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Facility Modeling Capability Demonstration Summary Report

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1. Executive Summary

A joint effort has been initiated by Los Alamos National Laboratory (LANL), Oak Ridge National Laboratory (ORNL), Savanah River National Laboratory (SRNL), Pacific Northwest National Laboratory (PNNL), sponsored by the National Nuclear Security Administration's (NNSA's) office of Proliferation Detection, to develop and validate a flexible framework for simulating effluents and emissions from spent fuel reprocessing facilities. These effluents and emissions can be measured by various on-site and/or off-site means, and then the inverse problem can ideally be solved through modeling and simulation to estimate characteristics of facility operation such as the nuclear material production rate. The flexible framework called Facility Modeling Toolkit focused on the forward modeling of PUREX reprocessing facility operating conditions from fuel storage and chopping to effluent and emission measurements.

The cornerstone of the flexible framework, which is used to link individual modules representing process operations, is a python-based code referred to as Cortix. Cortix provides the integration architecture for time-dependent communication between modules, with the flexibility to add or eliminate future modules written in most any programming language. This approach is intended to allow the user to add or remove their own modules as needed. Individual modules representing process operations were physics-based with an emphasis on the relationship between nuclear material production and that of the related emission signatures produced. This approach departed somewhat from past nuclear facility simulations that were more interested in nuclear material production than that of effluent and emissions.

Initial modules being developed were focused on that necessary for validation of a nuclear fuel reprocessing simulation. These modules included fuel shear, fuel dissolution, separations, acid recycle, and off-gas abatement. The dissolution, acid evaporation, and acid distillation modules contributed to heat emissions. Heat emissions can be related to the nuclear material production rate. An atmospheric dispersion model was used to disperse the off-gases and particulates over distance. Atmospheric dispersion can be used to relate environment samples to stack emissions, which can then be related to the nuclear material production rate. The separations module based on solvent extraction emphasized the formation of hydrolytic and radiolytic species that can enter the vessel off-gas (VOG) system.

Preliminary validation of the model was accomplished by simulation of the Nuclear Waste Vitrification Project (NWVP) which was conducted at the Department of Energy (DOE) Hanford site from 1976 to 1979. The NWVP objective was demonstration of high-level waste (HLW) vitrification of reprocessed oxide spent fuel from the Point Beach PWR. The validation objective for this effort reported here was simulation of the NWVP operations prior to vitrification, with the intent to estimate effluent and emissions. NWVP operations were based on batch dissolution of ~41 rods, or equivalently ~ 86 kg-U. Separations were based on PUREX-type solvent extraction with pulsed columns, the cascade configuration is shown in **Figure 1**. The modeled off-gas system was similar to **Figure 2**. Acid recycle was accomplished by evaporation followed by fractionation (i.e. distillation).

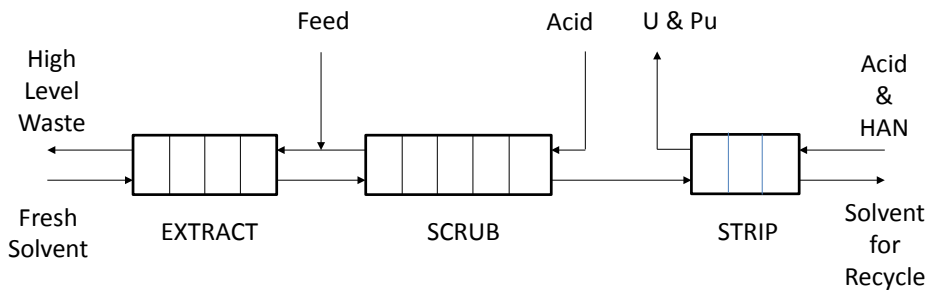

***Figure 1**: NWVP Solvent extraction cascade configuration*

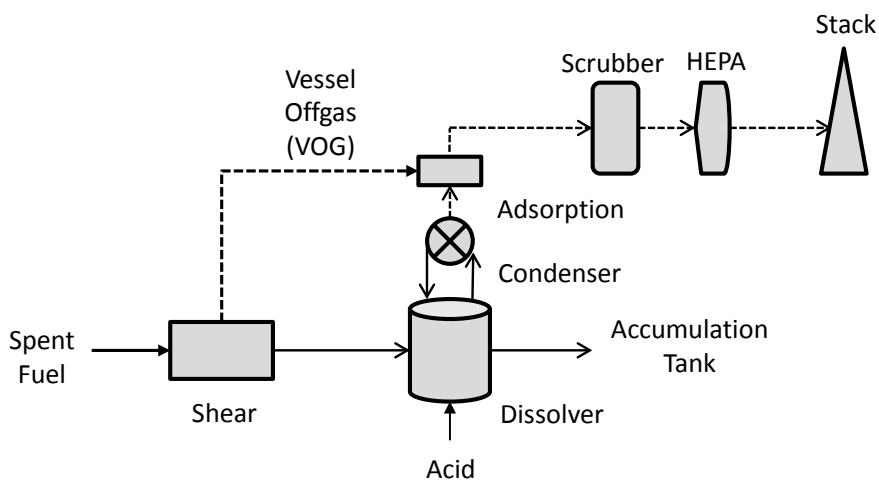

***Figure 2**: Off-gas abatement system*

Preliminary validation results comparing the simulation time dependent evolution of off-gas with that of the NWVP are shown in **Figure 3**.

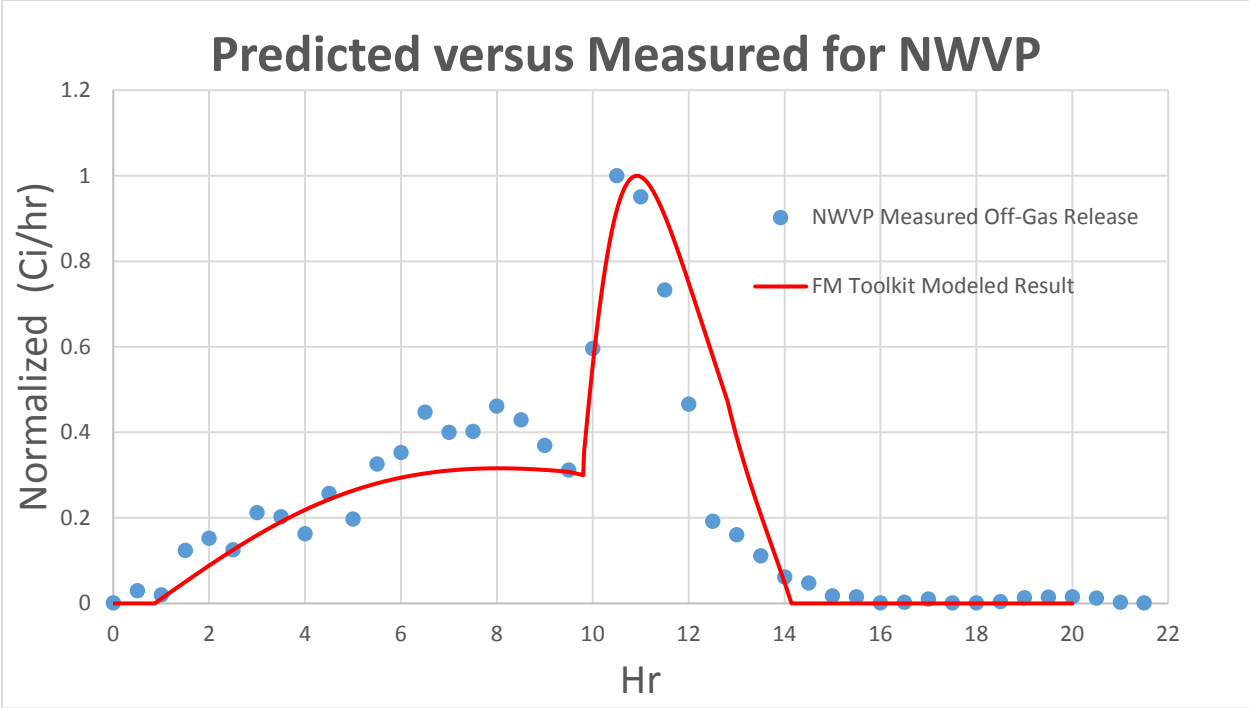

Figure 3: *Time dependent evolution of off-gas*

In conclusion, the validation results showed excellent agreement with the NWVP data. While we recognize the preliminary nature and limited extent of this validation, it does demonstrate the potential for the concept of building a flexible toolkit to simulate reprocessing facilities. As additional modules are developed, a complete toolkit for simulating different types of reprocessing facilities (and indeed additional nuclear operations) can be envisioned. This will be the foundation for a capability to solve the inverse problem (i.e. utilizing a limited set of emission and effluent measurements to infer nuclear material production rates). Uncertainty quantification (UQ) and sensitivity analysis (SA) is required on all modeled variables to produce a level of confidence associated with the inferred quantities. The mathematical theory regarding the sensitivities and predicted uncertainties of computational modules within the FM Toolkit have been reported [1] [2]. The papers constitute the first steps towards a theory section within a future computational module that implements the adjoint sensitivity method.

2. Facility Modeling Objective

The goal of the Facility Modeling (FM) project has been to develop a flexible modeling toolkit and methodology to accurately model important signatures from key steps in a reprocessing facility. The forward modeling toolkit is an essential element of a capability to relate nuclear facility signatures to the production rate of nuclear material in the facility.

Models have been developed for the processes in the facilities and the emissions from the facilities: gases, particulates, liquid effluents, and waste heat. The intended end use of the facility modeling capability is to enable inverse modeling - infer details of proliferation activity from a set of available signatures, and quantify the confidence level of the inference. Multiple indicators that some aspect of nuclear proliferation is occurring at a suspect site give higher confidence than a single indicator, particularly when signals are weak (only marginally above background concentrations or not unique). Multiple indicators of different types such as gas emissions, liquid effluents, and thermal signatures can be evaluated by a Subject Matter Expert (SME). But a quantitative statement of the probability that a nuclear proliferation site has been detected requires multiple forward runs of a facility model (that is ideally suited to application of an inverse algorithm) that simulates the suspected production process. Since the relationships between measurements of facility emissions and observations of activity patterns and frequencies are generally coupled, complex, and non-linear, to estimate production rates (with associated confidence details) automation of the forward facility model in an inverse algorithmic fashion is warranted.

Given the complexity and variability of processes involved in reprocessing, and the numerous flow sheet options that are possible, a toolkit software paradigm has been developed for a generalized facility modeling capability. The toolkit, called Facility Modeling (FM) Toolkit, is comprised of a set of computational modules and an interfacing application environment. Each computational module represents an individual process (i.e. unit operation) in the facility. Individual modules are linked in an application environment to compose an integrated “system-level” facility model for any particular reprocessing technology of interest. Details of the toolkit and integration architecture are described in Toolkit Concept.

2.1 Overview of Facility Modeling

In order to successfully develop the FM Toolkit a wide variety of technical aspects were brought together. The top six technical areas of expertise required are:

Chemical separations – PUREX process and associated flowsheet aspects.
Radiolysis and hydrolysis of reagents – modeling of signature formation chemistry
Internal facility modeling – modeling of chemical unit operations such as dissolvers and solvent extraction equipment.
External facility modeling – modeling the transport of chemicals and energy within the atmosphere.
Integrating platform – software employed to interface between analysts and computer modules.

Uncertainty Quantification and Sensitivity Analysis (UQ/SA) – UQ/SA work was performed by University of South Carolina under a separate contract. The mathematical theory regarding the sensitivities and predicted uncertainties of computational modules within the FM Toolkit have been reported [1] [2]. The papers constitute the first steps towards a theory section within a future computational module that implements the adjoint sensitivity method.

Subject matter experts (SMEs) from Los Alamos National Laboratory (LANL), Savanah River National Laboratory (SRNL), Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory (PNNL) and the University of South Carolina (USC) covering the first five of these areas were incorporated into the FM Toolkit development team. As shown in **Table 1** below, organizations often contributed expertise in multiple areas.

***Table 1**: Institutional Subject Matter Expertise*

Technical Area	***Institutional Expertise***
Chemical separations	*LANL, SRNL, PNNL*
Radiolysis and hydrolysis of reagents	*ORNL*
Internal facility modeling	*ORNL, LANL*
External facility modeling	*SRNL*
Integrating platform	*ORNL, LANL*
Uncertainty Quantification and Sensitivity Analysis (UQ/SA)	*USC*

Validation of every aspect of the toolkit is impractical given the scarcity of pertinent data and budget/time limitations. Expert opinion taken from SMEs was also utilized to fill in data gaps where necessary. In particular, SRNL and PNNL SMEs with direct first-hand knowledge of the operations that generated the validation datasets proved extremely valuable.

An example of a large-scale reprocessing facility is shown in **Figure 4** (specifically, the SRS H-Canyon facilities). Possible locations of external (i.e., thermal and chemical) signatures are also indicated in the figure. The facility shown in the figure is but one of many different types of facilities that may require analysis. Differences could arise in scale and footprint of the facility, the particular type of reprocessing technology being considered, or the signature mitigation (or abatement) technologies employed. The FM project developed a capability for end-to-end modeling of key unit operations in an aqueous reprocessing facility with the flexibility to also incorporate additional unit operations as they are developed. The FM project was a capability demonstration; as additional models are developed, the toolkit will evolve into a versatile application for proliferation detection (i.e. inferring “small-scale” facilities where only limited external signatures potentially exist).

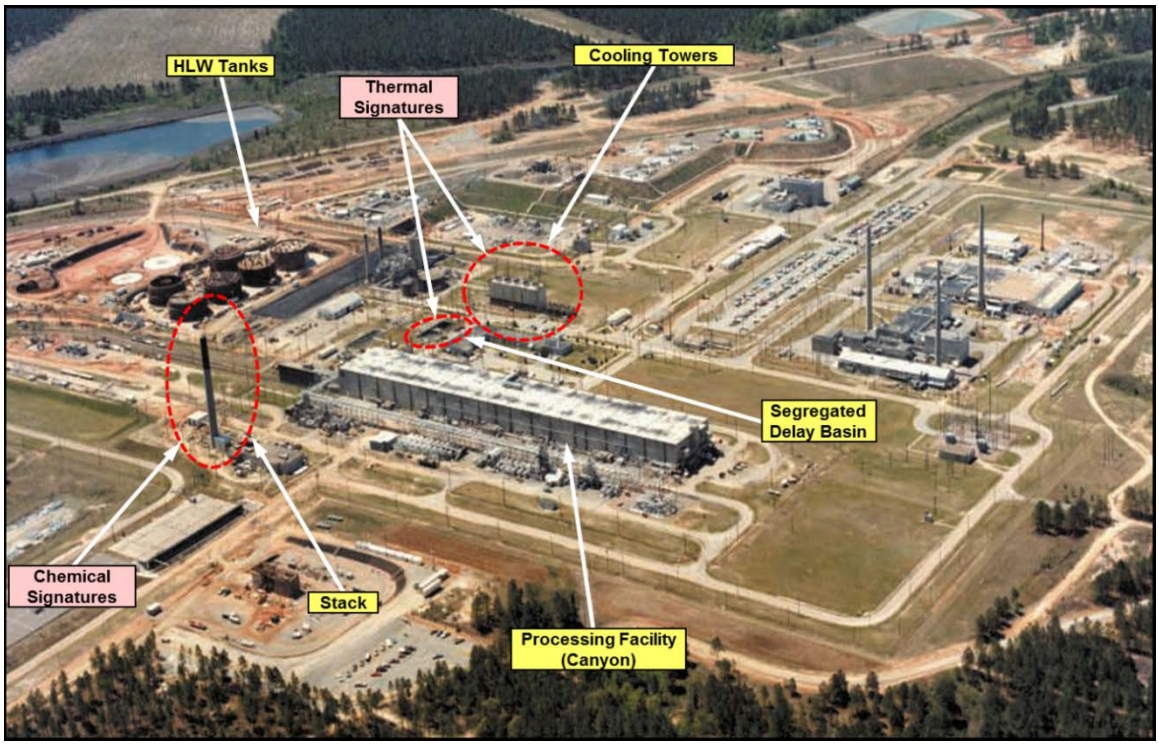

***Figure 4**: Example of a Large-Scale Reprocessing Facility, showing ancillary buildings and possible locations of exterior signatures (photo of SRS H-Canyon operations).*

A schematic cartoon illustrating the potential external signatures of interest along with the typical material flows occurring between key ancillary buildings is provided in **Figure 5**. For the FM project a variety of external signatures were considered for exploitation. These candidate external signatures are:

Gaseous and particulate emissions from stacks. The elevation of the stack release can be defined at the ground level or any height above ground.

Thermal energy emanating out the throats of cooling towers. Multiple (i.e., single or banks of) cooling towers operating under water recirculation mode is possible.

Thermal energy being deposited in local surface water basins (not being addressed in initial FM toolkit release). Typically associated with segregated once through water.

Radiation from a variety of onsite sources (not being addressed in initial FM Toolkit release). For example, above ground sources from nearby liquid waste storage tanks and retention basins.

Facility activities such as movement/transfer of material entering/exiting facility (not being addressed in initial FM Toolkit release). For example, the periodic shipments of irradiated fuel material to the loading area of the processing facility.

The majority of gaseous emissions are detected downwind of the stack at ground level fixed receptors. Multiple fixed (i.e., stationary) receptors are considered whose above ground elevations can vary. Fixed receptors can also be located directly at the stack exit if warranted. Fixed receptor sampling times and collection efficiencies are inputs to the FM Toolkit. Thermal energy outputs are measured by exhaust throat temperatures exiting one or more operating cooling towers. In order to compute these external signatures meteorological conditions are also required and are inputs to the FM Toolkit. Typically, these

meteorological conditions are extracted from more detailed weather modeling algorithms that are external to the FM Toolkit.

From the FM modeling perspective, modeling components (referred to in this report as modules, *i.e.*, a module is a computer program for the solution of a mathematical model of a subsystem) either exist internal to the facility (e.g., the canyon building in **Figure 5**, such as the dissolving unit operation) or external to the facility (such as the cooling towers). Materials, gaseous emissions, particulate emissions, and thermal plumes are mass/energy exchanges along specific system boundaries. The various potential external signatures are highlighted in **Figure 4** and **Figure 5** by red shaded circles.

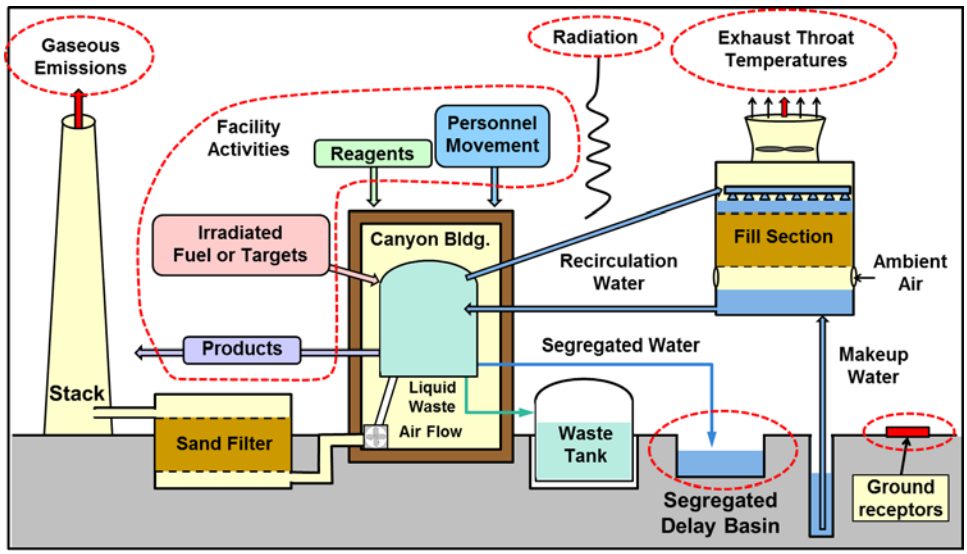

***Figure 5**: A schematic cartoon illustrating the potential external signatures of interest along with the typical material flows occurring between key ancillary buildings.*

External chemical signatures released at the stack originate from a variety of internal facility sources. These potential internal sources and some of the typical nomenclature employed are provided in **Figure 7**. **Figure 7** is a generic and very simplified view of a processing facility that focuses on the off-gas signatures. The nomenclature being employed here is:

DOG (dissolver off-gas) – Represents those unit operations generally involved in the decladding and dissolving processes of the irradiated fuel. Decladding involves either mechanical shearing or chemical dissolution depending upon the fuel type being considered.

HECOG (headend cell off-gas) – Represents the various sources (such as component leakages) that occur between those unit operations generally contained within the cells prior to solvent extraction.

VOG (vessel off-gas) – Represents the sources associated with those unit operations involved in the co-extraction, partitioning, and stripping of U and Pu from the dissolved irradiated fuel.

COG (cell off-gas) – Represents the various sources (such as component leakages) that occur between those unit operations generally contained within the cells associated with the PUREX process).

SOG (stack off-gas) – Represents the composite sum of all off-gas sources that exit the stack. Multiple stacks could be considered if warranted.

