the GUI are in blue and are compared to experimental data from Sandia National Laboratories (SNL) in red. The lines diverge as the pressure differential increases, but because this tool is meant for quick estimations of these calculations, this amount of error is deemed acceptable.

The results for the aerosol penetration fraction models the correct shape, with high penetration near the beginning of the blowdown but lower aerosol penetration as the pressure decreases and plugging begins. However, the calculator outputs unreasonably high penetration fractions, and the relationship between penetration fraction and particle size is incorrect. Although the calculator does not accurately determine aerosol transport, it serves as a framework for improved calculation scripts to be called by the GUI to perform the calculations and provide all of the features that have already been built.

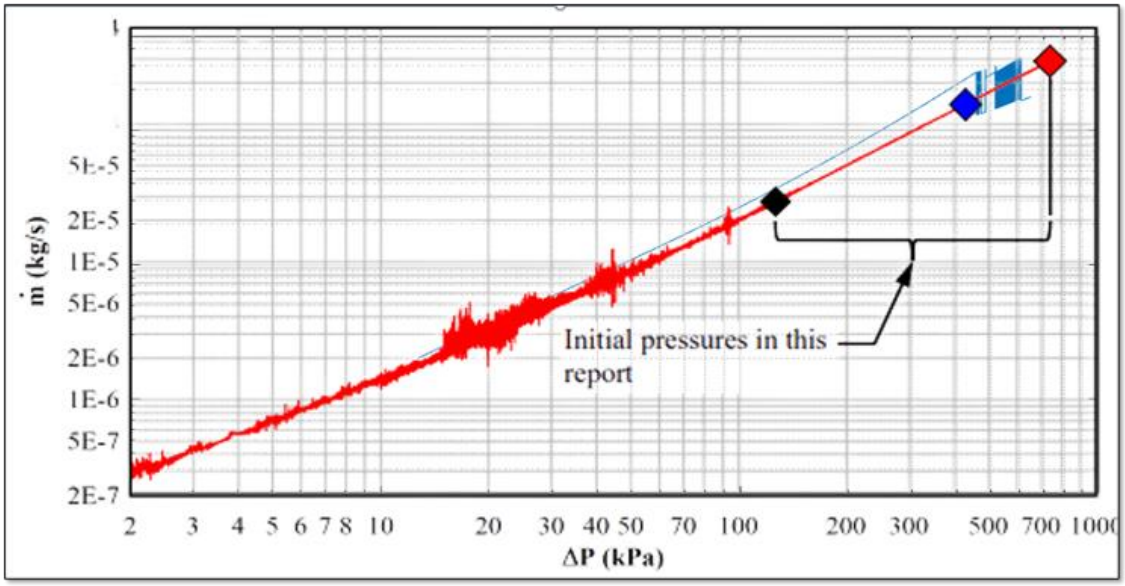

Figure 5. Mass flow rate vs. Pressure Differential graph showing outputs from the GUI (blue) compared to experimental data from Sandia National Labs (red) (Durbin 2018).

To maintain the approximately 1 s run time, a time step was chosen that scales inversely with the area of the crack opening. In the default case, the simulation runs over course of 3.2E+8 s, and the time step used is 7,600 s. As the area of the opening increases, the canister will depressurize faster so a finer resolution timestep may be used without compromising runtime. Additionally, for the rectangular geometry that suffered from instability at higher pressures, the timestep grows with each loop iteration so as to increase the resolution at the beginning of the blowdown when the pressure is highest.

$$\Delta t_{cylindrical} = \frac{5 * 10^{-7}}{Microchannel\ Area\ (m^2)}$$

$$\Delta t_{rectangular} = \frac{5 * 10^{-6}}{Microchannel\ Area\ (m^2)} + (1 * 10^{-5})(Current\ Loop\ Iteration)$$

As shown in Figures 6 and 7, the blowdown time and number of loop iterations follow a similar shape when plotted against the hydraulic diameter of the microchannel. Blowdown time is defined as the time it takes for the pressure in the canister to decrease from the starting value (typically 748 kPa) to 110% of atmospheric pressure. Due to the scaling of the time step, the number of loop iterations begins to flatten out more as the hydraulic diameter increases, which is in line with the goals of the GUI as resolution should not drop too low for larger microchannel openings. For very small microchannels both the number of loop iterations and the runtime increase sharply due to the high blowdown times. However, for

microchannels near the size of the ones tested in the experiments conducted at Sandia, which had a hydraulic diameter of 2.89E-5 m, the runtime and resolution are both sufficient.

