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Design of an Unattended Environmental Aerosol Sampling and Analysis System for Gaseous Centrifuge Enrichment Plants

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The resources of the IAEA continue to be challenged by the worldwide expansion of nuclear energy production. Gaseous centrifuge enrichment plants (GCEPs) represent an especially formidable dilemma to the application of safeguard measures, as the size and enrichment capacity of GCEPs continue to escalate. During the early part of the 1990's, the IAEA began to lay the foundation to strengthen and make cost-effective its future safeguard regime. Measures under Part 1 of 'Programme 93+2' specifically sanctioned access to perform environmental sampling by IAEA inspectors. During inspections, IAEA inspectors collect environmental swipe samples that are then shipped offsite to an analytical laboratory for enrichment assay. This approach has proven to be an effective deterrence to GCEP misuse, but this method rarely achieves the timeliness goal for high-enriched uranium (HEU) detection set forth by IAEA. Furthermore it is questionable whether the IAEA will have the resources to maintain pace with the expansive production capacity of the modern GCEP, let alone improve the timeliness in confirming current safeguards conclusions on facility misuse. New safeguards propositions, outside of familiar mainstream safeguard measures, may therefore be required that counteract the changing landscape of nuclear energy fuel production. A new concept is proposed that offers rapid, cost effective GCEP misuse detection, without increasing LFUA inspection access or introducing intrusive access demands on GCEP operations. Our approach is based on continuous onsite aerosol collection and laser-based enrichment analysis. This approach mitigates many of the constraints imposed by the LFUA modality, reduces the demand for onsite swipe sample collection and offsite analysis, and overcomes current limitations associated with in-facility misuse detection devices. Automated aerosol environmental sample collection offers the ability to collect fleeting uranium hexafluoride emissions before they are lost to the ventilation system or before they disperse throughout the facility, to become deposited onto surfaces that are contaminated with background and historical production material. Onsite aerosol sample collection, combined with enrichment analysis, provides the unique ability to quickly detect stepwise enrichment level changes within the facility, leading to a significant improvement in timeliness of verification results. We report in this paper our study of a conceptual GCEP environmental sample release and simulation results of a newly designed aerosol collection and particle capture system that is fully integrated with the Laser Ablation, Absorbance Ratio Spectrometry (LAARS) uranium particle enrichment analysis instrument that was developed at the Pacific Northwest National Laboratory (PNNL).

INTRODUCTION

Potentially valuable environmental signatures are continuously lost to the GCEP ventilation system. GCEP uranium emissions not collected by the ventilation system experience rapid diffusion before settling onto facility surfaces at trace levels. Such factors drive the sample analysis requirements toward offsite ultrasensitive laboratory measurements. Samples collected using surface swipes are also difficult to link to precise facility operation timelines. Conclusions from a new sample analysis finding can be mistakenly assumed to have occurred since the last onsite sample collection period, but the finding may have been generated previously. To date this approach has been a very effective GCEP verification regarding the absence of uranium enrichment to undeclared assays, but it is neither cost effective nor timely [1].

PNNL is developing a new GCEP safeguards technology that addresses these problems by providing a means to automatically collect environmental aerosol samples, targeting micron-size uranyl fluoride (UO_2F_2) particulates produced during atmospheric hydrolysis of trace uranium hexafluoride (UF_6) process emissions (i.e., due to minor leaks or maintenance within the cascade hall, process service area, and feed and withdrawal area). Continuous collection serves to acquire samples when and where they are generated (before loss or diffusion) and also provides the ability to timestamp and preconcentrate samples prior to analysis. This approach also significantly relaxes the analysis detection sensitivity requirements and provides a pathway to enable onsite sample analysis. The onsite aerosol collection is combined with an integrated uranium enrichment analysis system based on PNNL-developed Laser Ablation, Absorbance Ratio Spectrometry (LAARS). Our prior investigations have demonstrated the feasibility of LAARS analysis on aerodynamic sized particles of gadolinium, a convenient non-radioactive surrogate for uranium [2], and more recently on uranium-bearing materials [3]. This paper provides the latest results of our modeling, design, and fabrication effort focused on developing a GCEP onsite aerosol collection system based a rotating drum impactor (RDI) design.

ROTATING DRUM IMPACTOR

The conceptual environmental aerosol collector, based on a RDI design, is shown in Figure 1. The RDI was originally developed for remote long-term deployment to collect atmospheric particles for subsequent chemical analysis related to atmospheric science interests such as global climate change [4-6].

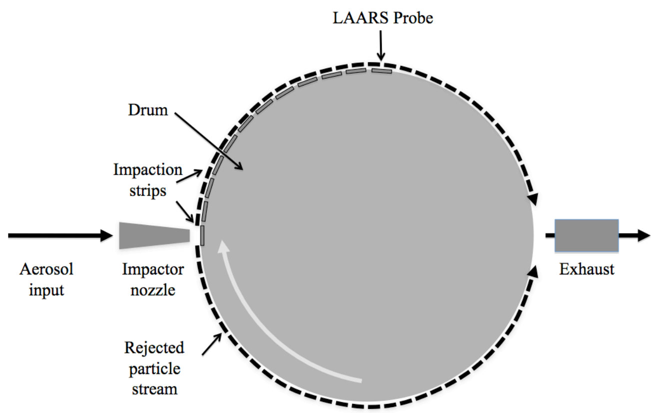

Fig. 1. The conceptual rotating drum impactor. Particles having the correct size range are impacted onto the surface of the drum, while the smaller particles are carried by the airflow around the drum to the exhaust.