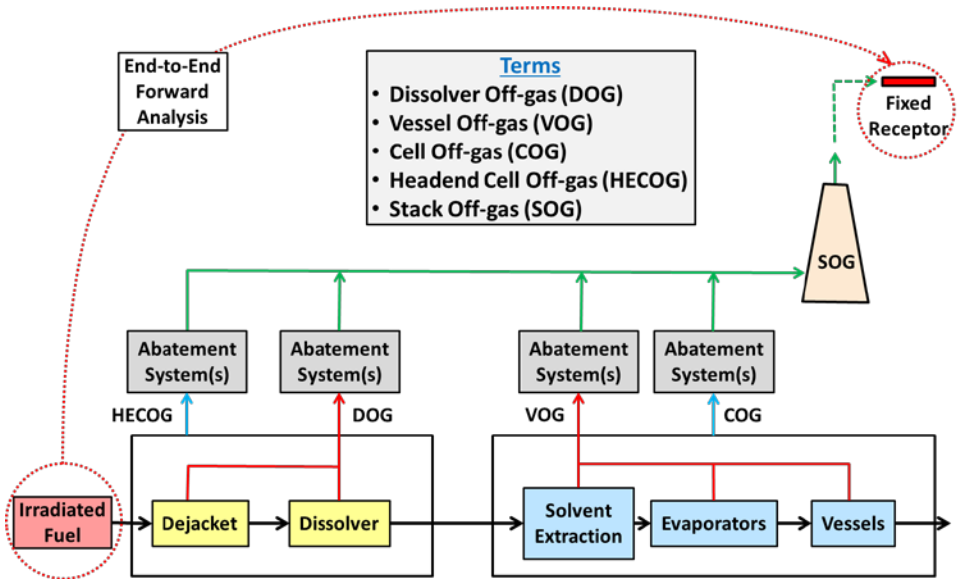

***Figure 6**: Simplified diagram of a process facility focusing on off-gas sources to the stack.*

As **Figure 6** suggests, gaseous emissions at the stack can result from a variety of contributing unit operations and can also be impacted by various downstream abatement systems. The amounts and types of off-gases that are released to the stack are fuel type dependent (as well as its burnup and cooling times). From the off-gas perspective **Figure 7** encapsulates the basic idea behind what we are calling an end-to-end forward analysis. Here irradiated fuel of known specifics (types, amounts, isotopics that are a direct result of burnup and cooling times, etc.) enter the facility from outside storage facilities and are then processed. During the processing of this fuel various off-gases are generated that end up exiting the facility through its external stack(s). Deployed downwind fixed receptors (with specified efficiencies and sampling periods) are monitoring for these gases where local meteorological wind-field and dispersive conditions are being accounted for. Here end-to-end implies that the analysis starts from irradiated fuel entering the facility (as input) and time-averaged gas species concentrations at downwind receptors are computed (key outputs of interest).

Ventilation systems, along with their optional abatement systems, can be configured in a multitude of ways. For example, the actual off-gas configuration for SRS F-Canyon is provided in **Figure 7**. The paths from the chemical dissolvers (i.e., DOG) and the hot canyon vessels (i.e., VOG) to the exit stack (i.e., SOG) are highlighted in **Figure 7**. For the DOG there are two possible paths being shown: one absent of abatement (shown by the cyan dashed line) and one with a silver nitrate reactor abatement system employed (solid red line). Note that detailed flowsheet aspects within the warm and hot cells are not defined in **Figure 7** and are somewhat different for SRS F-Canyon when compared to SRS H-Canyon and

would most likely differ some other proposed small-scale configurations. Thus, in general a multitude of configurations may be possible for both the chemical processing units (i.e., dissolvers, evaporators, condensers, vessels, solvent extraction, etc.) and their connections to the off-gas stack (i.e., abatement systems, piping connects, in/out vessel leakage, etc.).

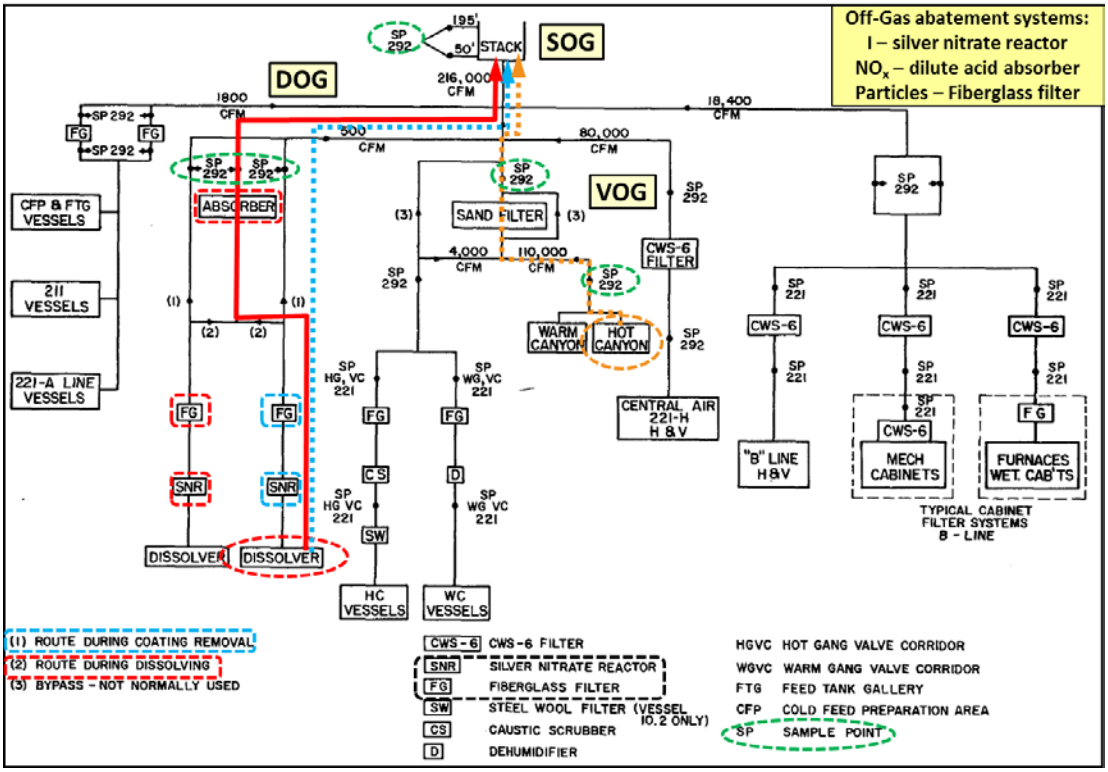

***Figure 7**: A look at SRS F-Canyon off-gas ventilation system highlighting key sources.*

For the release of the FM Toolkit the separations processes will be limited to application of the PUREX process. Benedict *et al.* provides an excellent discussion on the PUREX process and walks through the various key steps involved [3]. When employing a PUREX process there are several key steps generally taken [3]:

Decladding – Chemical or mechanical removal of cladding material.
Dissolution – Bring the U and Pu into an aqueous nitric acid solution.
NO_x Absorption – Recovery of nitrogen oxides.
Off-gas Treatment – Typically high-efficiency systems to mitigate releases
Primary Decontamination – Removal of majority of fission products.
Plutonium (Pu) Partitioning – Separation of Pu from U.
Uranium (U) Purification – Final polishing/purification of U.
Plutonium (Pu) Purification – Final polishing/purification of Pu.
Solvent Reuse – Degradation product removal for solvent reuse.
Aqueous Waste Processing – Preparation for permanent storage of liquid waste streams.

Nitric Acid Recovery – Recovery of nitric acid for reuse by evaporators.

All process facilities, small versus large scale, when using the principles of the PUREX process to reprocess irradiated (spent) fuel, will contain several of the same types of unit operations. The key steps modeled within the FM Toolkit are highlighted within the list above. These steps were chosen due to the signatures that are generated from the respective unit operations. Modeling results of the listed processes, compared to actual measured results are presented in **Section 3** for two use cases – small scale oxide fuel PUREX reprocessing with Hanford's NWVP and larger scale metal fuel PUREX reprocessing with SRS F-Canyon. The PUREX process includes a solvent extraction operation that can be performed employing a variety of staged contact equipment (mixer-settlers, pulse columns, centrifuges, etc.). There are three basic steps involved as illustrated in **Figure 8** [3]:

Co-extraction step – Separation of Pu and U (co-extraction) from fissions products.
Partitioning step – Separating Pu from U.
Stripping step – Moving Pu or U from the solvent into the aqueous phase.

The FM Toolkit contains a library of "building blocks" that represent most of the possible unit opts highlighted in **Figure 8**. Given the modular nature of the FM Toolkit, upgrades to the toolkit can be achieved through the addition of modules that address future unit opts not already available.

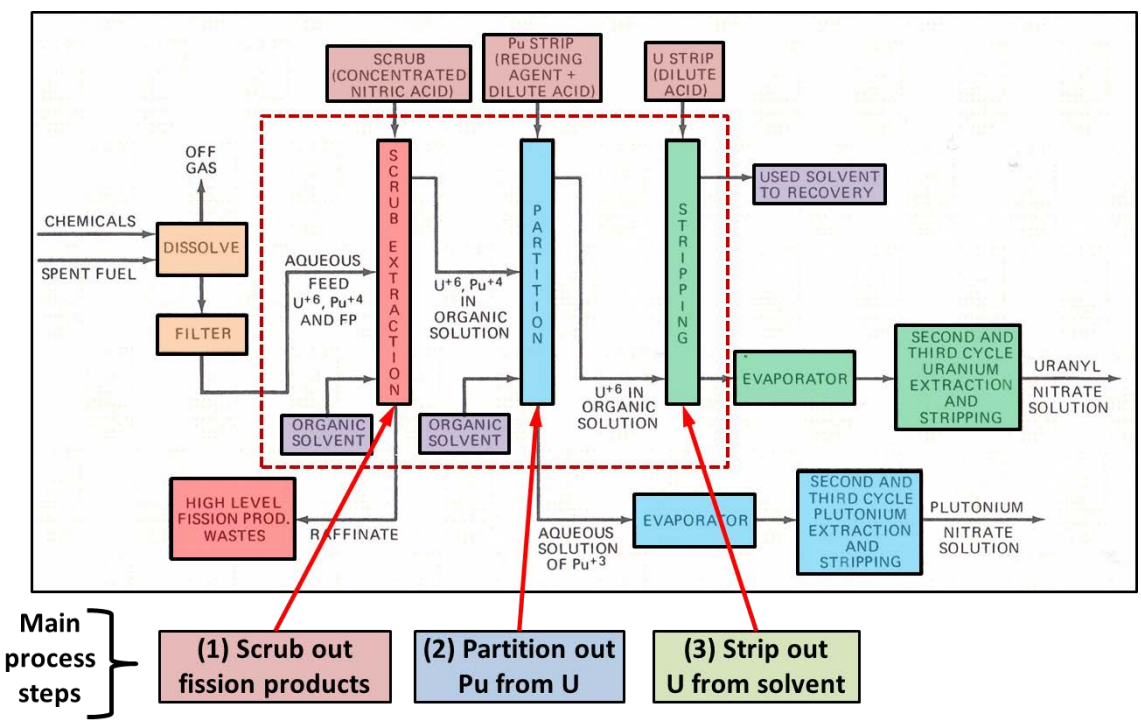

***Figure 8**: Basic outline of the PUREX process used for spent fuel reprocessing.*

As **Figure 6** and **Figure 7** might suggest, a wide range of possible flowsheet configurations are possible when assessing the Pu production aspects of a facility based on available external signatures. To reduce

the number of possible flowsheet configurations and to enhance production estimates, the exploitation of multiple external signatures is critical. These multiple external signatures will in general be partially correlated thereby reducing their individual contribution to the estimated confidence level.

To demonstrate a very simple application of the exploitation of multiple, partially correlation signatures, the following hypothetical facility is considered. Assuming that it has been detected and confirmed that this undeclared facility is producing Pu material, three downwind gaseous signatures and one thermal external signature have been observed. The following assessments can be inferred from each external signature viewed in isolation:

Gas-1 – Are we dealing with reprocessing of spent fuel? Detection of potential Pu production is the result of this observable.

Thermal-1 – Amount processed constrained by energy used? A one-sided argument.

Gas-2 – What type of irradiated fuel being processed? Indication of either fuel cladding or core materials.

Gas-3 – What sort of chemical flowsheet is involved? Impact of abatements systems.

When viewing the above information within the FM Toolkit, the initially broad range of possible flowsheet configurations should be reducible to a much smaller subset. For example, assuming all possible configurations to represent our sample space, each signature above will potentially reduce this likely subset of configurations. The rectangular region shown in **Figure 9** represents the entire sample space of possibilities. Each signature represents a subset of this space consistent with that signature's information. For example, Gas-1 (shown as a circle) reduces the possibilities down to only those configurations within the grayed in region. The Thermal-1 signature further reduces the possibilities based on the constraint of energy utilization. The Gas-2 and Gas-3 signatures continued to refine our estimate of this particular facility's configuration. The net result is the intersected region (shaded in red) shown in **Figure 9** which represents only those configurations that are consistent with all of the available signatures.

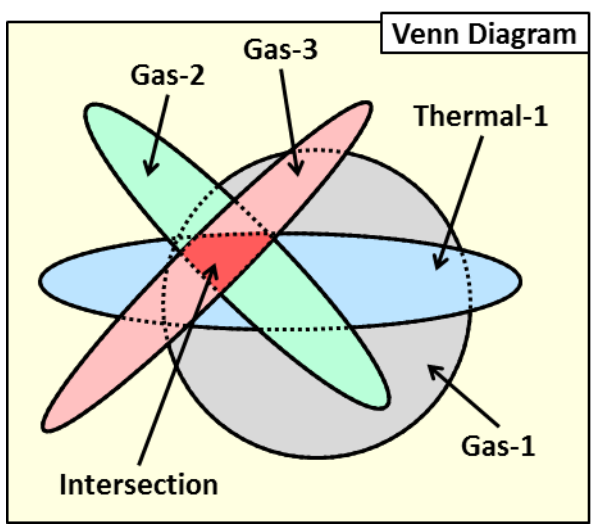

***Figure 9**: Simple Venn diagram "like" cartoon to illustrate multiple, partially correlated signatures.*

In theory, the area within the intersected region will become much smaller than any of the individual regions associated with one signature. By greatly reducing the possible number of configurations, the estimated production rates will have higher confidence levels. The addition of new potential signatures will impose additional constraints that should provide higher confidence levels as well. However, in practice, the creation of a systematic algorithm to accomplish this task can be very challenging to achieve. It is reasonable to expect that such an algorithm will evolve over time as new concepts are tested and vetted.

There is a list of candidate external signatures being considered. The temporal nature of their stack exit for chemical species (or throat temperatures for thermal energy) is key aspects that can provide substantial information, especially when considered together, in inferring production intent and quantities. For the chemical signatures the advective and dispersive characteristics of atmospheric transport significantly weaken their temporal information as measured at downwind fixed receptors.

Timing of signatures is considered to be very important. We believe that such data when plotted on a consistent time basis does provide some level of holistic interpretations to be made. In practice, data gaps will exist and the intent of the algorithm is to help analysts bridge these gaps. o see how temporal information can be employed to assess production aspects, again the same hypothetical facility as discussed above is considered. In this example we will assume that five external chemical signatures and one thermal signature are being monitored. We will just consider their responses at the stack exit and cooling tower throat. In addition to these external signatures information is made available about one key facility activity (i.e., the movement of irradiated fuel into the facility at particular times).

With the periodic movement of irradiated fuel into the facility, a repeating set of external signatures is anticipated to occur. This material movement activity sets a timeline and helps anchor the analyses that follow. The temporal behavior at stack exit (i.e., SOG) is provided in **Figure 10** for all monitored external signatures. The following assessments can be inferred from each external signature viewed, not in isolation, but as a set of multiple, partially correlation signatures:

- **Gas-1** – Detection of potential Pu production is the result of this key observable. The peak of this gas species establishes a time anchor that should be confirmable with observed facility activity. Its release is dominantly the result of fuel dissolution and releases through the DOG.
- **Gas-2** – The timing of this chemical species with respect to Gas-1 indicates that it's a precursor to dissolution and strongly indicates the type of irradiated fuel being processed. It directly establishes the cladding material. Cladding characterization (e.g., aluminum-based versus zirconium-based cladding) is accomplished here based on combined information: (1) the type of chemical species monitored and (2) its timing with respect to Gas-1. It predominantly releases through the DOG.
- **Gas-3** – A slight timing delay of this chemical species exists with respect to Gas-1. Fuel core characterization (e.g., metal versus oxide core) is accomplished here based on combined information: (1) the type of chemical species monitored and (2) its timing with respect to Gas-1. It predominantly releases through the DOG.
- **Gas-4** – A range of possible timing delays exits with the possibility of multiple peaks between each irradiated fuel charging. The magnitudes of these peaks are strongly dependent upon the various abatement options that might be operative. Potential temporal behaviors are shown

for abatement options: (1) no abatement; (2) Ag reactor(s); and (3) Hg addition. It predominantly releases through the DOG and VOG.

Gas-5 – A range of possible timing delays exists with the possibility of multiple peaks between each irradiated fuel charging. This particular chemical species relates to radiolysis and hydrolysis of solvent (i.e., degradation modes) and releases into the VOG from sources such as: (1) 1st cycle solvent extraction; (2) 2nd cycle Pu purification; (3) evaporation steps; and/or (4) dissolving step. This signature helps in addressing the arrangement of unit ops within the working flowsheet.

Thermal-1 – This thermal signature helps to further establish quantification in the amount of Pu being produced. Since energy utilization depends upon efficiency factors, only an upper bound estimate on production is achievable from this signature alone (i.e., a one-sided constrained).

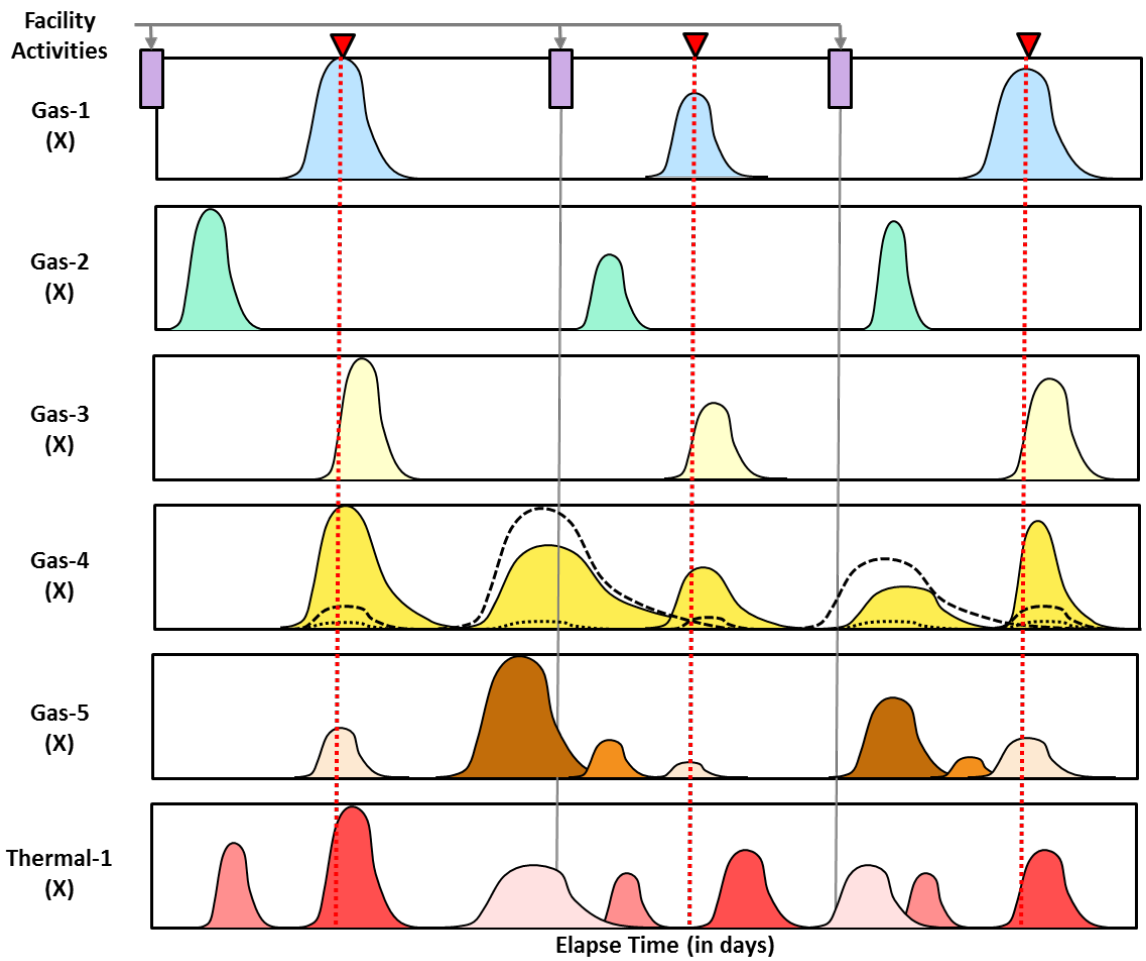

***Figure 10**: Example temporal nature of multiple, partially correlated signatures.*

The above examples illustrate the usefulness in: (1) exploiting multiple, partially correlated signatures and (2) making use of their coupled temporal nature. To take full advantage of these various sources of information, physics-based models must be considered.

Therefore, to fully exploit these multiple, partially correlated signatures, requires a flexible, modular, and integrated forward-modeling capability such as the FM Toolkit. The motivation behind the design of the FM Toolkit can be expressed by:

Why Flexible? – To rapidly evaluate multiple candidate flowsheets.

Why Modular? – To rapidly evaluate different flowsheet variants, subset of operations within a flowsheet, and different sequencing or timing of operations; to identify possible processing scenarios that best explain the set of observations (i.e., best match to multiple signatures); and to quantify confidence in numerical estimates which requires uncertainties to be quantified in individual unit operation models.

Why Integrated? – Unit operations and their correspondent signatures are coupled physically, chemically, and temporally.

3. FM Demonstration of Capability

Most of the FM Toolkit demonstration efforts in this project were focused on the Nuclear Waste Vitrification Project described in the next subsections. In addition, work as applied to the SRS F-Canyon dissolver was carried out for metal fuel; this is described in the final subsections of this section.

3.1 Introduction - Validation by Nuclear Waste Vitrification Project (NWVP)

The Nuclear Waste Vitrification Project (NWVP) was chosen for validation of the FM Toolkit due to its well-characterized (1) off-gas emissions, (2) facility description, and (3) small scale facility operation [4]. The NWVP was initiated to provide a demonstration of the vitrification of high-level liquid waste (HLLW) from spent fuel discharged from an operating light-water reactor (LWR). In fiscal year (FY)-1978 the "Monitoring Methods for Particulate and Gaseous Effluents from Waste Solidification Processes" project was modified to that of characterizing the vitrification related off-gas emissions associated with the NWVP, this also included the off-gas related to reprocessing prior to vitrification [5]. The NWVP was located on the Department of Energy (DOE) Hanford site, built and operated by the Pacific Northwest National Laboratory (PNNL), started in April 1976 and terminated in June 1979.