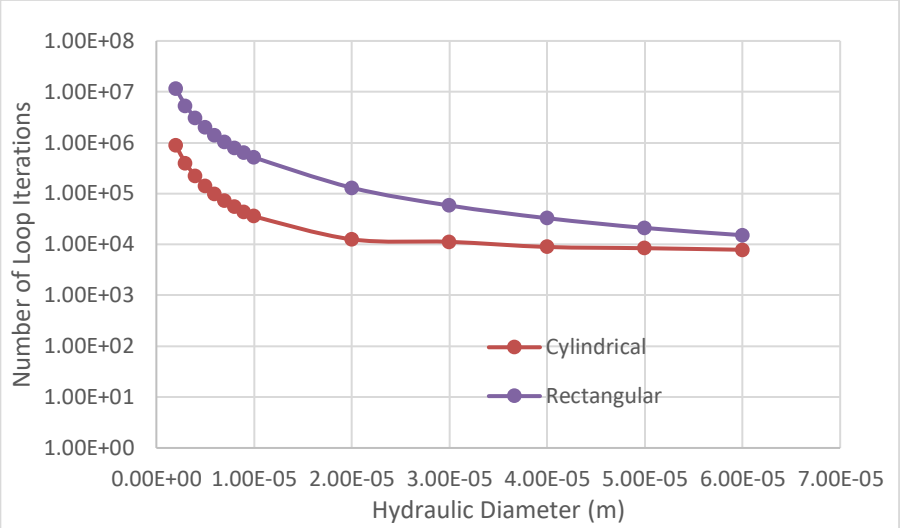

Figure 6. Graph of the number of loop iterations vs. the hydraulic diameter for both cylindrical and rectangular geometries.

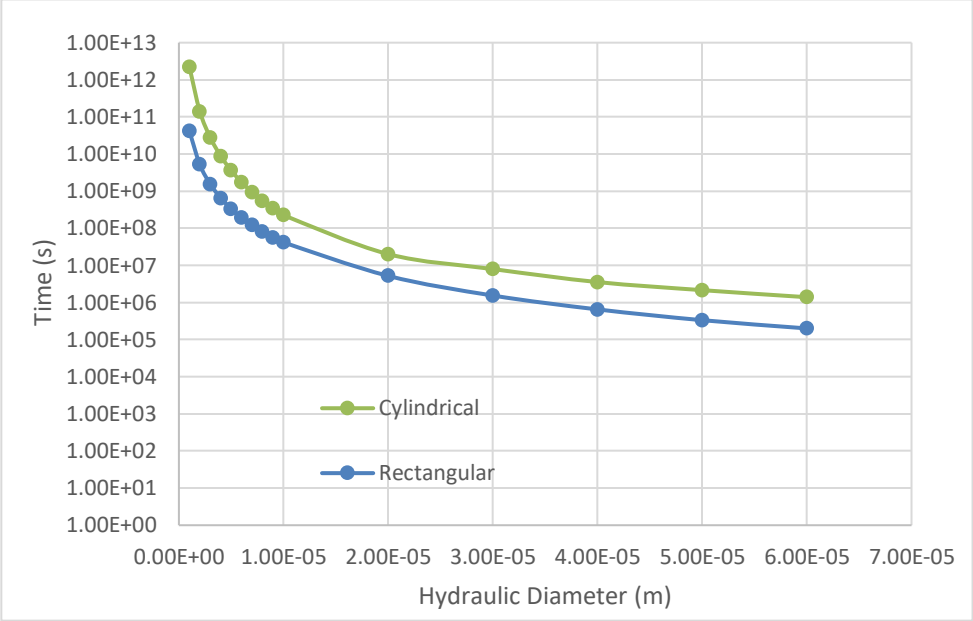

Figure 7. Graph showing the relationship between blowdown time and hydraulic diameter.

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5. SUMMARY AND FUTURE WORKSCOPE

A phenomenological model is currently under development that enables the calculation of aerosol transport and retention in leak paths and also accounts for plugging formation. The model assumes a one-dimensional flow through a hydraulically equivalent leak path. The description is dynamic, with changing duct geometry due to plugging, and it is also mechanistic. An extensive validation exercise (particle diameters: 0.01–10 μm and pressure difference up to 700 kPa) is in progress based on comparisons with experimental measurements and empirical correlations. The model predictions are in approximate agreement with experimental data and are reasonably representative. Qualification testing will be undertaken after the algorithm is developed further and validated.