In our design a rectangular impactor nozzle directs particle-laden air onto the impactor drum surface. Particles having a specific size distribution will impact and stick to the impaction surface. Smaller particles (typically < 1 μm diameter) will not have enough inertia to strike the impaction surface, but rather follow the airflow around the drum surface to be exhausted from the collection system. The collected particle distribution is optimized for agglomorated uranyl fluoride aerosols having large enough particle size (> 1 μm) to provide sufficient LAARS signal for accurate isotope analysis. The system collects airborne particles for a preset time interval, at which point the drum rotates to expose a new rectangular strip on the drum. A drum having sufficient diameter and height could provide time-resolved particle sampling and integration on a daily or weekly basis over a period of one year. The drum could also be segmented along the axial dimension to separate the collected samples into three equivalent regions, one each for the onsite LAARS measurement, offsite analytical laboratory analysis, and for host state confirmation if required.

In operation, the aerosol collection system acquires facility environmental particle samples for a preset integration period (i.e., day, week). The particle collection is then paused and the sample chamber is evacuated and then backfilled to ~1300 Pa with argon. The rectangular impaction strip is raster scanned using LAARS to detect and analyze the spatial distribution for enriched uranium particles. The drum is rotated one measurement resolution unit (typically 1 to 4 ablation laser spot diameters) and the next line is scanned. This process is repeated until the entire impaction strip is analyzed. Within 10 to 20 minutes, the entire impaction strip is scanned and the U-235 abundance distribution is determined. The sample chamber is then returned to atmospheric pressure to begin the next particle collection cycle.

ROTATING DRUM IMPACTOR MODELING

ANSYS FLUENT (Canonsburg, PA. http://www.ansys.com/) computational fluid dynamics (CFD) software was used to determine design operation parameters for the RDI design. The two-dimensional model used for the flow calculations is shown in the XY plane as indicated in Figure 2. If this model is extended in the Z direction (out of the page) it produces the features of the three-dimensional geometry of the rotating drum impactor critical for modeling. These include a vertical channel for the entrance, a vertical channel for the exit and a cylindrical annulus around the rotating drum. The flow passes through the entrance slit into the cylindrical channel around the rotating drum and out the exit. The two-dimension simplification allowed computational time reduction, while permitting a dense mesh for the CFD calculations to ensure high accuracy. FLUENT is capable simulating the three-dimensional geometry, if required. Flow path segmentation was used to optimize the meshing method for each rotating drum impactor section and to provide the highest meshing density in the critical region around the impactor.

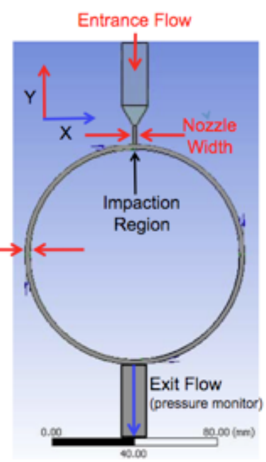

Fig. 2. The two-dimensional RDImodel used in the ANSYS FLUENT computational fluid dynamics analysis. Critical parameters that were evaluated are shown in red.

A steady-state pressure-based model was used with planar symmetry as indicated by the two-dimensional model. The effect of gravity on the x, y, or z direction did not produce any observable effects as expected for these particle sizes and the transit times involved. A discrete-phase model was used to include the particles. As a first approximation, particles were modeled as spheres having the mass of uranium and geometrical diameters between 0.1-2.5 microns. For calculating the behavior of particles in a gas, aerodynamic diameter is the relevant quality. For solid spheres, the aerodynamic diameter is equal to the geometric diameter. Any deviation in morphology results in an aerodynamic diameter that is less than the geometric diameter. This approach has been used to characterize the aerodynamic behavior of soot particles [7]. Here aggregated soot particles were taken as spheres divided by a shape factor that varies between 1 and 3, depending on the extent of aggregation. The same approach was taken for these calculations due to the uncertainty in the composition and morphology of uranyl fluoride particles created during various release scenarios, where a sphere of solid uranium serves as an upper bound for particle shape and density. Particles of lower density with different shapes, including aggregates, can be modeled as uranium spheres of smaller diameter. In our studies, it was found that particle behavior was indistinguishable between particles <0.1 micron, validating choice of 0.1 microns as the lower size limit used in the modeling calculations.