3.2 NWVP Hardware

For FM Toolkit validation and demonstration of capability only the reprocessing portion of the NWVP, not the vitrification operation, was of interest. Process operations for the reprocessing portion of

NWVP, used for the FM Toolkit validation, are shown in **Figure 11**. Process stream numbers shown in **Figure 11** were used as identification for the related computation modules developed within the FM Toolkit. Both material and energy balances were modeled for the FM Toolkit validation, more specifically material balances for significant process streams and energy balances relevant to the cooling tower. Equipment designs and specifics of the facility layout are described in detail in the referenced reports, and therefore, will not be discussed here.

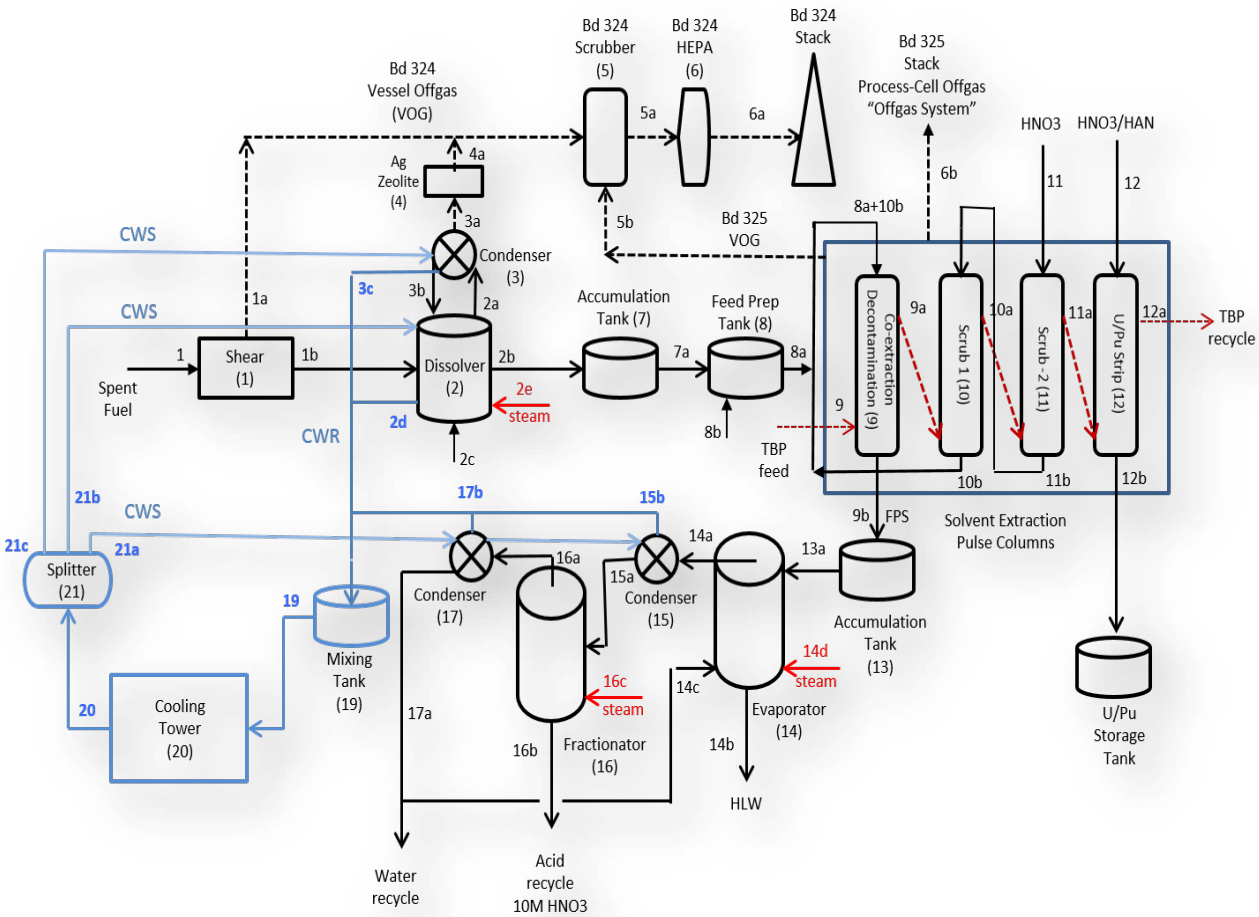

***Figure 11**: Process flow model of NWVP used for FM validation*

Chop/Shear

A commercial hydraulic shear was modified for use by increasing its resistance to radiation, see **Figure 12**. The cut fuel pieces dropped down a chute into a basket located in the dissolver. A vessel off-gas (VOG) system vented to the facility off-gas system, which included the gases and fines released during chopping. The facility off-gas system then treated and/or trapped these emissions prior to release from the facility stack.

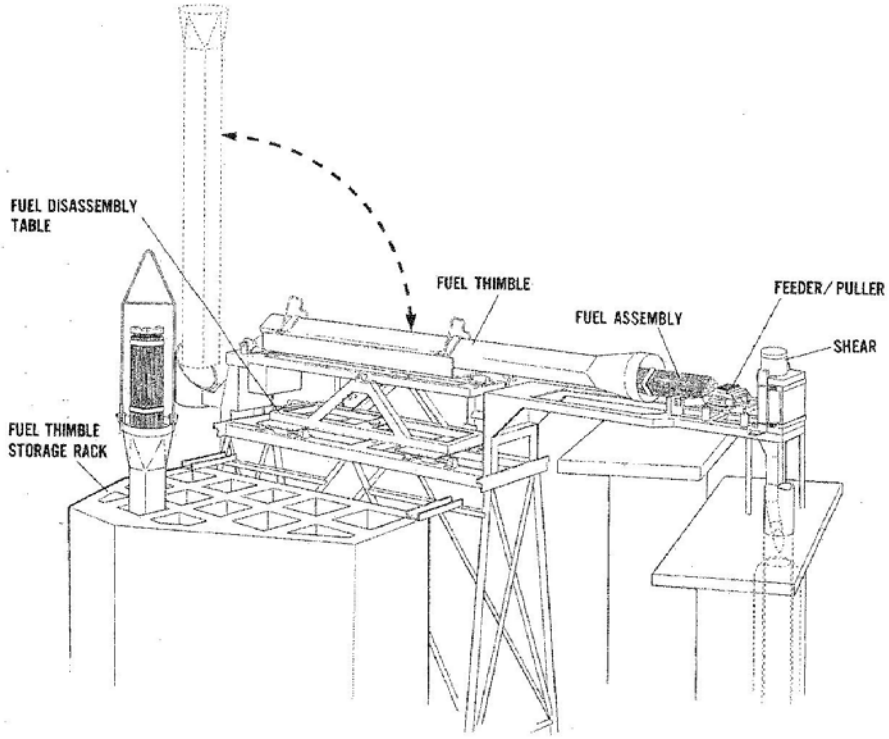

***Figure 12**: Chop/shear system*

Initially, the FM Toolkit preliminary chopper module chopped fuel according to the NWVP validation efforts and dropped fuel segments into the dissolver at the request/signal from the dissolver. All of the gases monitored for stack emissions were contained within the solid structure of the fuel segments within the dissolver for the NWVP validation efforts. Exact gas phase speciation within the chopper had to be developed to account for small gas and particulate release as chopping occurred. Currently, the chopper can model the speciation and gas phase dynamically at the specified operational time of the validation case being tested. As fuel is being chopped during a simulation, gas phase release from the plenum and the fuel during chopping is modeled and passed through the off gas system. Small perturbations in the gas being released during chopping can now be seen through the stack.

The FM Toolkit contains a FUEL_DEPOT module which creates a *FuelBundle* object to represent a used fuel bundle and its solid/gas species. In the absence of available speciation for used nuclear fuel, the *FuelBundle* object fills the gap of creating solid species as follows: uranium oxide, plutonium oxide, minor actinide oxides, fission product oxides, volatile fission product (trapped in the solid phase), and remaining fission products. In the gas phase of the fuel bundle (plenum), gas phase species are created as volatile fission products. FUEL_DEPOT generates all species, molecular formulae, their molecular masses, and corresponding radioactive information, using rigorous nuclide data from the periodic table supplied as a support module in the FM Toolkit framework, Cortix. The periodic table contains stable and unstable isotopes data for a large number of fission products. This periodic table has been extensively modified to include radioactive nuclides. FUEL_DEPOT uses the flexibility of the *Specie* module to define many fictitious species for practical use while retaining all the original information of

the nuclides making up the species. Much work was devoted to the FUEL_DEPOT module to simplify the number of nuclide species present while keeping an accurate representation of all involved quantities.

Dissolver

The NWVP batch-type dissolver consisted of a primary vessel with an auxiliary reservoir, and a dedicated off-gas system, see **Figure 13**. The dissolver off-gas system consisted of a demister, condenser, steam heater, and silver zeolite bed, see **Figure 14**. A perforated basket into which the fuel pieces drop fit within the primary vessel of the dissolver.

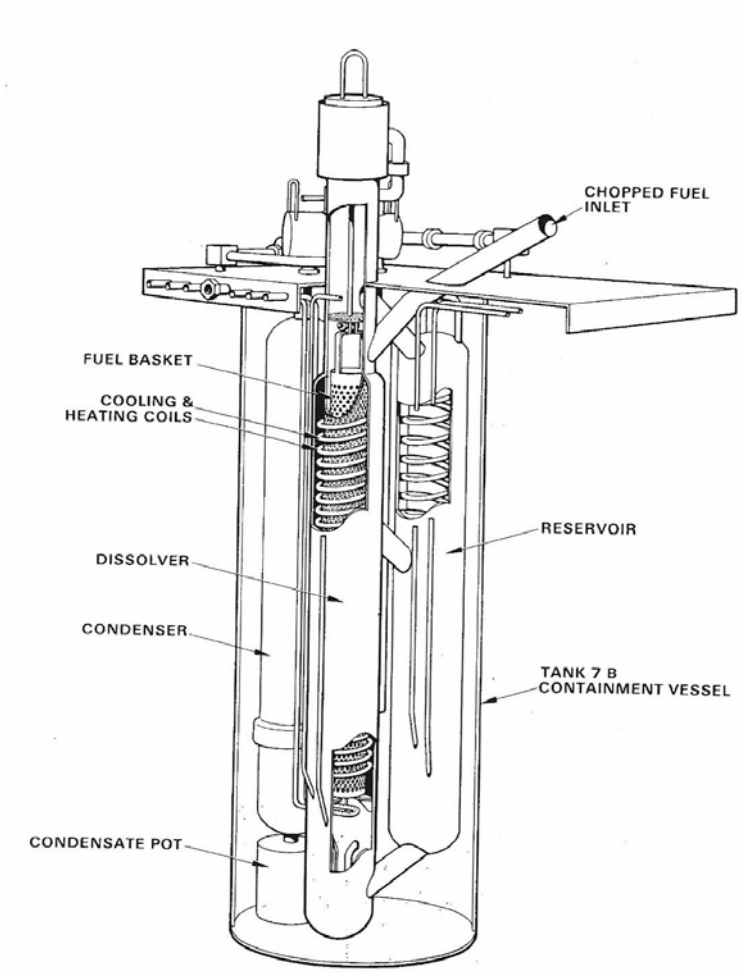

***Figure 13**: Dissolver*

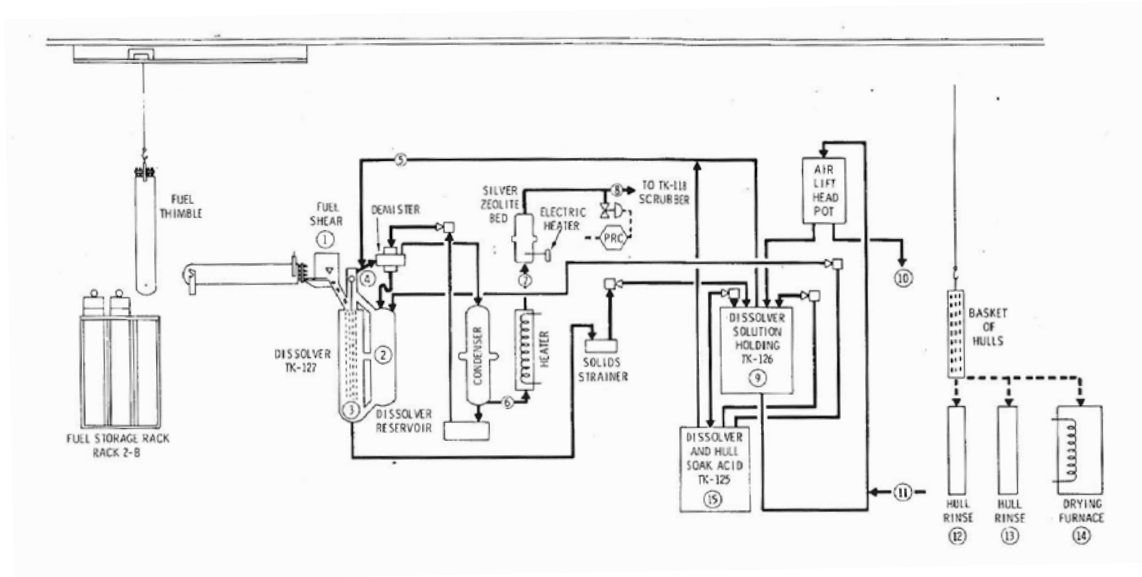

***Figure 14**: Dissolver off-gas system*

Volatile fission products were added as new species in the *Dissolver* module and the disengagement of volatiles from the liquid into the vapor are better modeled as a mass transport rate model. Combining this model with information extracted from the NWVP report (specifically, the gradual addition of nitric acid to the dissolver before the heating up of the dissolver contents) resulted in a much improved result being obtained as shown in **Figure 15**. The figure shows the measured release of a specific signature noble gas plotted over the entire dissolution batch time for a specific NWVP batch which will be called Case D for classification reasons. Here the improved mass transport model for volatile fission product release allowed for a better representation of the Gaussian peak (compare the blue curves to the red curves), and the position of the peak and magnitude (blue curve) match the experimental data more accurately. As shown in **Figure 15**, the modeled signature data within the FM Toolkit closely matches the experimental data from NWVP.

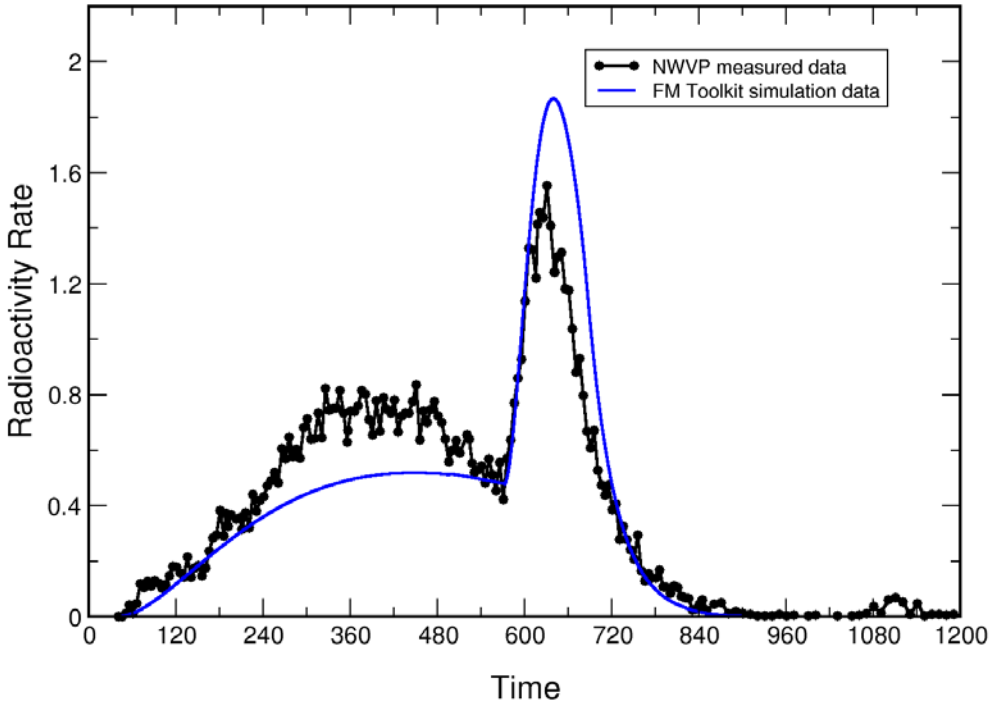

***Figure 15**: FM Toolkit Comparison of a Signature Noble Gas Stack Release vs Experimental NWVP Dissolution Batch Data*

Additionally, to allow for better validation simulations of the FM Toolkit capability when compared to NWVP experimental data, improved modeling of the condensation model for the dissolution system has been carried out. The condensation model takes into account transient (non-equilibrium) production of nitric acid, and a condensation pot added to the model to better represent the NWVP condenser. The SUNDIALS (Suite of Nonlinear and Differential/Algebraic Solvers) package (2.6.2; LLNL) is used with the condenser (CVODE) and dissolver (CVODE) modules. The solvers are more robust and faster and the CVODES solver can be used to calculate sensitivities (forward and/or adjoint) without any additional algorithm development. In view of the evolving nature of the modules and models, and the non-linearity of the chemically reacting models, advantage is taken by using numerical Jacobian calculations provided by SUNDIALS. The use of SUNDIALS in the FM Toolkit modules is an essential element for the successful prediction of gaseous signatures from the dissolver off-gas system.

Facility Off-gas System

Downstream of the dissolver off-gas system was the NWVP facility off-gas system consisting in order of (1) silver zeolite adsorption filter, (2) packed-bed caustic scrubber, and (3) HEPA filter(s). Following the HEPA filter, the emissions were vented through the facility stack.

The packed-bed caustic scrubber was modeled with the following simplifications:

- The model is applicable only to a diluted inlet gas stream washed with an uncontaminated liquid inlet stream;
- The modeled scenario operates at steady state;
- The column does not get clogged by particulate or precipitate;
- The column work at ambient pressure and temperature.

Flowing inert gas is mixed with sprayed caustic water where inert and non-inert gases absorb according to their solubilities and/or by chemical reaction with the caustic. The FM Toolkit scrubber module has been modeled for an inert gas where absorption depends only on solubility not chemical reaction. For the case of low concentrations in the gas for the inert species, Henry's Law applies for solubility. Utilizing Henry's Law, one can determine the concentration of inert species in the exhaust of the scrubber as a function of the gas flow rate into the scrubber, concentration of the inert in the gas flowing into the scrubber, Henry's Law constant for the species in the scrubber solution, total pressure of the gas flowing into the scrubber, and the mass flow rate of scrubber solution bleed from the scrubber bottoms.

The performance characteristics of HEPA filters has been defined by the U.S. Department of Energy (DOE) for filtration of CAT 1 and 2 facilities, shown by **Figure 16** [6]. This requirements state that only 0.03% of 0.3 microns diameter particulates shall escape the filter. Furthermore, the most penetrating particulates in typical HEPA filters used in CAT 1 and 2 facilities have been found to be slightly greater than ~0.2 microns diameter. **Figure 16** shows a typical particle diameter normal distribution based on (1) 0.03% of 0.3 microns diameter particulates escape filter and (2) ~0.2 microns diameter particulates as most penetrating.

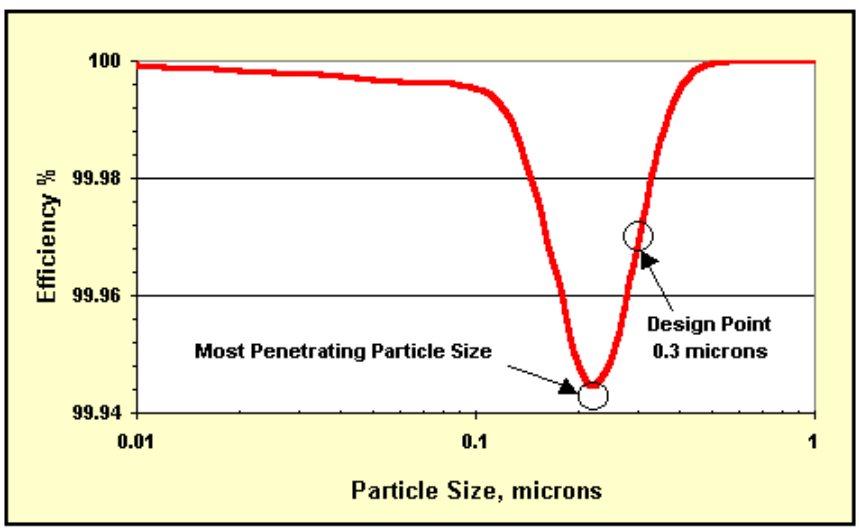

***Figure 16**: Performance of HEPA filter for DOE CAT 1&2 Standard*

(0.03% of Pre-filtered particles at 0.3 microns)

By observation, the mean particulate diameter for the characteristic distribution of **Figure 16** is ~0.22 microns and the standard deviation for particulate diameter is ~0.07 microns. Knowing that 0.03% of 0.3 microns diameter particulates escape filter, the FM HEPA module can determine the mass flow rate of any diameter particulates escaping filter given the mass flow rate of 0.3 microns diameter particulates entering the filter.