Further progress in refining the model will require (1) adding additional crack geometries, deposition mechanisms and realistic particle size distributions and (2) additional experimental studies for model validation. It is recommended that future experiments address:

- Flow rates for various pressures to provide insight into flow regimes and develop friction factor correlations in a microchannel.
- Measurements of aerosol concentration in a canister, including polydisperse aerosol, to better understand aerosol reduction processes such as coagulation and deposition as well as for model validation.
- Tests that result in the complete plugging of a microchannel to better understand aerosol plugging conditions and for model validation.

The GUI built in this project performs blowdown calculations quite well, with reasonable agreement between the results from the calculator and the results from models from Oak Ridge National Laboratory (ORNL) as well as the experimental data obtained by SNL. Current work is focused on correcting the errors in calculating the aerosol penetration fraction as well as addressing the instability that tends to appear at higher starting pressures. Future work on the GUI development will involve integrating newer features and modifying the functions as the code is updated. Currently, the model is limited to a flexible 1D mechanistic model that can simulate various aerosol deposition mechanisms, either separately or in combination (Figure 8). This can provide unique insights into the behavior of each mechanism and can help identify the main mechanisms, including importance ranking, that drive deposition and plugging in SCCs. However, further model development and fidelity can be achieved by including:

- additional deposition mechanisms
- additional dimensions (from 1D to 2D)

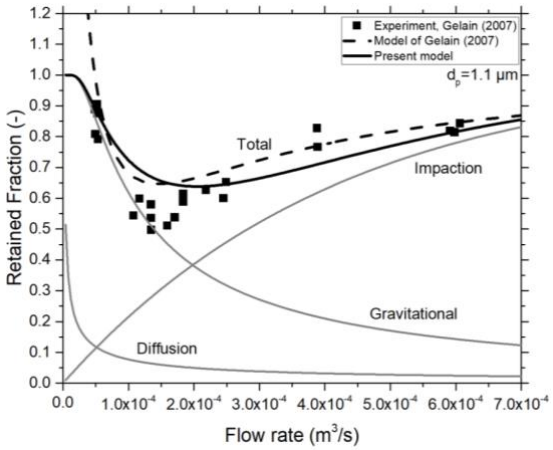

Figure 8. Model results and comparison of aerosol deposition mechanisms with experimental measurements (Chatzidakis, 2019).

The work package and experiments conducted at Sandia National Laboratories are looking to progress toward more realistic crack geometries. This is also a priority in the modeling approach and ORNL is currently working on adding divergent crack geometries into the model (Figure 9). Furthermore, a sensitivity analysis is planned to have different degrees of divergence (crack geometries) and its effect on aerosol diffusion. The final goal is to simulate a realistic crack geometry.

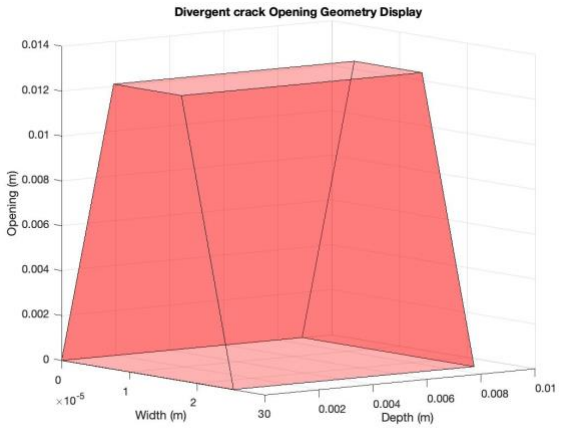

Figure 9. Divergent slot geometry used in the Sandia experiments generated from the GUI 3D plot feature.

In addition to the above, there is a strong interest in converging the sibling pin aerosol collection and characterization study (Montgomery 2020) and the current aerosol diffusion modeling efforts at ORNL. This is aimed to bridge the gap in the aerosol source terms used in the model. Furthermore, the experimental tests will be modified as necessary, based on the parameters required for the model.

Advances in this area hold the promise of improving the accuracy of consequence assessments and reducing the uncertainty of radiological consequence estimations by taking the filtering effect of leak path aerosol deposition and plugging into account the source term.

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