Separations

Portions of the separation system used for FM Toolkit validation included (1) the feed makeup tank, (2) pulsed columns (see **Figure 17**) representing extraction, scrub, and co-strip of the uranium and plutonium (see **Figure 18**), and (3) product and waste collection tanks. NWVP made use of ion exchange columns for partitioning of the uranium and plutonium; however, this operation was not included in the development of the solvent extraction FM Toolkit module. The solvent extraction module consists of equilibrium stages with “fixed” distribution coefficients. It differs from classic solvent extraction models such as Solvent Extraction Processes Having Interacting Solutes (SEPHIS) due to (1) the use of these fixed rather than variable distribution coefficients, (2) interpolation of the concentrations gradients within a bank (i.e. extract, scrub & strip), and (3) the additional feature of gamma and alpha radiation exposure for the reagents. Differences (1) and (2) simplify the classic modeling approach of an iterative material balance solution with variable distribution coefficients, while (3) enables the determination of the radiation profile necessary to estimate relevant signatures. Classic solvent extraction models such as SEPHIS simultaneously solve a set of equations for all stages in all banks in an iterative fashion. The solution is complicated by the fact that the distribution coefficient is variable, dependent on the concentrations of nitric acid (HNO3), tributyl-phosphate (TBP), and other extractable species. Because both the TBP and nitric acid also become complexed with species other than Pu, most significantly uranium, the set of material balances should be solved simultaneously for all significant species. It is because of these complexities that the simplified solvent extraction module described here was used to expedite demonstration of the overall forward modeling capability. The solvent extraction model can be used to represent pulsed-columns, mixers-settlers, centrifugal contactors, and/or any solvent extraction equipment where the number of equilibrium stages are known. Because cesium decontamination was measured for NWVP, it was used to demonstrate computation of the gamma exposure. Similarly, plutonium recovery for NWVP was used to demonstrate computation of the alpha exposure.

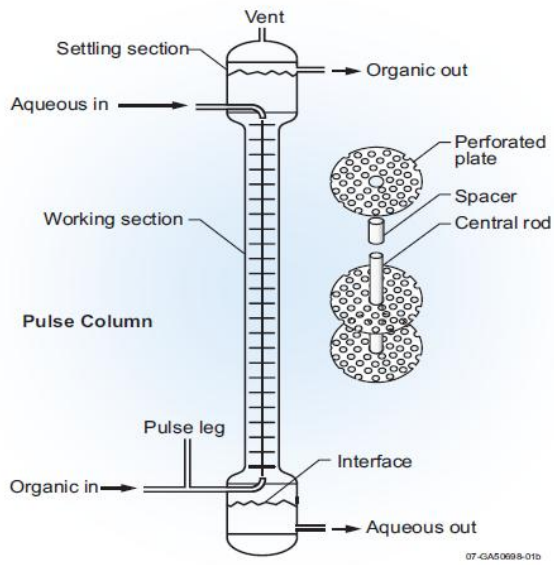

***Figure 17**: Example pulsed-columns*

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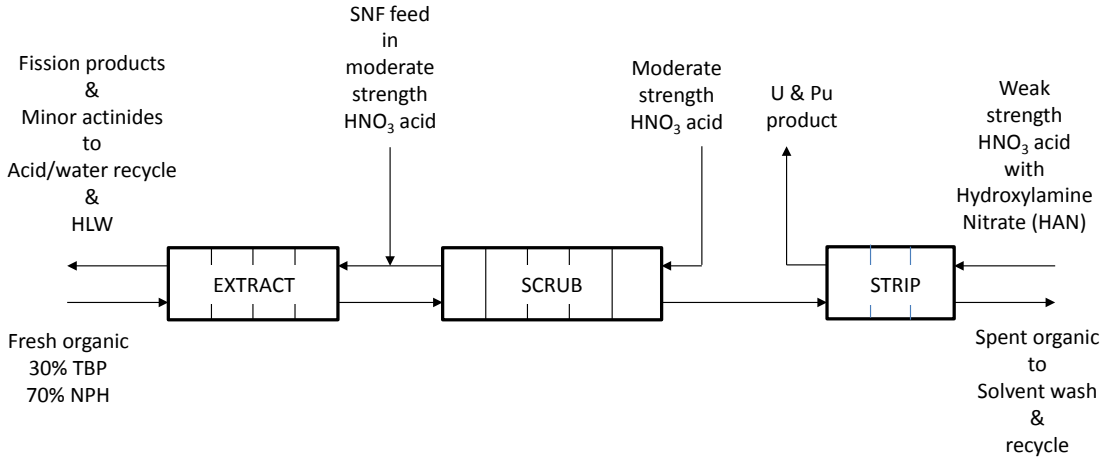

***Figure 18**: Extraction, scrub, and strip of solvent extraction*

The values determined for **Figure 18** are used to solve for the concentration and radiation profiles throughout the extraction, scrub, and strip banks. The radiation profile per bank, for NWVP conditions, determined using the solvent extraction module is shown in **Figure 19** for total Pu and ^{137}Cs, where total Cs consists of predominantly ^{137}Cs after long cooling of spent fuel.

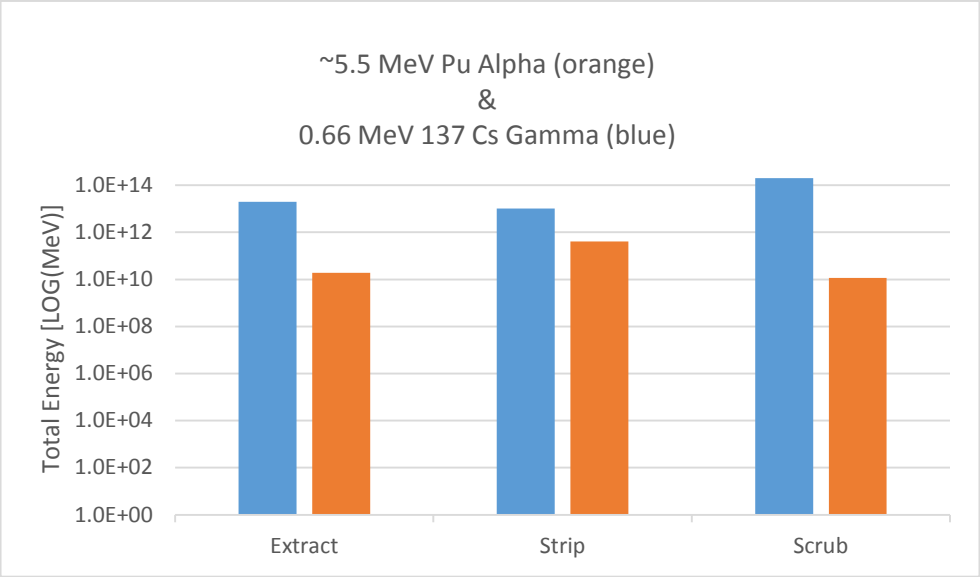

***Figure 19**: Radiation profile for NWVP conditions due to Pu and 137Cs*

Acid Recycle

Acid recycle was included in the model validation because of its significant contribution to the facility overall energy consumption and consequent facility heat rejection through the cooling towers. Acid recycle was accomplished by (1) evaporation to concentrate the HLW and then (2) fractionation (e.g. distillation) of the evaporate distillate to produce water both for recycle and near-azeotropic nitric acid.

Solvent Recycle

Solvent recycle was not included in the initial model validation; however, it was to be included in future validation efforts.

Cooling Towers

Cooling towers for process heat rejection were included in the validation, the source of heat to these towers for the FM Toolkit validation consisted of the dissolver and acid recycle operations. The modeling of the external thermal signature is accomplished using the *CTTool* module (released version 1.0). Details for *CTTool* (v1.0) can be found in Aleman and Garrett [7]. In this reference many of the details concerning the algorithm and validation efforts can be found.

The *CTTool* module models mechanical draft cooling towers (MDCT's) that are widely used by industry and proliferant sites to dissipate moderately large waste heat loads (a few megawatts up to about 100 megawatts). The temperature of the air being exhausted from the MDCT is a measure of the amount of thermal energy being removed from the process cooling water, although ambient weather conditions and cooling water and air flow rates must be known or estimated to calculate the rate of thermal energy dissipation. It should be possible to derive information about the plant or facility operating status from accurately calibrated thermal imagery of MDCT's. The *CTTool* module is designed to operate in several modes: the benchmarking mode (SRS A-Area, F-Area or H-Area cooling towers), the recirculation water

mode or cooling tower calculation only, and the remote sensing mode for single cooling tower throat or multi-cells (throats and exposed decking materials). The *CTTool* module in the facility modeling capability demonstration flowsheet is executed in the recirculation water mode as shown in **Figure 20**.

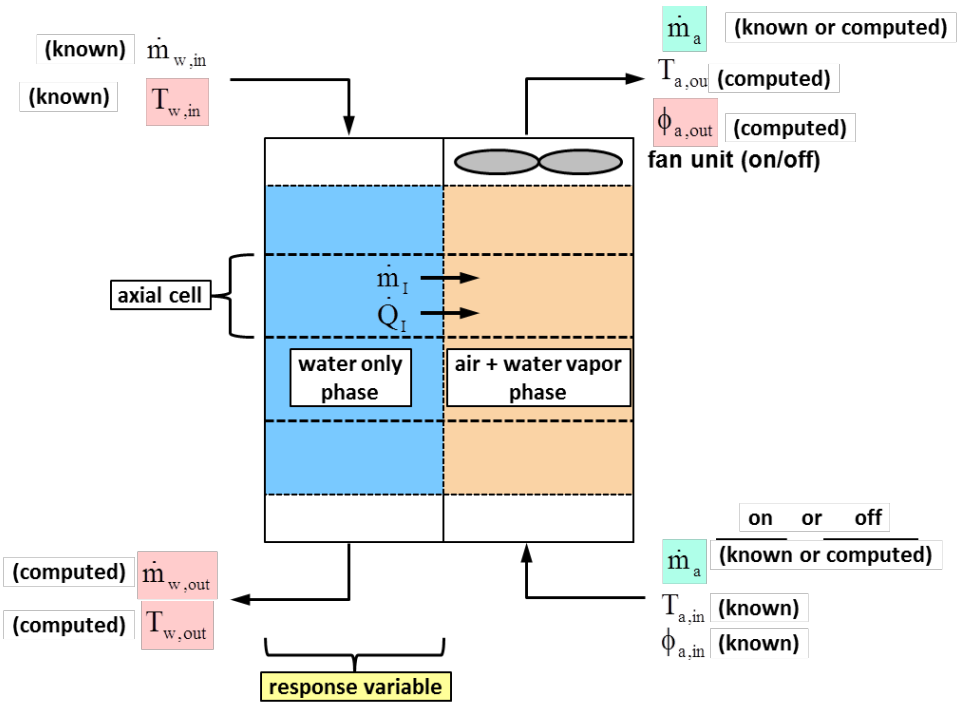

***Figure 20**: Counterflow cooling tower in recirculation mode*

Stack

The NWVP stack served as the termination of facility processing and the source for the plume. It was the location of sampling for the source that contributed to the plume module. One very simple cartoon of the overall process is shown in **Figure 21**. The stack contributes potential signatures through downwind collection of pollutant material at fixed receptor locations.

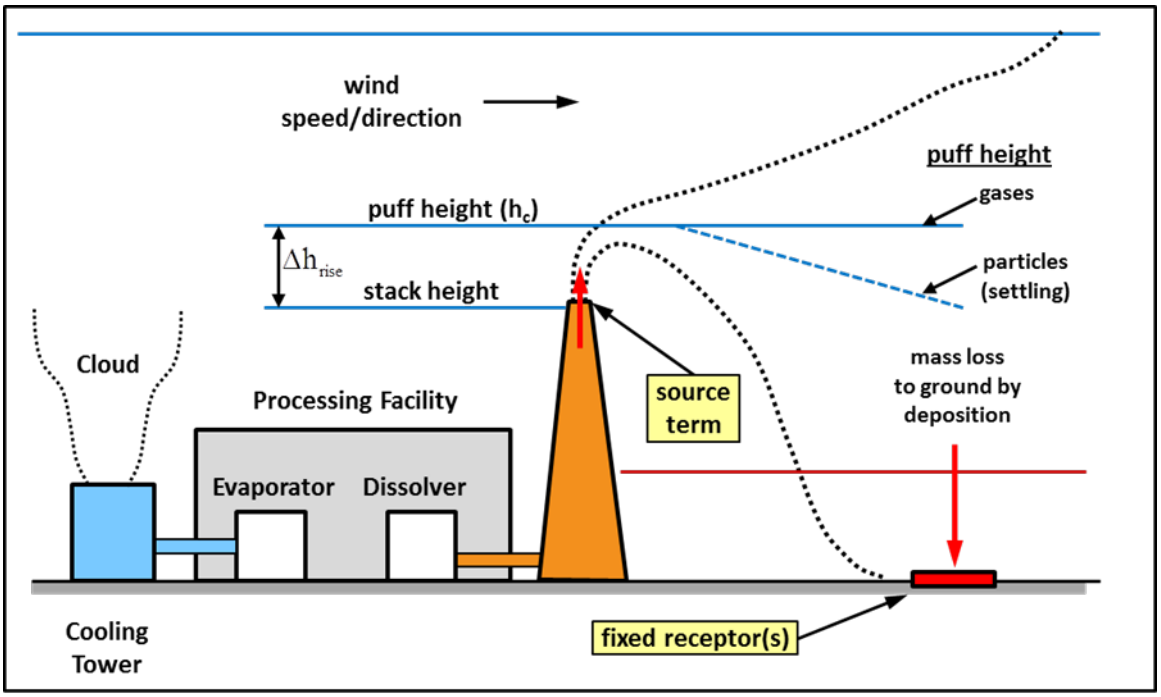

***Figure 21**: High level conceptual view of processes modeled in the FM program*

The pollutant's atmospheric fate and transport from its emission source (elevated stack) to its ultimate deposition and/or dispersal downwind is modeled by *PUFF-FM*. Analysis is limited to a single species which may be either a soluble gas or an insoluble distribution (by size) of particles subject to 1st order loss (radioactive decay and/or wet deposition). The ultimate predicted responses of most interest are downwind time-sample average concentrations and cumulative deposition at spatially fixed receptor locations near the ground. *PUFF-FM* is primarily focused on tracking the release of a finely divided series of puffs from a single source location. Multiple downwind fixed receptors are numerically monitored as response variables outputted for further analyses. Forward modeling of the NWVP data for one batch generated gas concentration signatures at different fixed receptor locations. The gas signature and receptor response is dependent on the distance from the stack and the operational parameters of the module as shown in **Figure 22**. The peak concentration of the off-gas occurs during the heating of the nitric acid for dissolution of the oxide fuel. No other signatures are noticed until this event occurs, which closely couples the timing of the dissolver operation with peak signature generation. Moreover, the signature concentration is directly affected by the downwind distance the receptor is from the stack. Understanding the temporal, chemical and physical aspects of the dissolver as well as placement of the receptor can help infer specific chemical processes that may be occurring.

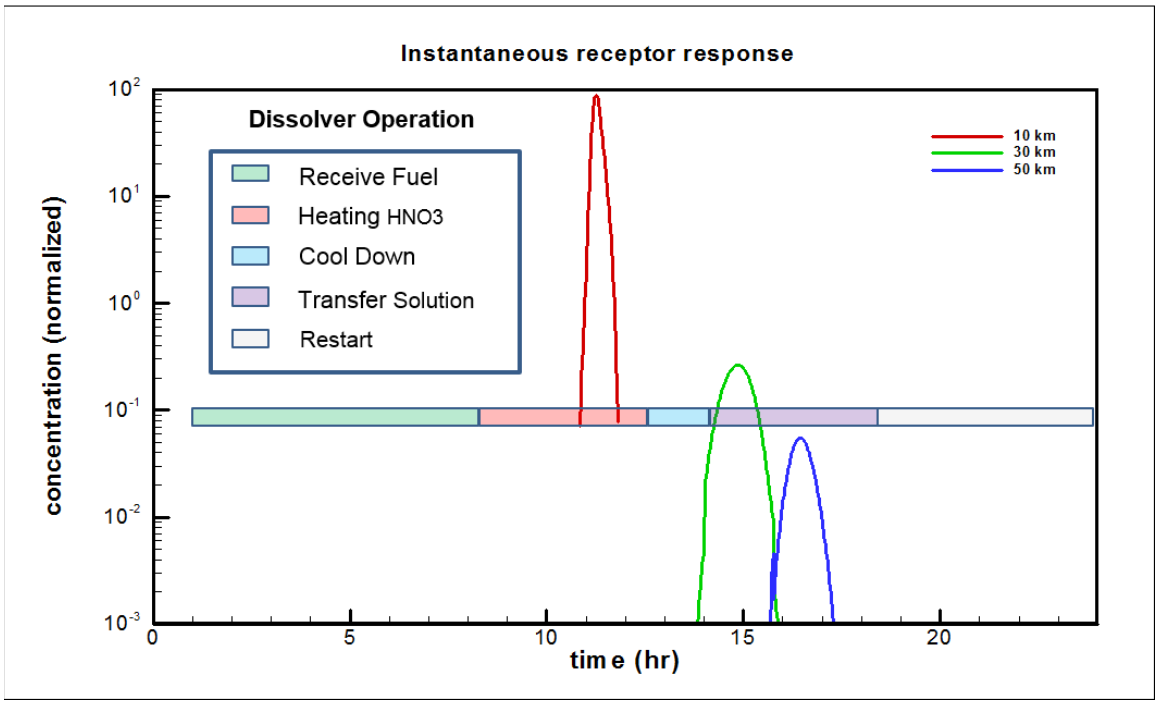

***Figure 22**: Instantaneous plume concentrations at fixed locations for a gas signature of interest.*

3.3 NWVP Operations and Results

The specific NWVP "use case" used for FM Toolkit validation was focused on a relatively short-duration of consistent operation. Since the NWVP was a one-time (albeit multi-year) demonstration of technology, not a production operation, continuous integrated operation of the full suite of equipment was problematic at times. Consequently, operating conditions for FM Toolkit validation were selected based on consistent operation of the fuel shear/chopper, dissolution, solvent extraction, and off-gas systems. Additionally, not only was consistent operation necessary for validation, but also thorough off-gas sampling was required at both the stack and downstream (in the plume) locations.

In Building 324 the chop/shear and dissolution operations were conducted 24-hours/day and 5-days/week, in Building 325 the solvent extraction operations were conducted 24-hours/day and 7-days/week. Fuel rods were pulled from an assembly and fed into a feeder-puller with a manipulator, average time to shear a batch of 40 to 42 pins was eight hours and was equal to approximately 86 kg-U per batch. The 40 to 42 pins represented a single dissolver batch. Because the production rate for the dissolver was greater than the throughput of the solvent extraction system, and through the judicious use of feed accumulation and makeup tanks, the solvent extraction system was operated continuously. However, occasional problems with the chopper/shear, dissolver, off-gas system, and other equipment did interfere with continuous plume generation during an individual batch. Data from these runs were rejected for the purpose of FM Toolkit validation. Four specific batches were chosen for validation of the FM Toolkit modules. The four batches were chosen based on minimal mechanical issues with the operation of the chopper/shear and dissolver. Upon detailed review of the four batches, only three of

them contained enough representational data for validation and comparison. The fourth case involved two different fuel assemblies being processed within the same batch, which complicated the NWVP off-gas sampling data. The validation involved comparing the concentration in curies per hour of a noble gas signature of interest over the entire dissolution time for that batch. Complete dissolution of the fuel rods took approximately 18 hours for each batch before the solution was transferred to holding tanks for further reprocessing. **Figure 23**, **Figure 24** and **Figure 25** show the results of the signature of interest compared to data collected and reported in NWVP documents. Specific concentration values are omitted for classification purposes and the results are normalized. Specific detailed results can be provided through classified avenues upon request.

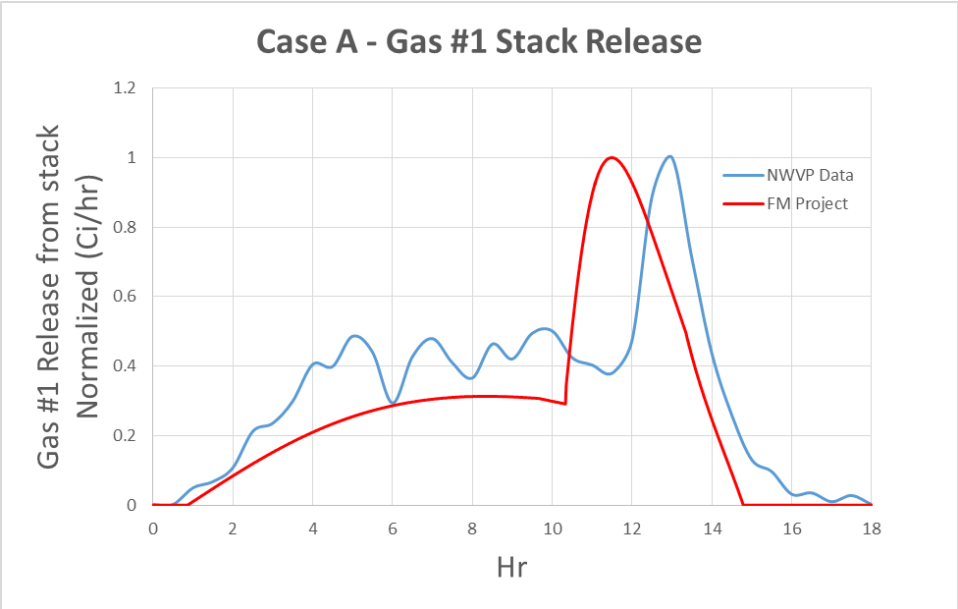

***Figure 23**: Comparison between NWVP Data and FM Modeled Data for Noble Gas Signature Concentration as a function of Time – Batch A*

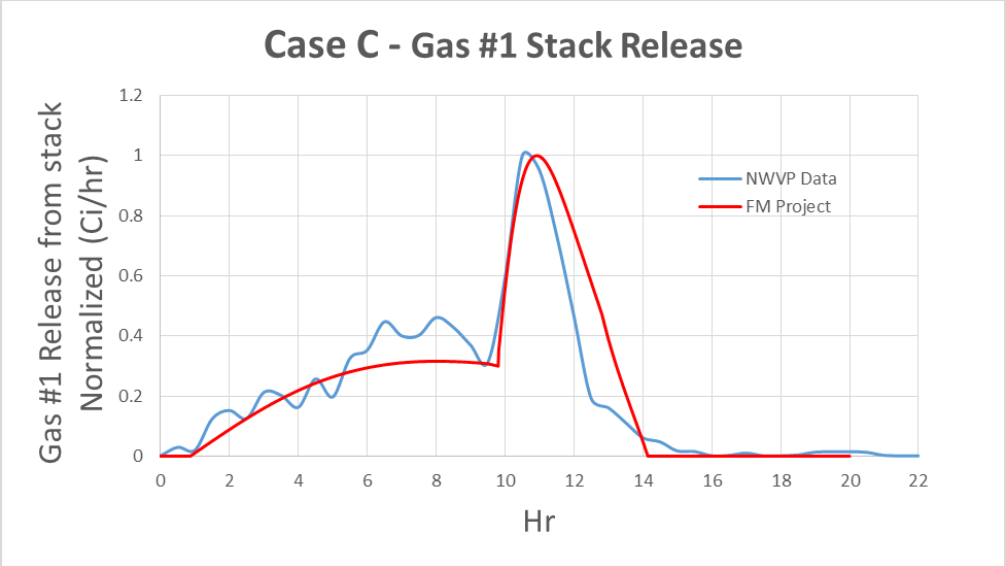

***Figure 24**: Comparison between NWVP Data and FM Modeled Data for Noble Gas Signature Concentration as a function of Time – Batch C*

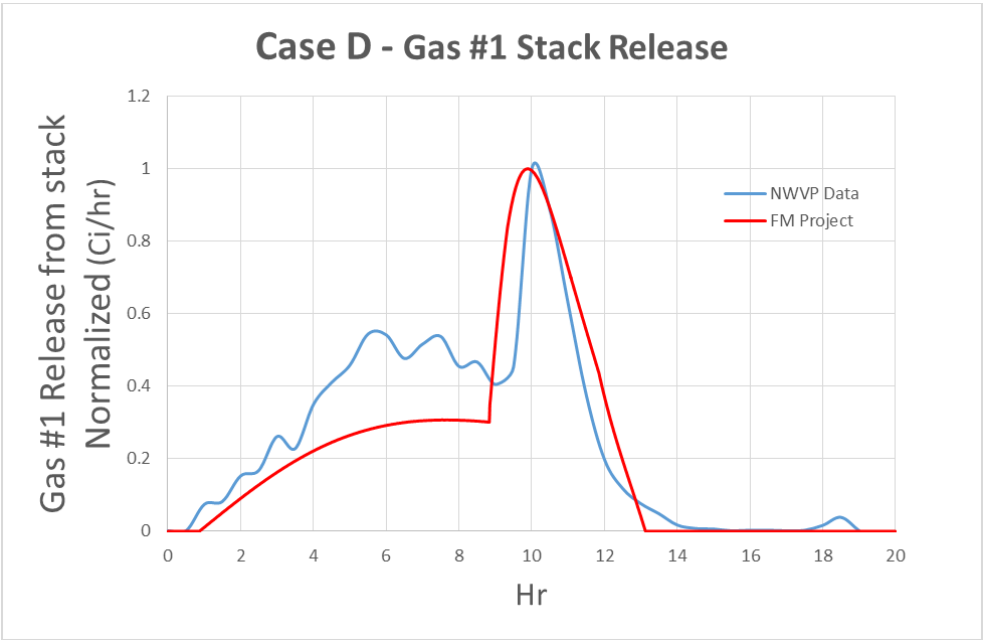

***Figure 25**: Comparison between NWVP Data and FM Modeled Data for Noble Gas Signature Concentration as a function of Time – Batch D*

Excellent agreement between actual NWVP data and data modeled by the FM Toolkit can be seen within these three cases. Additional gases of interest are being modeled within the FM Toolkit, but limited information is available for detailed temporal stack releases of those signatures within the NWVP.

3.4 Introduction – Savanah River Site (SRS) F- Canyon Dissolver

Typical PUREX feed consists of uranium metal slugs clad in aluminum. SRS's reprocessing facility (Bldg.221-F 'Canyon') utilizes an annular dissolver which represents an additional nuclear safety measure. The slugs are shipped in buckets from one of the reactor areas (P, K, or C) in a shielded railroad car called a cask car.

The aluminum cladding, or coating, is dissolved by metering in 50% NaOH in the presence of $NaN0_3$. The $NaN0_3$ suppresses the formation and evolution of hydrogen during the dissolving of the aluminum. The alkaline solution of aluminum, called coating waste, is transferred to waste. The declad metal slugs are rinsed twice with acidified water. The uranium is dissolved in two separate cuts with 50% $HN0_3$ and the raw metal or dissolver solution is transferred to an accountability tank for sampling and analyses which become the basis for uranium and plutonium accountability. The dissolution proceeds by maintaining the temperature at the highest value (~80C) while maintaining adequate vacuum in the dissolver for one hour, then raising the temperature to the boiling point of the solution until dissolution is complete.

The dissolver solution is processed through head end in batches. A batch is transferred to a headend strike tank and "struck" by adding gelatin and simmering; this removes silica by forming a gelatin-silica polymer. Silica is removed to prevent formation of solids which cause emulsions in solvent extraction equipment during later processing. The digested solution is transferred to a centrifuge run tank, then to a centrifuge where the polymer and any other solids are removed. The solids within the centrifuge are washed with dilute acid to remove any uranium and plutonium residues, then slurried to high activity waste neutralization tanks; there, the solids are combined with high activity waste, neutralized, and transferred to waste storage. The clarified dissolver solution is received in an adjustment tank and adjusted to the correct valence of uranium and plutonium and acid adjusted for feed to the first cycle of solvent extraction.

The annular dissolver, shown in **Figure 26** and **Figure 27** below, consists of an outer annulus section (approximately 10 by 13 feet), an inner annulus section (8 by 20 feet), and a center section (6 by 20 feet). The lower portion of the center section contains an 8-inch thick concrete neutron shield (5-1/2 by 8 feet), the upper portion is a de-entrainment section. (The two sections are physically separated with the lower shield portion never being in contact with the process materials.) The tank bottom is 1-1/2 inches thick and the sides are 7/16-inch thick; all metal components of the dissolver and condenser are 304L stainless steel.

The outer annulus contains two sets of heating and cooling coils (one lower and one upper) with 350 ft2 surface area per set. All nozzles are connected to the top of the outer annulus. The inner (fuel crib) and outer annuli are separated by a series of metal bars (7/8 inch by 2 inches by 13 feet) spaced 1/2 inch apart. The fuel crib has a grating 13 inches from the bottom to prevent slugs from going to the bottom of the dissolver and to allow room for a uranium heel. An air sparging ring is located at the bottom of the dissolver to aid in the circulation of the solution. There are four charging ports (90° apart) located on top of the inner annular section. A charging chute, with a mechanism to tip the buckets, fits on top of two charging ports at once and directs the slugs into the fuel crib.

A condenser (4 foot OD x 6.5 feet) is attached to the side of the dissolver tank at the top. The condenser tube bundle is sealed with a metal gasket ring. The ring has a bail attached for remote installation. The noncondensable off-gases pass through the condenser to the off-gas heater-reactor. The condensate drains back into the dissolver tank.

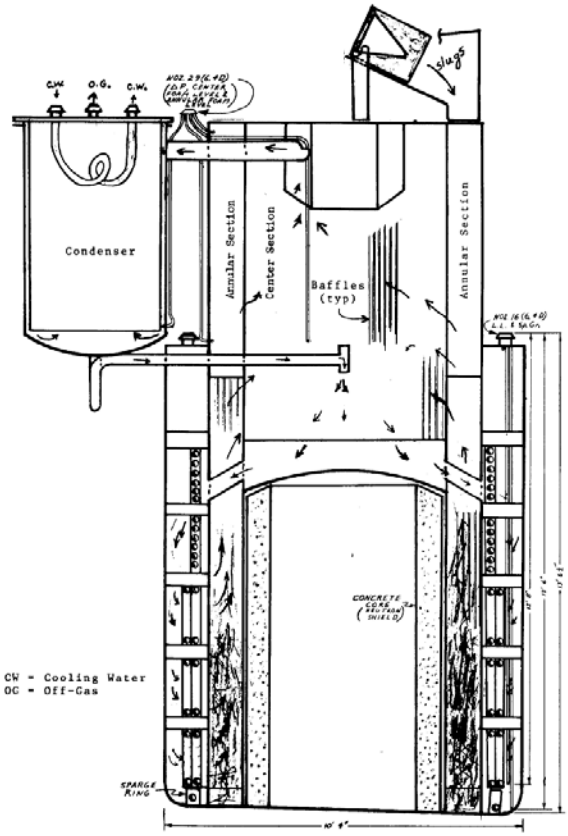

***Figure 26**: SRS Annular Dissolver*

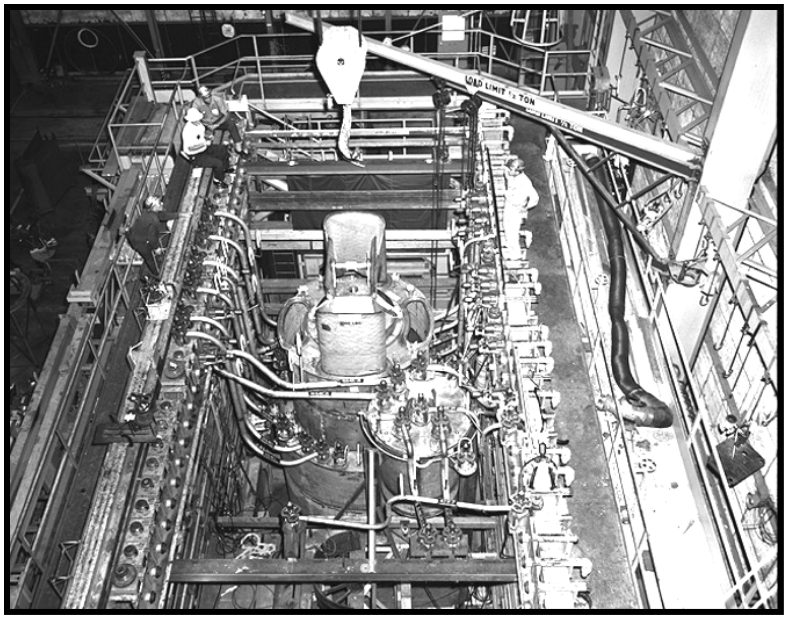

***Figure 27**: Photo of SRS Annular Dissolver in Construction Mock-Up Shop*

3.5 Operation and Dissolution Results

A typical dissolution operation of the SRS Mark 31A (uranium metal) fuel slugs entails many buckets charged into the dissolver. Fuel slugs are usually discharged from a reactor at about the same burnup rate to achieve uniformity of plutonium content. Each bucket of slugs holds about 1.5 tons of uranium. The time for the loading of such a charge including the movement of one bucket from the canyon storage area to the dissolver loading chute takes approximately 1.2 hours. About 5 buckets of slugs are dispensed into the dissolver aqueous solution of $NaNO_3$ (50 wt%) for a full charge. The solution is then heated to boiling point during the period of 1 hour and a metered aqueous solution of NaOH is introduced into the dissolver at about 4.5 L/min. A volume of about 0.15 L per kg of uranium sets the total time and volume of the caustic solution introduced into the dissolver. As the solution is introduced, the dissolution of the aluminum coating starts immediately, hence NaOH is consumed at the same time it is introduced for a period of several hours. The reaction produces H_2, NH_3, H_2O, $NaAlO_2$ and $NaNO_2$ in liquid phase and their corresponding volatile counterparts in the vapor phase. The vapor phase is swept with air inleakage through a bleed-in valve which controls the vacuum on the vapor above the liquid surface. The dissolution process is kept at boiling temperature for about 1.5 hours until the aluminum coating has been fully removed from the double hollow cylinder slugs. At this point, the solution is cooled to 40 ºC within about 1 hour and the contents transferred to a storage tank.

This operation sequence was modeled with the Facility Modeling toolkit by implementing the following operation status of the dissolver module DISSOLVER_ME: '*not-ready', 'receive-fuel', 'heating-cladding', 'base-addition', 'hold-temp', 'cooling-down', 'transfer-cladsolution', 'restart'*. The sequence of statuses and their timing for a decladding simulation of about 7.5 tons of uranium is presented in **Figure 28**.

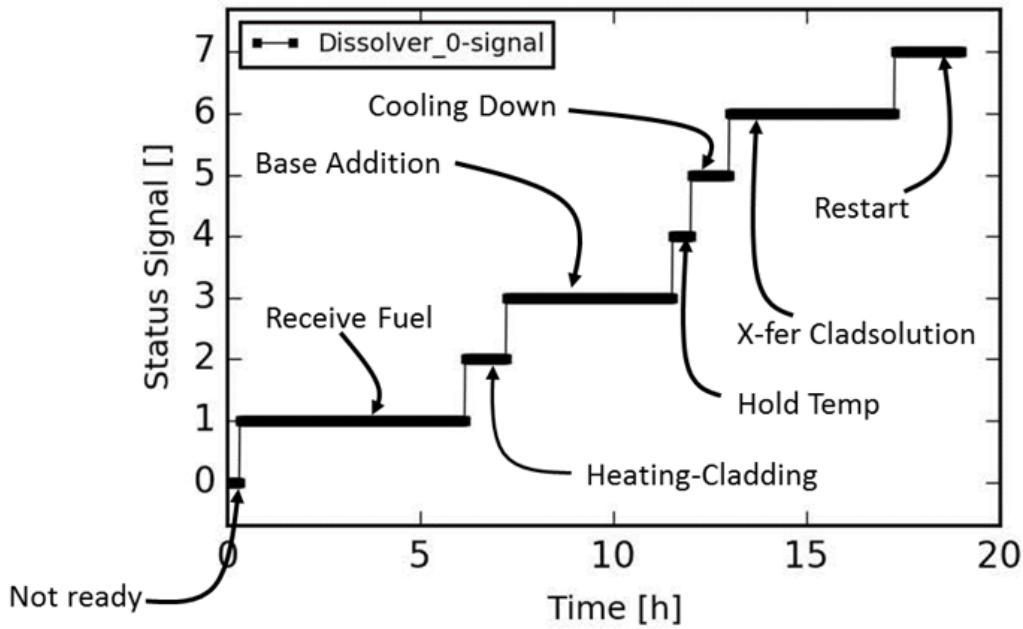

***Figure 28**: Status signals of the dissolver during decladding operation (from left to right): 'not-ready', 'receive-fuel', 'heating-cladding', 'base-addition', 'hold-temp', 'cooling-down', 'transfer-cladsolution', 'restart'.*

The fuel (uranium) mass load into the dissolver and the Al cladding dissolution are presented in **Figure 29**

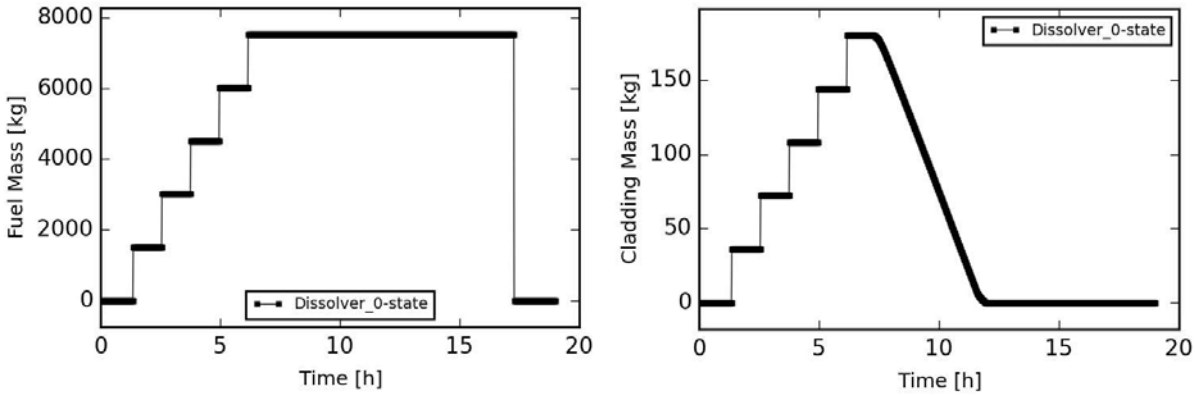

***Figure 29**: Left, uranium load in the dissolver (5 fuel buckets). Right, loading and dissolution of the Al cladding*

.

The feed and consumption of NaOH, and the generation of H2 in the vapor phase of the dissolver follows in **Figure 30**.

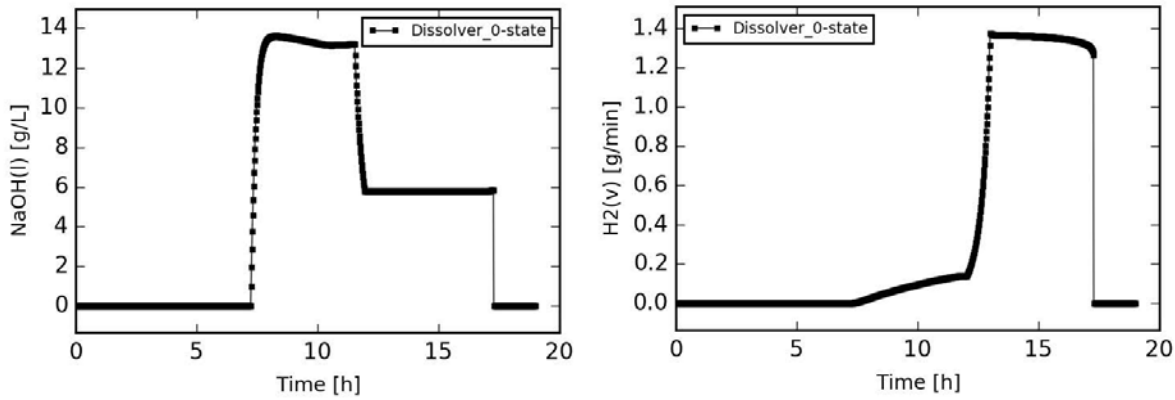

***Figure 30**: Feed and Consumption of NaOH and generation of H2 vapor*

The sodium hydroxide solution is introduced at about 7 hours from the beginning of the operation (left, **Figure 31**). When injection of the basic stream stops it is quickly consumed while the rest of the Al cladding is dissolved. The generation of H_2 gas (**Figure 30**) is reduced by the formation of NH_3 from $NaNO_3$.

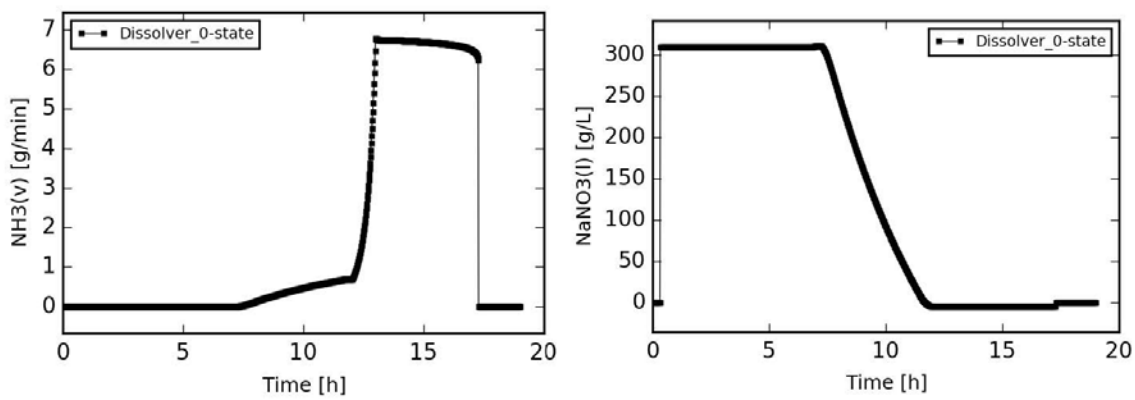

***Figure 31**: Production of ammonia (left) from sodium nitrate (right) to mitigate the production of hydrogen gas*

Following the dissolution of the cladding, a more convoluted operational/chemical process begins to dissolve the metal fuel slugs. At the end of the cladding dissolution, the caustic solution is transferred to a holding tank and the naked slugs are rinsed with water twice to prepare for the acidic dissolution at boiling temperature. The dissolution of the fuel is carried out in two “cuts.” Each cut is separated by a solution transfer of the liquid content of the dissolver and a rinse with water which is also transferred to a holding tank. **Figure 32** shows the combined status evolution of the simulated cladding dissolution and first-cut metal dissolution; note that the simulation was interrupted at 35 h while transferring the dissolved fuel to a holding tank.

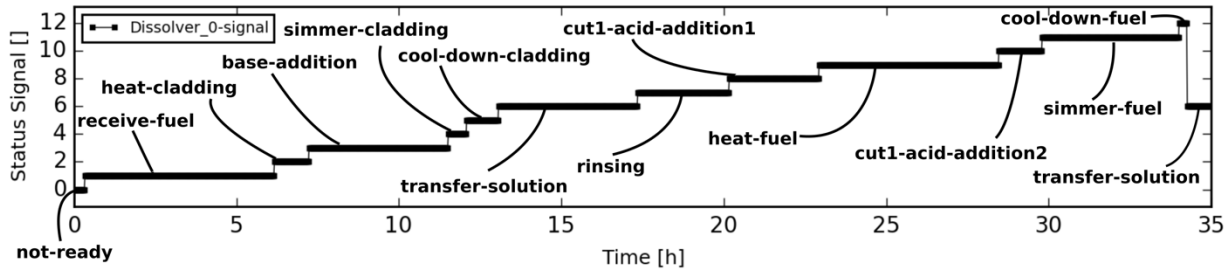

***Figure 32**: Status of a combined simulation of decladding and first cut of metal dissolution. Statuses are as follows: "not-ready", "receive-fuel", "heat-cladding", "base-addition", "simmer-cladding", "cool-down-clad", "transfer-solution", "rinsing", "cut1-acid-addition1", "heat-fuel", "cut1-acid-addition2", "simmer-fuel", "cool-down-fuel", "transfer-solution."*

Simulation results are provided up to the first cut dissolution because the experimental data at hand is only available for the first cut dissolution operation in the F canyon dissolver. Of interest to this work, is the emission of traceable gases. **Figure 33** shows the predictions for a noble gas of interest. The simulation shows 2 major humps. The first, starting at about the 23-h mark indicates the release of gas during the increase in temperature up to the boiling point. The second emission peak at about the 30-h mark is a consequence of the second injection of acid. These results were obtained using various approximated values for many parameters in the model. Experimental data for these parameters are either unavailable or uncertain. A best effort was done to estimate values for these parameter based on various reports related to the DP-500 SRS report. The specific rates in **Figure 33** are dependent on the fuel composition used in FuelDepot. It is uncertain what the particular conditions of the simulated Mark 31A fuel slugs for irradiation and decay were. Therefore, a scaling of the volatile radionuclide content in the fuel composition data at hand was performed before comparison against the experimental data shown in **Figure 34**; the total release of gas was made to match the experimental data (*i.e.* the area under the curve).

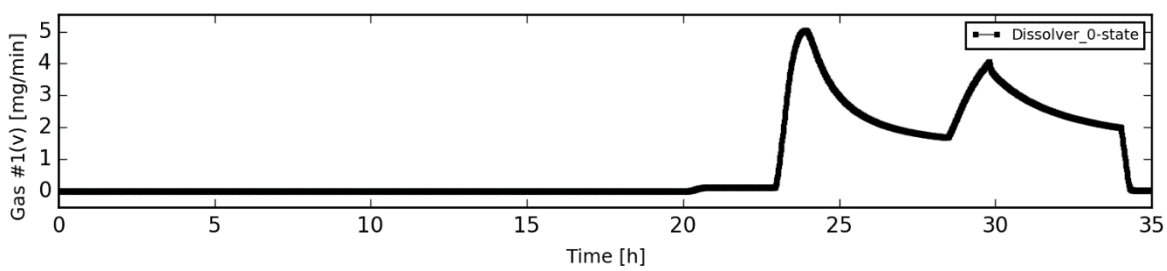

***Figure 33**: Simulated generation of a gas of interest at the dissolver vapor phase outflow.*

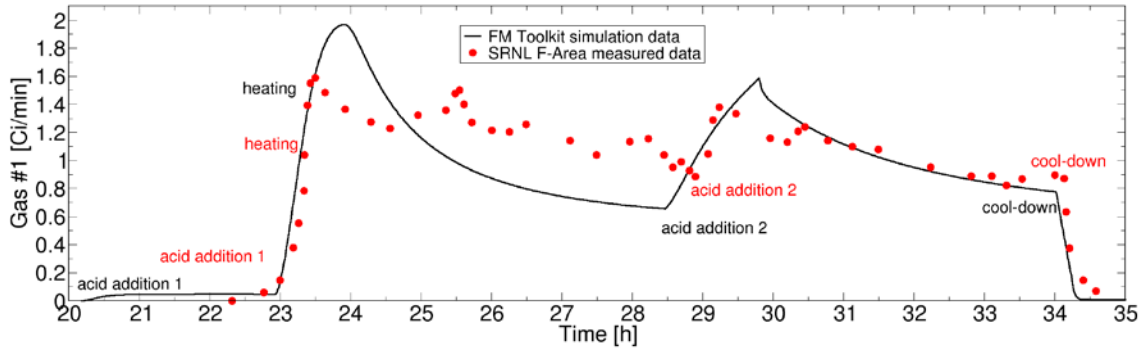

***Figure 34**: Comparison of the simulated data corresponding to Figure 33 against experimental data SRS. The red dots represent measurements at the stack of the facility. Simulated values are shown for the vapor phase at the outflow of the dissolver-condenser unit.*

Comparison of the model results to experimental data for the gas of interest emission at two different points in the facility reveals a good agreement. Details of the dissolution operation for the experimental data presented here are either not available or difficult to retrieve from log books. In the simulation, educated guesses were made based on a typical operation of the dissolver (DP-500); this could have been different from the experimental operation at hand. Nevertheless, the agreement is satisfactory and the model could certainly better capture the experimental data if enough information about the operation and conditions of the experiments were available. When dissolving 7.5 tons of heavy metal (as in this case), operations are often slightly modified to assure safety and these seemingly small changes could lead to patterns in the time-dependent data acquired. For instance, in between peaks (**Figure 34**) the experimental data shows a fluctuation above the model prediction. This may have been due to variations on sparging and/or ventilation (applied vacuum) of the dissolver during the first acid addition which is violent and generates foaming. Here operators may be cautious and make adjustments on the fly which are reflected in the time-dependent data measured. Since the model uses a constant sparging rate and constant vacuum, the resulting model prediction is a rather smooth decaying curve (24 h to 28h). This seems to be the case for the second acid addition after the second peak (31 h to 34 h) wherein model results and experimental data are in much better agreement. We present these results and hope they generate questions about the path forward to improve experimental validation procedures. The operation of a large-scale dissolver is complex and we have succeeded in assembling a computation model with various components that represent the operation. The integration of experimental validation and simulation remains to be exercised with the current computational tool.

4. Toolkit Concept

Facility modeling is the task of deriving and coupling various mathematical models of physicochemical processes associated with the recycling or reprocessing of used nuclear fuel within the facility envelope and its exterior [8], [9]. In view of the complexity and number of processes involved in fuel recycling, the mathematical modeling approach taken here is multi-tiered as shown in **Figure 35**; wherein the top tier consists of a set of spatially decoupled models characterized as:

- reduced modeling;
- time-dependent (time is the only independent variable);
- time-coupled.

This tier is referred to as the plant-level (or facility-level) tier. Specifically, we implement here a particular approach for time-coupling of plant-level mathematical models that allows for building an overall facility model by composition. Therefore the derived computer-aided solution of any facility model can be obtained by coupling computer programs, called **modules**. A module (for short) is the computational realization of a mathematical model of a significant physicochemical process in a particular tier (such as a dissolver/dissolution), see **Figure 35**.

Tiers are formed by levels of theory. We seek to explore mathematical commonality in each tier to:

- re-use a mathematical architecture in different processes;
- enable coupling of consistent quantities;
- share common solution methods;
- incorporate sub-scale calibrations of reduced models.

This enables **modules** to be coupled by time-dependent data, see **Figure 36**, either asynchronously or at a specified synchronized facility simulation time. Furthermore modules are standalone computational units that implement external requirements for coupling with other modules, **Figure 36**. A set of modules combined with a particular coupling strategy defines a facility application. The simulation of a facility model entails the time evolution of the underlying system of equations for each module in the whole facility application.

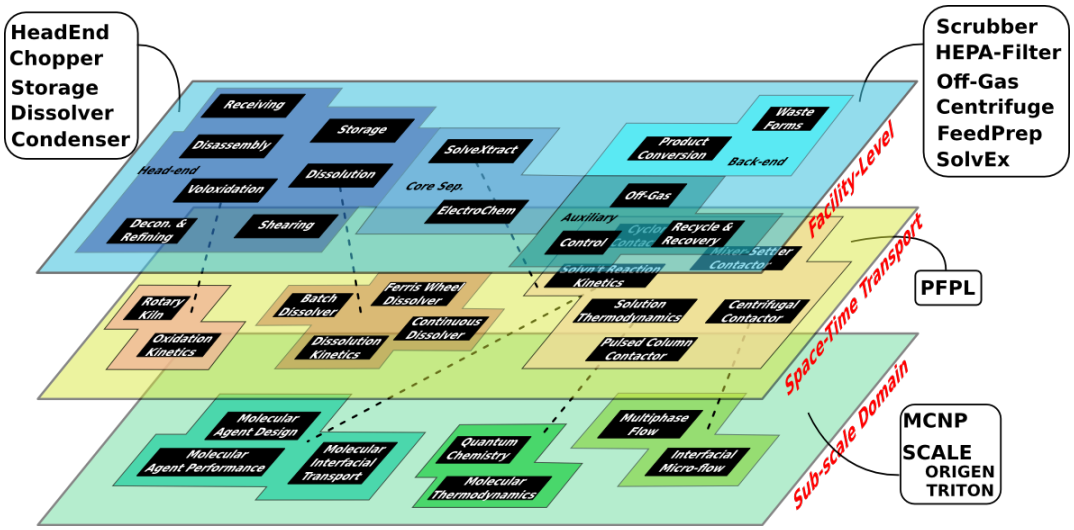

***Figure 35**: Tiered grouping of mathematical models and their corresponding computational modules for a generic facility [10].*

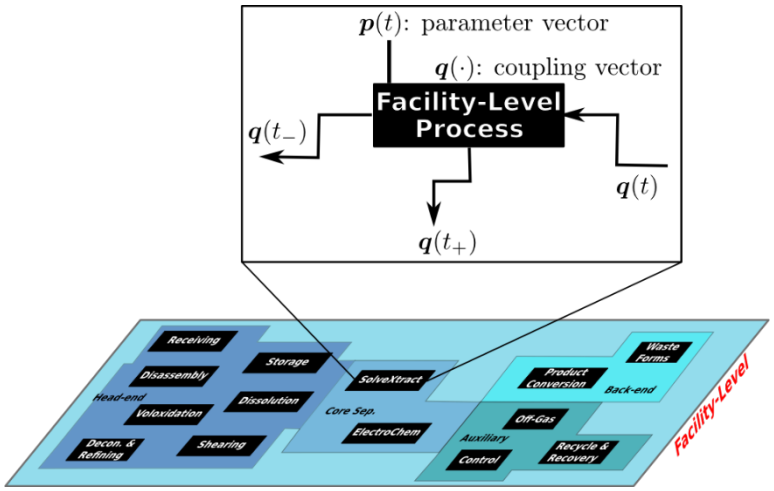

***Figure 36**: Generic facility-level computational module and its external time-dependent data. Modules exchange two types of time dependent data: parameters and coupling. The former includes all necessary controls of the underlying model including signals [10].*

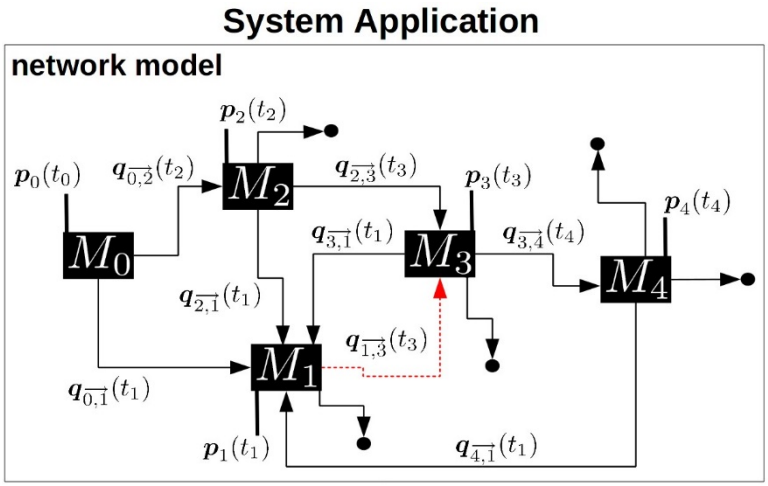

***Figure 37**: Facility application with network-coupled modules. A choice of modules and network connectivity defines a facility application. Modules couple through time- stamped data at different facility times. Therefore a wait mechanism synchronizes* t_i *[10].*

4.1 Design of the FM Toolkit

The most expedient form of implementation of a facility application, that is, a computer-aided program that computes a solution for a facility model, is a *toolkit approach*. A toolkit usage entails, first, the composition of a particular realization of a facility referred to as an *application*; second, the resulting application is used as a program to solve the underlying mathematical problem using computational methods. The intended usage calls for an *application developer*, and an *application user* where the former uses the toolkit to build an application and the latter performs simulation analysis using the application, see **Figure 38**.

The process of performing a facility simulation entails two steps:

1. build an application that models a desired facility type;
2. use the application to perform simulation studies.

A facility toolkit software consists of two components:

1. a collection (library) of facility-level modules;
2. an integration environment used for
    a. coupling modules to create an application,
    b. perform simulation studies (tasks) using an application.

The development of a facility toolkit software is central to the FM project since it enables the modeling and simulation of generic facilities.

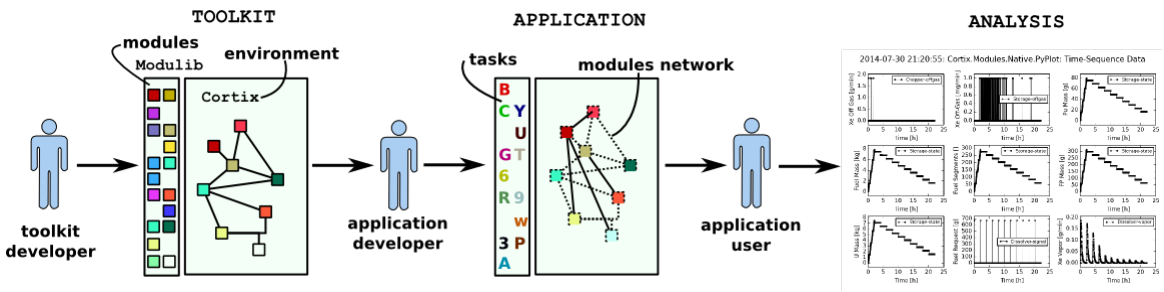

***Figure 38**: Toolkit development and usage workflow.*

4.2 Facility Level Modules

Modules are computational units that realize a mathematical model of a significant facility-level process. These modules must possess external behavior, shown in **Figure 39** in order to be coupled into a network of modules. This defines the architecture of the modules as follows:

Module M_i advances its own time t_i: time integration is module dependent.

1. A module needs a parameter vector $\boldsymbol{p}_i(t_i)$: *i.e.*, all parameters in the underlying mathematical model that may couple to other parameter vectors of other modules via signals or alerts.
2. A module may *require* one or more coupling vectors $\boldsymbol{q}_{j\to i}(t_i)$ from one or more modules: time-stamped data at t_i required from other modules, via the integration environment will force the module to *wait* for data.
3. A module may need to *provide* one or more coupling vectors $\boldsymbol{q}_{i\to n}(t_n)$ to one or more modules either in the *past* or in the *future*: a module needs to watch for a message and provide time-stamped data at t_n required from other modules via the integration environment.
4. A module may be coupled to as many other modules M_k as needed: a module cannot be self-coupled; furthermore, high degree of connectivity will lead to cycles which will require future investigation on how to resolve such cases.

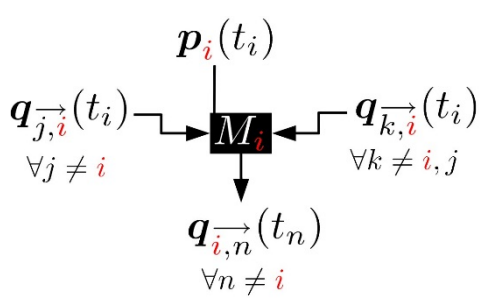

***Figure 39**: External data and behavior of facility-level modules. Modules use and provide data at different facility times. Modules advance time according to their own time integration scheme [10].*

It is instructive to adopt a common mathematical theory underlying the facility modules. This has been advanced in the form of a unifying theory of chemical thermomechanical network transport and a list of questions has been created to guide the communication of model derivation as follows.

1. **Plant-Level Localization.** What plant-level module does the model relate to? If a corresponding module has not been listed in the plant-level tier, propose where it should be placed starting with the sub-system.
2. **Physicochemical Phenomena.** What physicochemical phenomena the model is trying to capture?
3. **System Description.** Describe the system including the principal length and time scales, and how it couples to the outside.
4. **Mathematical Model.** Describe the mathematical model including the simplifying assumptions. For instance, if the model is a partial differential equation set, what are the independent variables, depended variables, domain, boundary conditions, etc. as fully as possible.
5. **Data.** What data is used and what data is provided?
6. **Solution Method.** What is the solution method of the mathematical problem?
7. **Verification Method.** What is the verification method of the solution of the mathematical problem?
8. **Uncertainty.** How is the uncertainty of the solution assessed?
9. **Fidelity Improvement.** How is the model fidelity improved? What is needed and why does the model require higher fidelity?
10. **Model Transferability.** Is the model transferable? If so, what other systems is it applicable to? Apply above questions 1 to 10 recursively.

4.3 Toolkit Environment

The toolkit environment (**Figure 38**) is the component that enables the construction of a network of modules. Therefore its primary function is to connect modules, and to configure them so that parameter vectors $\boldsymbol{p}_i(t_i)$ are properly provided to their modules, and so are the coupling vectors $\boldsymbol{q}_{i\to(\cdot)}(t_{(\cdot)})$ and $\boldsymbol{q}_{(\cdot)\to i}(t_i)$. The environment initializes the modules at the initial facility time $t0$ and executes them. The modules themselves keep track of the facility time as they time-integrate their individual sets of governing equations and either request/provide data at predetermined times or at variable times when needed.

The necessary and sufficient roles for the environment are the following:

1. Build and maintain the network representation of the coupling between all modules;
2. Provide information necessary for the initialization of all modules;
3. Allocate and manage computing resources for running modules;
4. Launch the execution of all modules;
5. Monitor the execution of all modules.

The foregoing results in three basic architecture elements for the development of the toolkit:

Modularity

Computational modules are stand-alone units that must implement the external behavior required by the toolkit integration environment and synchronization approach. The implementation requirements are minimal and driven by the needs for obtaining and providing data in the course of a coupled simulation.

Connectivity

The connectivity of modules is an enabling operation for coupling the underlying mathematical models. It must not interfere with the results of the scientific simulation, *i.e*, it does not introduce any additional errors or approximations to the underlying models.

Synchronization

Time integration is controlled by modules and their internal algorithms. A facility time evolution is controlled by each module starting from the same reference initial time. The exchange of time-stamped data between modules effectively enforces the synchronization of all coupled modules.

The last two elements combine to form what we call the **coupling** architecture element.

4.4 Integration Environment Selection Criteria

The focus of the FM Toolkit was to develop an architecture for an integration environment that enables the coupling of computational modules. Three main categories were addressed for the development of the FM Toolkit: limiting impact of environment choice on module development, ease of use for the end product, maintainability of the integration environment.

Limiting impact of environment choice on module development

As the goal is to develop the capability to couple diverse models, it is necessary to minimize overhead on developing corresponding modules. A major limitation on the usefulness of the toolkit will be the availability of modules. Two main sub categories were identified within this:

- Modularity: the FM Toolkit allows for models to be developed independent of each other which makes validation and verification of individual components simpler and more reliable.
- Limiting the requirements on module developers: Limiting the requirements allows for module authors to program in the languages they are most familiar with, and allows them focus on the development of the models instead of meeting the requirements of the environment. This capability was not directly addressed and the majority of module development occurs in the language of Python.

Ease of toolkit usage

The end use of the toolkit is to allow the specification of facility models out of a multitude of interconnected sub-models (**Figure 38**). It is desirable to specify the sub-model connections with minimal effort by the end user.

Maintainability of the integration environment

The environment should require little or no modification for including new modules or even different types of modules. The environment should be usable on multiple operating systems with minimal effort. It is envisioned that end users of this framework could be using Windows, Mac, or *nix operating systems. Ideally the behavior of the environment will be identical for users on all three systems with a single code base, and that updates to the operating systems will not require modification of the source code for the environment. While this topic was explored during the development of the FM Toolkit, the integration environment was limited to Linux based systems. To increase flexibility of distribution of the FM Toolkit, the linux based systems are ran on Virtual Machines which can be hosted and distributed on any operating system with no impact to the integration environment.

4.5 Requirements

The FM Toolkit was designed around three major requirements:

Extensibility

The central goal of the FM project is to develop an environment that can grow, so developing an architecture for the environment that is by its nature extensible is important. There are several aspects to this requirement.

Modularity

Modules must be able to be developed with a high level of independence by diverse authors at a number of locations.

Flexibility

Module developers will be domain experts with varying degrees and types of programming experience. It is important for them to be able to focus on the development of their modules and not on requirements imposed by the integration environment. As such module development should be allowed in as many programming languages as possible and without any requirements as to the programming techniques that are to be used. Within the FM Toolkit, modules can be developed with different programming languages and “wrapped” to work with the integration environment, but the easiest module development occurs with Python 3.x.

Scalability

The environment must not require modification to accept new modules. End users of the environment do not have to modify the environment in order to include new modules, and are able to develop and include their own modules.

Access to source code

In order to ensure the extensibility of the usage of the environment it is important that the source code be available for future modification.

4.6 Toolkit Approach

The FM Toolkit consists of two primary components:

1. an integration environment: Cortix,
2. a library of modules: User developed and specified.

Cortix has two roles: coupling of modules into a network and execution of tasks associated to a desired analysis. The coupling of modules into a network forms an application which is ready to be executed. Various studies can be done with an available application including variations in the network connectivity. Cortix has seven basic classes (**Figure 40**), used in composition programming style. Their roles are as follows:

Cortix

Creates one or more Cortix objects with their own configurations allowing for multiple instances of Cortix.

Simulation

Creates the overall simulation for each Cortix object, by holding one Application object and a set of Task objects.

Application

Holds a set of Module objects and a set of Network objects.

Task

Creates objects that execute a run.

Module

Builds module objects and their data transfer ports, and defines how modules are executed in parallel (shared memory mode).

Network

Defines the connectivity of a set of modules for each network object in the Application object.

Launcher

Loads every computational module at runtime, spawns one thread per module, and executes all Cortix drivers by incrementing the facility time for all modules; this is the “beating heart of Cortix.”

CortixDriver

Advanced programmable interface (API) for guest modules. All public methods of this class need to be implemented following the concepts of connectivity in the Cortix approach.

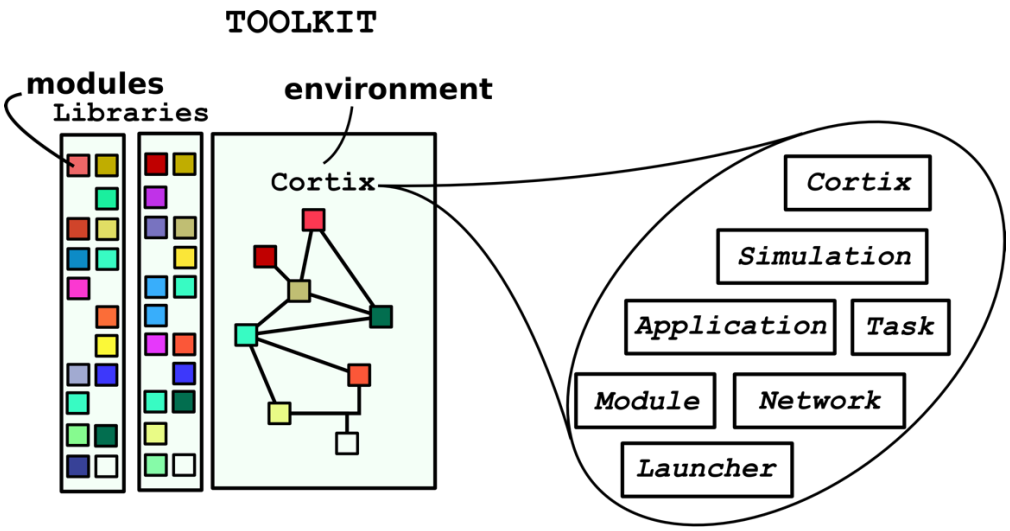

***Figure 40**: Cortix principal classes. Sketch of Cortix components.*

In order to satisfy the modularity and connectivity architecture elements for coupling modules the Launcher executes any module through the common CortixDriver API. For example, guest modules can be of any type as follows:

Stand-alone modules

This type of module has minimum requirements from the CortixDriver. The guest module developer needs to implement two kinds of data *ports*: *use port*, which obtains data needed by the module; and *provide port*, which makes data available to other modules. A stand-alone module is an executable file and the developer can implement the Execute method of CortixDriver as system call execute. This module type allows for use of legacy guest codes when the original developer is no longer available.

Native modules

This type of module integrates directly into the CortixDriver API. The development of the module must be in the same programming language as Cortix (Python) and modules will be executed by threads. This is the preferred method of new module development using the FM Toolkit.

Wrapped modules

This type of module support is useful for codes written in different programming languages where some of the API functions are used for code development. For example, the Cooling Tower module code CTTOOL is a FORTRAN based code that was “wrapped” to work with other modules for the NWVP effort.

4.7 Toolkit Implementation

Cortix is configured by one single XML file (say, cortix-input.xml) with various parameters needed for six basic classes of Cortix (Simulation, Application, Tasks, Networks, Modules, Launcher). Cortix is written in Python 3.x language (using its libraries) and implements communication between modules via file input-output (IO). In-memory data exchange fits the design model of Cortix but it is not implemented at this time. Three preferred file formats are supported at this point: XML, tabular, and Python shelve; the parsing of these file types requires Python libraries.

A key configuration of an application created by Cortix is the communication pattern between modules. To this end, Cortix defines the following files:

> cortix-param.xml: this XML file is created by Cortix for each task and must be read by all modules involved in the corresponding task.
>
> cortix-comm.xml: this XML file is created by Cortix and specifies what the ports of a module need to do to obtain or provide data. One file per module is created by Cortix.
>
> module-status.xml: this XML file is created by modules and expected to be read by Cortix so it reports on the overall running status of the application.

Cortix creates the output file directory for all tasks of all simulations. The above-mentioned files are written in their proper directories, and all temporary data output generated in the course of various tasks (runs) executed by Cortix are organized in a directory tree.

Cortix uses the Python logging facility and it reports extensive logging information for its components and detailed logging information for native modules through the CortixDriver class. All logs are kept organized in separate files by Cortix in the running working directory. These files are invaluable source of information of the flow of data exchange between modules.

5. Documentation of Modules

Table 2 below is a current listing of the modules developed in the FM Toolkit. The list indicates the current status of the module, its intended purpose, and the particular fuel type being addressed.

***Table 2**: Listing of FM Toolkit internal and external unit operation modules.*

ID	Function	Fuel Type	Module Name	Process Involved	Status (ID)
Internal Modules					
1	Fuel Storage	NA	FUEL_DEPOT	Used fuel specieation. Storage of Fuel, awaiting reprocessing, sends and receives signals from CHOPPER	operational
2	Decladding	LWR, CANDU, etc.	CHOPPER	Mechanical shearing of fuel pins	operational
3	Dissolution	oxide	DISSOLVER_OX	Chemical dissolution using HNO3 plus heat	operational
4		metal	DISSOLVER_ME	Chemical dissolution using NaOH, and HNO3 plus heat (pot design)	operational
5	Condenser	NA	CONDENSER	Condenser for dissolution (condensers on evaporator and fractionator are included within the respective module)	operational
6	Dissolution Storage	NA	STORAGE_TANK	Storage of dissolution products	operational
7	Feed Preparation Tank	NA	FEED_PREP_TANK	Feed preparation tank before solvent extraction	operational
8	Scrubber	NA	SCRUBBER	Off gas treatment	operational
9	Solvent Extraction	NA	SOLVENT_EXTRACTION	Solvent extraction pulse columns	operational
10	Raffinate Storage Tank	NA	AQUEOUS_STORAGE_TANK	Tank for the storage of the raffinate from the solvent extraction module	operational
11	Evaporation	NA	EVAPORATOR	Acid Recycle	operational
12	Cooling Water Return (CWR) mixer	NA	CWR_MIXING_TANK	Combine the CWR from various other modules before sending into the cooling tower	operational
13	Cooling Water Splitter	NA	SPLITTER	Splitter for sending cooling water supply (CWS) back to various modules	operational
14	Off-Gas Treatment	Na	FILTER	High-efficiency p[articulate (HEPA) filters for radioiodine absorption	operational
15	Off-Gas Treatment	NA	OFF_GAS	Placeholder for any additional off-gas treatment that may be needed	operational
External Modules					
16	Atmospheric transport	NA	PUFF-FM	Species transport from stack exist to fixed downwind receptors	operational
17	Cooling Tower	NA	CTTool	Forced or natural draft cooling of thermal energy by water based cooling towers	operational

The following subsections address each module listed in **Table 2** separately in the order listed.

FUEL DEPOT Module

The FUEL_DEPOT module is used as Fuel assembly storage before reprocessing steps begin. Currently, ORIGEN-6.2 data is used as input into the module for a selected fuel assembly, but the FUEL_DEPOT module is flexible to use any other source of data for fuel assemblies including excel or data files; this is the case for metal fuel irradiation data provided in a tabular format. The FUEL_DEPOT module provides data for the CHOPPER Module when fuel needs to be sheared. FUEL_DEPOT also provides fuel directly to the DISSOLVER_ME module in case of bucket of fuel slugs used in the SRS F canyon dissolver.

Currently, the module works with as many fuel assemblies as needed or as many buckets needed. There are no specifics regarding the geometry of the fuel storage racks within the FUEL_DEPOT module and the amount of assemblies that can be stored. It is also assumed that there is no heat exchanger for cooling within the fuel depot because such capacity will not be needed since the fuel assemblies used have been cooled from 4 to 6 years. The central data structures created in FUEL_DEPOT are the FuelBundle and FuelBucket containers with chemical species in both solid and gas phases. Therefore a simple assumption is made on the swelling of the fuel for computing the volume of the irradiated fuel from the original pre-irradiated fuel data provided to ORIGEN-6.2 or other irradiation/decay simulator. FUEL_DEPOT also provides a simple oxide speciation for fuels that do not provide such information. The oxide species are built from the isotopics list and the following solid phase species: UO_2, PuO_2, MAO_2, FPO_{1-x}., VFP, and RFP. U stands for the combination of all U isotopes, similarly for Pu, MA is the combination of minor actinides, and FP is the combination of all non-volatile fission products. The stoichiometry coefficient x is computed automatically from the original isotopics input. Also, the combined species, MA and FP, preserve the list of isotopes and their molar fractions. VFP stands for volatile fission products and RFP, for remaining fission products. In addition, a gas phase is also created for the plenum gas evolved from the solid matrix. For the case of metal fuel, FUEL_DEPOT creates similar metal species. Since radioactivity of the species is a necessary input information, FUEL_DEPOT will calculate radioactivities of all nuclides in the fuel specification if this information is not available in the original input data.

Fuel Assembly Parameters within the Fuel Depot include:

Table 3: Parameters for the Fuel Depot Module

Item	Units
U-235 Enrichment	Weight %
Assembly weight	kilograms
U mass per assembly	kilograms
Overall Assembly Length	inches
Bundle Array Size	# x #
Fuel Rod Pitch	inches
Fuel Rod Number	#
Fuel Rod Length	inches
Fuel Rod Fuel Length	inches
Fuel Rod O.D.	inches
Fuel Rod Wall Thickness	inches
Fuel Rod Cladding Material	Based on Assembly
Fuel Composition	grams
- nuclide concentrations	
Fuel Gamma	watts
- nuclide gamma power	
Fuel Radioactivity	curies
- nuclide radioactivity	
Fuel Power	watts
- nuclide thermal power	

CHOPPER Module

Head-end mechanical operations lead to the escape of noble gases from the reprocessing facility. Noble gas inventory in the fuel to be processed developed on the burn history. Gas release during plenum puncture depends on the gas transport into the plenum during and after irradiation. The segregation of

fission-gas isotopes between the fuel-solid and the plenum space in the fuel rod is caused by the transport of the fission gases out of the fuel solid and the radioactive decay of the fission gas. The fission gas that has escaped to the plenum space is necessarily older than the gas that is more recently created in the fuel solid, therefore the isotopic vector for a particular fission product element will be different between the plenum and the fuel solid. Gas release during chopping of the fuel is diffusion-controlled. A mechanistic model is used to predict the gas release from the crushed fuel (due to the shearing operations) based on the known isotopic concentrations in the fuel solid, a log-normal particle size distribution of the crushed fuel, and the surface release of the fission product gases from the particles. The gas release from this crushed region then undergoes a diffusive flux from the sheared ends of the segments of fuel rod into the environment. The particle size distribution (PSD) is based on available data from direct observation of spent fuel shearing operations. The PSD is a potential source for signatures of small scale reprocessing. The model for chopping fuel begins with removing fuel bundles from a fuel depot or storage rack. An individual fuel rod is then chopped into many segments. The initial shear produces two fuel segments from a double balanced cutting design and the fuel segments are dropped into the dissolver. The chopper module produces data for the particulate mass, off-gas mass, fuel segment mass composition and size for all fission gases, volatile and non-volatile species at each time segment specified for the reprocessing operation. In addition, decay heat, gamma radiation power density and radioactivity density information for the all isotopes is obtained for each chopped segment. Alpha radiation power density will be added in the future to supply radiolytic source data for downstream modules.

The CHOPPER module is connected upstream to the FUEL_DEPOT module. The CHOPPER module creates a data request to the FUEL_DEPOT module and receives a fuel assembly with geometric and composition specifications. Downstream the CHOPPER module is connected to the DISSOLVER_OX module and the SCRUBBER module. At the data request of the DISSOLVER_OX module, the CHOPPER begins shearing the fuel assembly by grouping fuel rods according to a configuration parameter (say 4 for the NWVP simulation). Fuel segments are sent to the dissolver in a continuous way as long as the status of the dissolver is in the receiving mode. The resultant plenum puncture and gas release data is passed to the SCRUBBER module for processing. Particulates from the shearing action of the CHOPPER module are passed to the SCRUBBER module. Fuel solid composition is determined from ORIGEN-6.2 calculations for a given reactor burn and decay cooling time. Time dependent data is passed at a given time step frequency.

DISSOLVER_OX Module

The purpose of the dissolver module is to estimate the type and time-dependent quantity of relevant gases released to the dissolver off-gas system, and generate the aqueous feed for the separations portion of the flowsheet. The off-gas estimate is based predominantly on modeling two primary mechanisms. First, the rate of spent fuel dissolution which dictates the rate of fission gas release from the fuel. Second, the chemistry of the released fission gases within the dissolver solution and consequent formation of volatile molecular off-gas species. The dissolution rate of spent fuel has been studied and semi-empirical correlations have been developed and published. The chemistry of off-gases

within the dissolver solution is complex and while it has been studied significantly in the past, complete models that include interaction of the many species within the dissolver are few if any.

Two salient features implemented in the dissolver module are as follows: a) the progressive dissolution of chopped fuel segments as they first arrive in the dissolution acid; b) interfacial transport of the disengaging vapor from the dissolution broth. The first features allows for the simulation of the NWVP process where the chopping of the fuel and subsequent loading into the dissolver takes as long as 8 hours to complete. During this time the dissolution of fuel proceeds at ambient temperature but it is sufficient to dissolve about half of the uranium load. Therefore, this is a critical and challenging aspect of the process to be simulated. The DISSOLVER_OX module successfully predicts the initial dissolution of the fuel by evolving the dissolution of each chopped fuel segment fed to the dissolver. At one time there are as many as 4000 fuel segments in various stages of dissolution completion. The module solves for the differential equations with varying number of unknows with time. That is, as a fuel segment is fully dissolved, the unknowns are removed from the set of equations to be time integrated. This is a substantial savings in computational time and the DISSOLVER_OX module carries out this dynamic task automatically. On the second new feature introduced in the dissolver model, the disengagement of vapors from the liquid phase into the gas phase advances a interfacial transport approach so the relaxation time for vapor release can be controlled. This allows for introducing into the model the effect of sparging on the rate of vapor release. Note that sparging is accounted for in the kinetic of dissolution of the solid matrix but it is also important for the vapor release which is of central importance for signature modeling.

Connections for the DISSOLVER_OX module include the upstream CHOPPER module for acquisition of fuel, and downstream the dissolver condenser (CONDENSER) followed downstream by a silver-zeolite filter and then SCRUBBER modules for off-gas, and also downstream the STORAGE_TANK module for accumulation of the dissolved spent fuel.

DISSOLVER_ME Module

The metal fuel dissolution module is very similar to its oxide counterpart. The main difference is the additional decladding dissolution and its caustic chemistry, and the operation of the metal fuel acidic dissolution carried out in what is referred to as “two cuts.” Similarly to the oxide dissolution, slugs are individually dissolved all at the same time creating as many unknowns as there are slugs in the liquid phase. The FuelSlug container manages solid phases (cladding and fuel) information, and geometry information of the double hollow cylindrical slugs as they are consumed in the caustic/acidic solution. Once the Al cladding is fully consumed, the bare uranium metal slugs are dissolved with nitric acid. This step is very similar to the oxide dissolution process in the DISSOLVER_OX module. In fact, the exact same nitrate species are formed in the nitric acid solution in both processes. However, because the metering of the nitric acid occurs by gradually flooding the bare slugs, an account of partial dissolution of slugs as they get wetted by the acid is developed. This is essentially implemented in the same way as gradually dropping fuel segments into a pool of nitric acid as it happens in the DISSOLVER_OX module. The DISSOLVER_ME module has been fully implemented and tested for the Al decladding operation up to the first-cut dissolution process to allow for comparison to experimental data as explained in Section 3.5.

CONDENSER Module

The CONDENSER module was assumed to be a total condenser. The temperature is adjusted such that the vapor/liquid ratio is at the bubble point temperature therefore the distillate is all liquid and is at a specific fixed rate. Future versions of the condenser can include total condensers with refluxed liquid at specified rates, specified reflux ratios, or partial condensers.

The dissolver condenser model uses the same vapor-liquid reactions and transport equations as the dissolver but at room temperature. The basic reaction mechanism allows for the formation of HNO3 under consumption of NO2. The model is still evolving to include a liquid film formation and fission products of interest.

STORAGE_TANK Module

The primary dissolver accumulation tank model includes a vapor-liquid mass transfer mechanism for the generation of vessel off-gas. The source term for mass balance in the vapor-liquid phases is created as a departure from vapor-liquid equilibrium conditions via a relaxation time parameter. Vessel off-gas is generated by both air bubbling and vapor ventilation. The model also accounts for mixing nitric acid from flushes of the dissolver and temperature control; typically cooling of the tank contents.

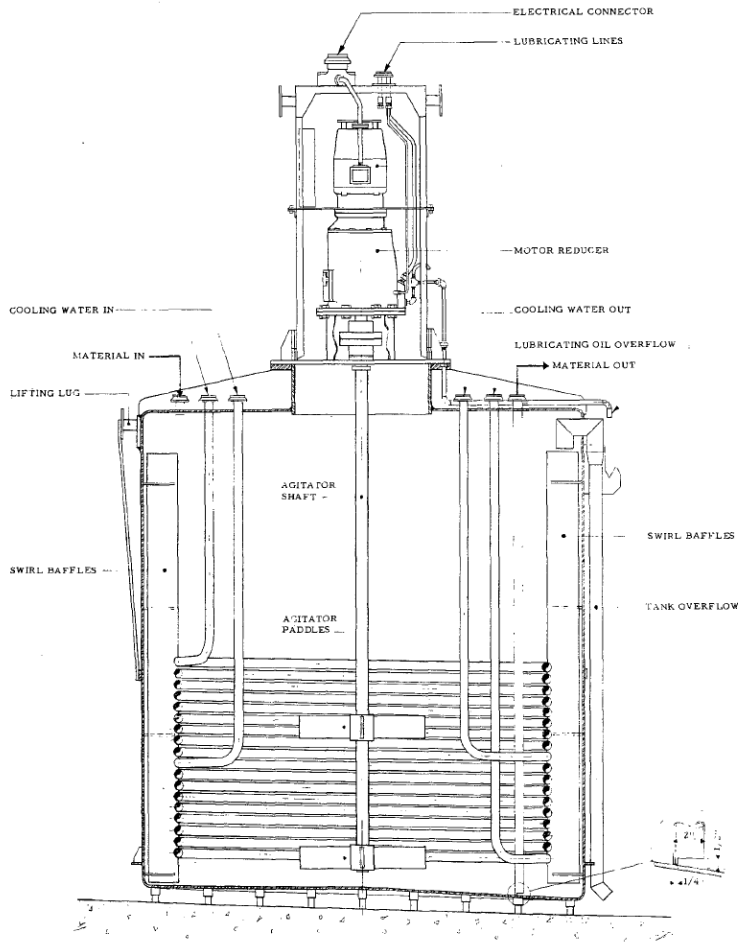

***Figure 41**: Typcial Storage Tank*

FEED_PREP_TANK Module (Feed to solvent extraction)

The solvent extraction feed adjustment tank is functionally similar to other tanks but it is advantageous to have a separate module implantation to account for some of the specific operations in the solvent extraction system. The feed preparation tank model includes a vapor-liquid mass transfer mechanism for generation of off-gas via a bubbler, and vessel ventilation. There are multiple additions of nitric acid streams or valence adjustment streams implemented. Also there is temperature control of the feed.

SCRUBBER Module

Models have been developed to account for both particle collection and gas absorption (to include chemical reaction) for a reprocessing facility off-gas system. These models include a scrubber unit for both particle collection and gas absorption, and a high-efficiency particulate (HEPA) filter for additional particle collection following the scrubber. Particle collection in the scrubber is based on absorption into

a liquid, and in the HEPA filter adsorption onto the filter media surface, and in both cases is predominantly dependent on the particle size. The gas absorption in the scrubber is based upon (1) gas solubility in the scrubber liquid and (2) chemical reaction between the gas and scrubber liquid. For both particle and gas, the degree of absorption and adsorption have been defined here as efficiencies for the "high-level" model. The efficiencies are then dependent upon the scrubber design, gas solubility, and reactive chemicals in the scrubber solution.

The SCRUBBER module is connected upstream to many modules including: DISSOLVER_OX, CHOPPER, and SOLVENT_EXTRACTION. Future upstream modules will include the EVAPORATOR and FRACTIONATOR modules. Downstream the SCRUBBER module is connected to the FILTER module. The scrubber receives data, in the form of time dependent concentrations of species and volumetric flow rate from the upstream modules and based on the scrubber efficiency provides concentrations of species to the FILTER module.

SOLVENT_EXTRACTION Module

The purpose of the SOLVENT_EXTRACTION module is to estimate the type and time-dependent quantity of relevant gases released to the vessel off-gas (VOG) system. As defined here, the SOLVENT_EXTRACTION module includes a co-extraction/decontamination step, scrub step, partitioning step, and finally a strip step. For some separation arrangements additional extraction and strip steps are added to achieve higher purity Pu and/or U product. The co-extraction/decontamination serves to co-extract both the U and Pu from the aqueous-based dissolved spent fuel solution into an organic-based solvent, thereby achieving decontamination of the U/Pu by leaving behind the fission products in the original aqueous-based dissolver solution that becomes the High Level Waste (HLW). The scrub serves to further decontaminate the U/Pu in the solvent by contacting with aqueous-based moderate-strength nitric acid, which is then passed on to the co-extraction/decontamination step to be included in the HLW. The partitioning step serves to separate the U from the Pu in the solvent by contacting with an aqueous-based "reductant" that reduces the Pu in the solvent from the +IV oxidation state to +III. The Pu^{+III} state is not soluble in the solvent and then passes to the aqueous-based reductant phase while the U remains in the solvent. And finally, the strip step involves contacting the uranium in solvent with very low acid (nitric) water. This low acidity tends to reverse the complexation reaction required for uranium (or plutonium) solubility in the solvent, thereby causing the uranium to move to the dilute acid solution, see below. This type of reaction can be thought of occurring at the interface of the immiscible aqueous and organic solutions in a well-mixed vessel.

$$[UO_2^{+2}]_{aq} + [2(NO_3^-)]_{aq} + [2TBP]_{or} \leftrightarrow [UO_2\,(NO_3)_2.2TBP]_{or}$$

In each of these solvent extraction steps, the formation of volatile species will depend on the specific reagents available for reactions, and radiolytic and hydrolytic degradation of these reagents. Radiolytic degradation will consist of both alpha and gamma radiation, alpha principally being due to the actinides and gamma due to the fission products. As such, gamma degradation occurs almost exclusively in the

co-extraction/decontamination step and alpha occurs in all but the uranium strip step. Hydrolytic degradation will be greatest where the nitric acid concentration is the highest, that being the co-extraction/decontamination and scrub steps. Consequently, the type and time-dependent quantity of relevant gases to the VOG system will be different in each of the solvent extraction steps, and dependent on the residence time and temperature in each. While significant modeling has occurred in the past for estimating plutonium losses and decontamination in a solvent extraction system, little has been done for formation of off-gases.

Connections for the SOLVENT_ EXTRACTION module include the upstream feed adjustment (FEED_PREP_TANK) tank module where the dissolver feed is adjusted by diluting with water to the correct nitric acid concentration for solvent extraction, and downstream the SCRUBBER module for the off-gases, and also downstream the AQUEOUS_STORAGE_TANK module for accumulation of the fission products and a product accumulation tank for accumulation of the U and Pu products.

AQUEOUS_STORAGE_TANK Module (Feed to solvent extraction)

The AQUEOUS_STORAGE_TANK module was a generic holding tank module with unlimited volume. There was no modeling of agitators, bubblers, motors, cooling water, baffles or a specific size. The raffinate was completely mixed, and at equilibrium before passing raffinate to the evaporator.

EVAPORATOR Module

The evaporator model accounts for the thermodynamic equilibrium of nitric acid and water within the aqueous solution from solvent extraction. The evaporator is modeled as an equilibrium flash vaporization of the aqueous feed and can handle multi-stage evaporation where the liquid stream of one evaporator becomes the feed of the successive one. The gaseous mixture of nitric acid and water is treated as ideal, with the fugacity coefficient equal to one, since the evaporator operates at the sub-atmospheric pressure of 200 torr. Liquid equilibrium between nitric acid and water has a negative deviation from ideal behavior due to an attractive bond, or association with hydrogen bonds. This negative deviation from ideal behavior in the liquid phase contributes to a maximum boiling point azeotrope within the mixture of nitric acid and water. The extent of the non-ideal behavior within the liquid phase is captured through activity coefficients which are determined by the van Laar equation. The van Laar equation was chosen not because it is the most reliable equation for liquid non-ideal behavior, but because it is simple, easily understood, extendable and easily programmed for future models within the FM project. Vapor pressures used are determined by Antione's equation. Other species within the aqueous phase are treated by Henry's law since the species are at a significantly lower concentration within the feed from solvent extraction. The evaporator also contains a total condenser which takes the vapor product and condenses to a liquid at 65 ºC before sending the liquid to adjacent unit operations. Cooling water is used within the total condenser and heat calculations used for thermal signatures are estimated through enthalpy balances and the use of the Shomate equation for nitric acid and water.

The EVAPORATOR module is connected upstream to the AQUEOUS_STORAGE_TANK module and the SPLITTER module. The EVAPORATOR makes a data request to receive the aqueous raffinate from the AQUEOUS_STORAGE_TANK at approximately 194 ml/min and makes a data request to receive cooling water supply (cold water) from the SPLITTER module for the condenser which is built into the EVAPORATOR module. The EVAPORATOR is connected downstream to the CWR_MIXING_TANK where the cooling water return (hot water) from the condenser on the evaporator is passed to a mixing tank to be cooled and resupplied to other modules. The condensed vapor product of the evaporator will be sent downstream to the FRACTIONATOR.

CWR_MIXING_TANK Module (Recirc. Water junction)

The CWR_MIXING_TANK module only received cooling water return (CWR) from the evaporator condenser; therefore the final temperature was the exit temperature from the condenser without mixing of different CWR streams. The CWR whole mixture will be the same before heading to the cooling tower. The volume of the tank will be determined by the validation case. The final temperature of the CWR heading to the cooling tower will be determined through simple energy balances.

SPLITTER Module (Recirc. Water splitter)

The SPLITTER module assumes the incoming aqueous stream is incompressible and that only a pure splitting of this stream occurs into N specified outlet streams. This splitting operation is assumed to be an incompressible adiabatic process and no energy is being employed for the physical splitting process. Thus, isothermal conditions are maintained resulting in very simple mass and energy balance applications.

FILTER Module (HEPA Filter)

The HEPA filter removes particles from the gas stream according to the same mechanisms highlighted in the SCRUBBER Module section: The performance characteristics of HEPA filters has been defined by the U.S. Department of Energy (DOE) for filtration of CAT 1 and 2 facilities, shown by **Figure 16**. This requirements states that only 0.03% of 0.3 microns diameter particulates shall escape the filter. Furthermore, the most penetrating particulates in typical HEPA filters used in CAT 1 and 2 facilities have been found to be slightly greater than ~0.2 microns diameter. **Figure 16** shows a typical particle diameter normal distribution based on (1) 0.03% of 0.3 microns diameter particulates escape filter and (2) ~0.2 microns diameter particulates as most penetrating.

OFF-GAS Module (Additional treatment of off-gas streams)

Currently, the OFF_GAS module is a placeholder for any future off-gas treatment that may be needed beyond the FILTER module.

PUFF-FM Module

The atmospheric transport of a chemical species (either a gas or a particulate) from the stack exit to various fixed receptor locations is modeled using PUFF-FM. PUFF-FM is an upgrade of the existing code named PUFF-PLUME (developed by Garrett and Murphy, 1981). The mass (or activity) release rate (in g/s or Ci/s) as a function of time is supplied by the OFF_GAS Module as input into PUFF-FM. PUFF-FM breaks up this release rate curve into a series of finite puffs and then transports them based on inputted local meteorological conditions (e.g., temporal wind fields).

As input to PUFF-FM a series of fixed receptors are considered where time averaged sampling is employed along with species dependent detector efficiencies.

PUFF-FM solves the three dimensional atmospheric transport equations based on a separation of variable technique pioneered by Llewelyn [11]. The solution is analytic resulting in very fast run times. Numerical Eulerian or Lagrangian based models would take significantly more run time and are viewed to be unnecessary given the level of uncertainties associated with atmospheric dispersion.

PUFF-FM requires input such as:

- Species of interest and rate of release input source
- Metrological wind-field data
- Fixed receptor downwind locations
- Detector efficiencies and sampling times
- Temporal stack source term

The temporal stack source term is provided by a connection (one-way coupling) to the OFF_GAS module. The entire history of off-gas release is saved and an off-line simulation with PUFF-FM determines the dispersion of the species in the atmosphere.

CTTool Module

The deposition of thermal energy to the environment resulting from the application of cooling towers is modeled using the CTTool module. Based on the time constants of the internal facility unit operations, a steady-state mass and energy balance formulation for the cooling tower performance is acceptable. Thus, no delayed response to thermal capacity of the cooling towers (due to either load changes or atmospheric changes) is accounted for.

CTTool requires input such as:

- Geographical location of facility and UTC timing
- Metrological wind-field, humidity, and air temperature data

- Type, size, and number of cooling towers in bank
- Recirculation water flow rates and inlet temperatures

Since there can be multiple internal facility unit ops requiring cooling water, a common mixing vessel (junction) has been developed contained in the CWR_MIXING_TANK Module. The returning cooled water from the cooling towers is then split back out to the various unit operations using the SPLITTER Module.

6. Toolkit Distribution and Capability

The FM Toolkit is best distributed and run utilizing a virtual machine (VM). A VM is a separate and independent software instance that includes a full copy of the operating system and application software. By using a VM, users of the FM Toolkit do not have to worry about software dependencies required for the FM Toolkit to run. The VM, and ultimately, the FM Toolkit can be created, optimized and duplicated as needed allowing for the FM Toolkit to be proliferated as many times as needed and having convenient recovery options if a software crash does occur. As with any developed software there are tradeoffs that must be addressed. Using a VM for the FM Toolkit allows for easy maintenance, application provisioning and availability at the tradeoff of a lack of efficiency for accessing computer hardware for computations. Since the FM Toolkit utilizes the integration environment, Cortix, to manage a simulation, which is a network of modules (application) that is defined by a run time (task) a large reprocessing facility simulation simulated for several days can take up to an hour to run on a standard dual core laptop. Additionally, if a user has multiple virtual machines simultaneously running on a host computer, each virtual machine may introduce unstable performance which is dependent on the workload on the system by the other running virtual machines. The ease of distribution with all application software dependencies resolved outweighs the potential negative tradeoffs mentioned.

As discussed in section 4.6, the FM Toolkit consists of an integration architecture, Cortix, and Modules. Modules are coded representations of individual unit operations within a reprocessing facility. There isn't any requirement pertaining to the level of detail required for individual modules. It is up to the user to code and develop modules that represent the level of detail desired for the reprocessing scenario. Currently, a large amount of effort has been used to develop highly detailed physics based modules to represent key unit operations within a PUREX reprocessing facility that produce signatures of interest. These modules reside in a module library where users can connect modules together through the use of ports to represent a reprocessing flowsheet. The FM Toolkit capability is limited to the user's ability to design or use existing modules and connect them correctly through their respective ports.

Once modules are developed, the integration architecture, Cortix, leverages a single configuration file (XML file) to run the simulation. Please refer to section 4.7 for more details. The configuration file "ties" together all components of the simulation and a properly defined configuration file is critical for a successful simulation. The configuration file has various parameters needed for six basic classes of Cortix (Simulation, Application, Tasks, Networks, Modules, Launcher). Although the Cortix design has proven to be enormously flexible and extensible, the process of configuring a Cortix application by directly

authoring its configuration in XML was complicated, tedious, and error prone. Furthermore, authored applications could only be reused by cutting, pasting, and modifying XML files.

As a solution, we have implemented a GUI for Cortix that facilitates the configuration of a Cortix application using helpful forms and wizards as shown in **Figure 42** and **Figure 43**.

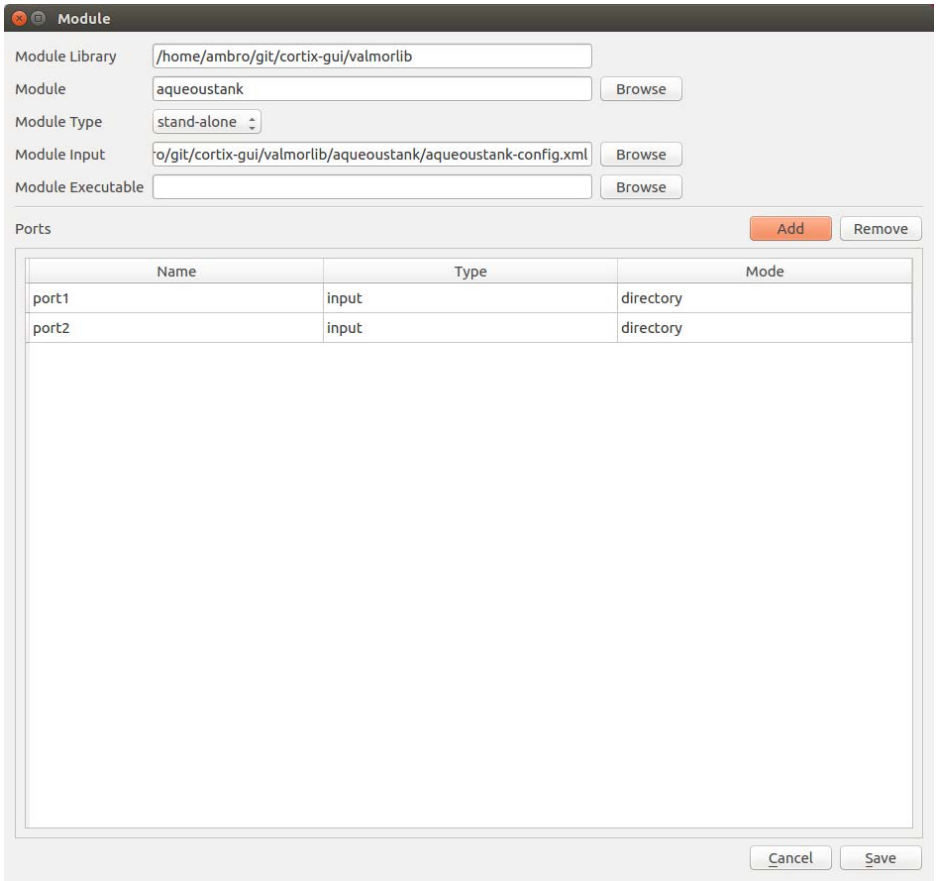

Figure 42: *Module Connection Wizard*

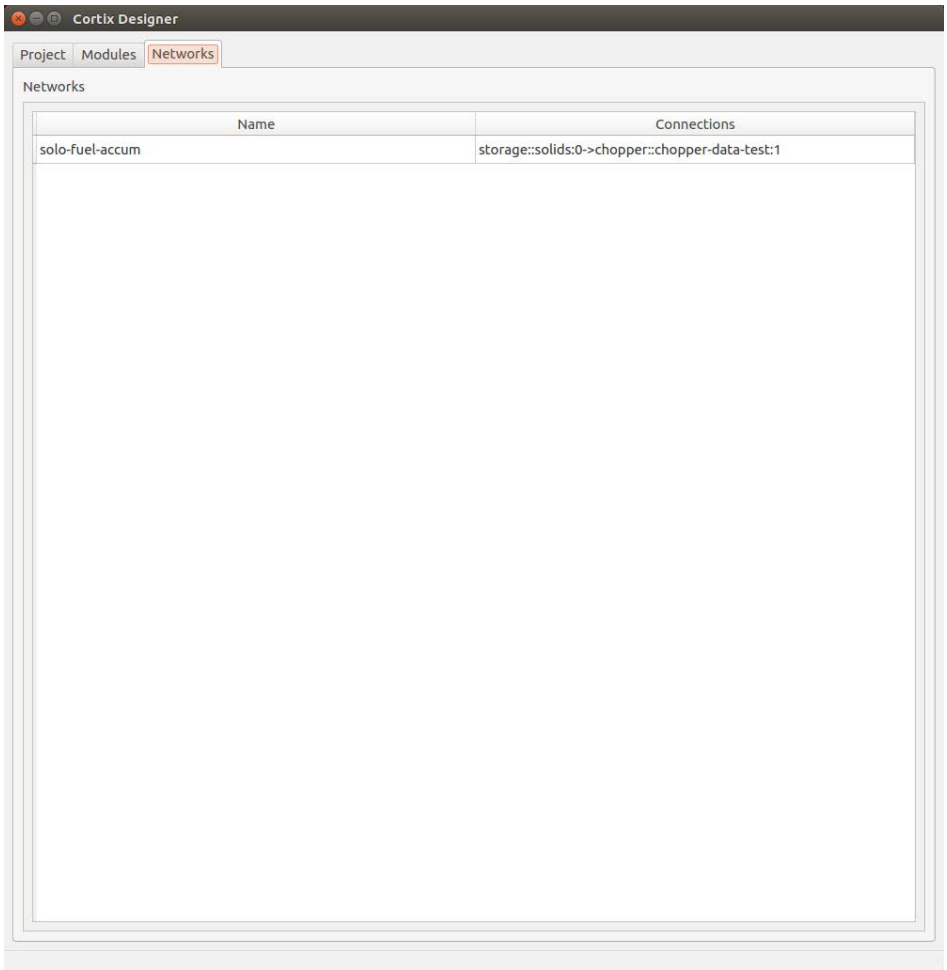

***Figure 43**: GUI for assisting with the creation of the Cortix configuration file, connecting Networks of Modules*

The tool provides information, in the form of tables and diagrams, at each stage of the authoring process so that an analyst can easily validate that the application is being configured is as intended, while avoiding errors that could cause the model to fail in ways that could be difficult to trace. The GUI is also backed by a database that allows analysts to store, retrieve, modify, share, and reuse applications, shown in **Figure 44**.

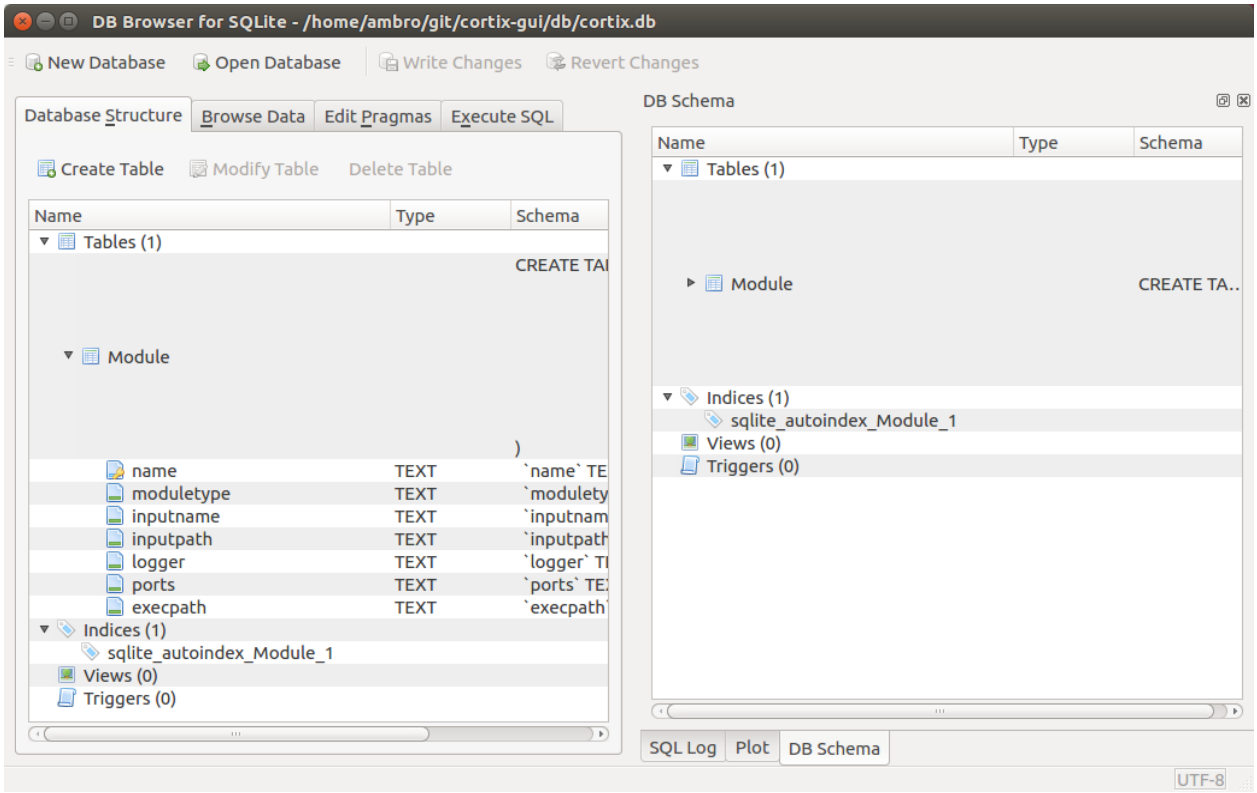

Figure 44: *Cortix configuration file Database*

The development of the Cortix GUI is an enhancement to an already sophisticated simulation model. One that is expected to substantially improve its usability.

7. Conclusion

The goal of the Facility Modeling (FM) project has been to develop and demonstrate a flexible end-to-end modeling capability to accurately model key unit operations in a reprocessing facility. Key model outputs relevant to proliferation detection are the facility signatures i.e. effluents and emissions, and thermal signatures.

A well-developed end-to-end forward modeling capability is an essential element of an overall capability to exploit measured signatures to infer production rates and related information for a facility. An end-to-end modeling capability requires development of individual models for each key unit operation that potentially generates exploitable signatures, as well as an overall framework that serves as a backbone to integrate the individual models. The FM project has developed an initial set of unit operations models: Fuel Storage, Chopper, Dissolver for Oxide Fuel, Dissolver for Metal Fuel, Condenser, Storage Tanks, Feed Prep Tank, Scrubber, Solvent Extraction, Raffinate Storage, Evaporator, Fractionator, Cooling Tower, and Stack and the overall framework that connects them together to produce an end-to-end model for the facility. While the set of individual models can necessarily evolve over time with the

development of additional unit operations models, the integration framework has been developed to be able to accommodate as many additional unit operation models as needed. **In this sense the overall project objective of demonstrating an end-to-end modeling capability has been successfully demonstrated.**

In addition to demonstrating the end-to-end capability, a second objective was to validate the capability using available data sets. We identified candidate flowsheets in the historical US reprocessing database that could be used to validate the models developed within the FM Toolkit. While significant data was located in various documents, we found deficiencies in the completeness of much of the data for potential use in validation exercises. Most facility data was incomplete, lacking detailed operational information, and more importantly lacking data regarding facility signatures of interest. A thorough examination of available data and discussions with plant SMEs at SRNL (H-canyon database) and PNNL (Hanford database) resulted in six candidates being identified for end to end validation efforts. The datasets are summarized in **Table 4**; the red highlighted cases were chosen for the validation and demonstration of the FM Toolkit demonstration. The Hanford Case represented a small scale scenario with sufficient data to validate and the SRS Case represented a large scale scenario with sufficient data.

***Table 4**: Potential End to End Cases for Validation Efforts*

	Large-Scale					Small-Scale
	Campaign Information					
User Case ID	**A**	**B**	**C**	**D**	**E**	**F**
Attribute (fuel)	***1996 (Mk-31)***	***1985 (Mk-15)***	***1975-1977 (Mk-16 & -31)***	***2013 (Danish Fuel)***	***1954-1959 (Mk-1)***	***NWVP (PWR)***
Fuel Type	Al/U	Al/U	Al/U and Al-U alloy	Al/(Al Alloy Silicon Based)	Al/U	Zr/UO2
Important Gases Measured	X	X	X	X	X	X
Feed Characterized	X	X	X	X	X	X
Thermal Data Available	X					
Distance to Receptors			X	X	(?)	X
Thermal Imagery	X					
	SRS Cases					**Hanford Case**
	Validation Case(s)					**Demo + Validation**

The results of the validation exercises were extremely encouraging as a demonstration of the facility modeling capability. Modeling toolkit predictions of signatures compared extremely favorable with measured data for the validation cases. Detailed results of the validation exercises are presented in Section 3.

The overall project objective of demonstrating a capability for modeling reprocessing facilities has been achieved, see Section 3. The forward modeling toolkit has been developed, the capability for integrating individual models has been demonstrated, and the toolkit has been successfully shown to match measured data in two validation cases. In addition the flexible, modular nature of the toolkit ensures that it can be used in the future as a foundation by interested developers or organizations to incorporate additional models for operations or facilities of interest and evolve the toolkit into a comprehensive tool for proliferation detection analysis.

The ultimate end use of such a comprehensive toolkit is to support inferences related to proliferation activities based measured data. In cases where the objective is to evaluate suspect, or clandestine, facilities, measured data will necessarily be somewhat limited. To support this goal, the forward modeling capability that has been successfully demonstrated in this project will need to be coupled with a future capability that quantifies the uncertainty in the forward models. The theory related to predicting uncertainties of computational modules within the FM Toolkit has been developed and demonstrated through preliminary applications to simple unit operations models [1], [2]. The work reported in these papers constitutes the first steps towards a future computational module for characterizing the uncertainties associated with FM toolkit predictions.

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