Air was used as the fluid medium, at a constant density and temperature, ensured by monitoring outlet pressure to ensure no pressure drop across the impactor model region. Critical parameters that were varied during the modeling are shown in Figure 1 in red. The entrance flow rate was varied from 5 to 80 L/min, representative of our best estimates for the GCEP application. 5 L/min was selected as a minimum flow rate needed for effective particle transport from a remote sampling location within the GCEP. 80 L/min was selected as an upper bound based on maintaining laminar flow and reasonable sampling pump size and power requirements. Two nozzle widths of 1 and 2 mm were used in the model and three chamber gap distances were evaluated, 2, 4, and 6 mm. FLUENT was first run under a given set of parameters including, flow rate, nozzle width, and cylinder gap spacing, until convergence was reached. The initial particle velocity was matched to the velocity of the air at the inlet and then model calculated subsequent changes in velocity and direction based on the particle parameters and the flow field. The flow rates explored were all within the laminar flow regime, allowing a simple coupled pressure-velocity scheme to be used. The spatial discretization scheme for calculating the flow field in the two dimensional model was least squares-based using standard pressure and second order upwind approximations. The solution convergence was monitored using the residuals and the stability of the pressure monitor at the impactor exit.

The impaction region was defined as a section of the drum immediately opposite and of the same width as the nozzle. Particles were tracked and the flux impacting on this region was monitored. Only initial contact with the impaction region was considered. Neither the particle bounce, nor the matte surface of the glassy carbon surface was considered in this analysis. Prior literature reports have demonstrated that Apiezon grease is effective at minimizing particle bounce [5] and our prior LAARS studies have shown that this grease has no effect on the enrichment measurement. It is therefore reasonable to conclude that if the particles reach the drum surface they will stick.

Data were acquired for one particle size at a time from 0.1 – 2.0 microns, which is circumspect of uranyl fluoride particle agglomeration [7]. 200 particles were monitored for each simulation. Smaller steps were used for the smaller particle sizes to account for the larger change in the ratio of surface area to mass as particle size changes. Determination of the most desirable particle size range will depend on specific deployment conditions of the final instrument. It was therefore desirable to target a particle size distribution likely to be encountered within the GCEP. Additional outcomes of this study were to form a clear understanding of the tolerance requirements for machining the components and the dependencies of the distribution cutoffs. A likely strategy is to engineering a particle size cut-off to exclude any background clutter particles, predominantly below 0.5 microns, that are observed in many environments. We also note here that impactors in general capture all particles above a given diameter determined by the particle and flow characteristics. If desired, larger diameter particles (e.g., > 5 microns diameter) can be eliminated by various means. All the figures shown here assume that all particle diameters > 2.5 microns are captured by the impactor, while not explicitly modeled.

Results from the models of a 2 mm nozzle with a 2 and 4 mm cylindrical gap are shown in Figure 3 (right and left). It is apparent that the flow rate has a significant effect on the particle diameter distribution mean location, as noted by the clear separation between each distribution. This effect becomes more pronounced at lower flow rates and larger gap size. Both of these configurations are strongly dependent on both flow and will required careful tolerance of the machined RDI components. In addition, fairly large flow rates are required to shift the distribution toward the 0.5-micron diameter cutoff regime.

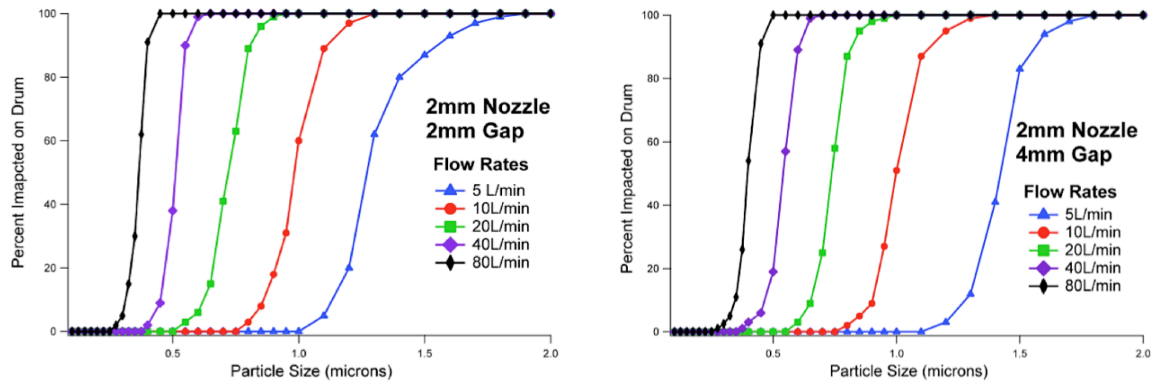

Fig. 3. Simulation results of the 2 mm wide nozzle, having a 2 mm chamber gap (left) and 4 mm chamber gap (right).

Figure 4 (right and left) show the simulation results for a 1 mm nozzle with chamber gaps of 2 and 4 mm. The 6 mm gap produced only minor variations compared to the 4 mm gap, so that simulation is not shown here. It is immediately apparent that this design configuration has a much smaller dependence on both the flow rate and the chamber gap. In addition, very modest flow rates are required to achieve a 0.5-micron cutoff. A nozzle design width <1 mm becomes increasingly difficult to machine, while a nozzle width >2 mm has an undesirable effect on the flow rate. The 4 mm gap simplifies the assembly of the drum within the chamber, without affecting the low flow characteristics.

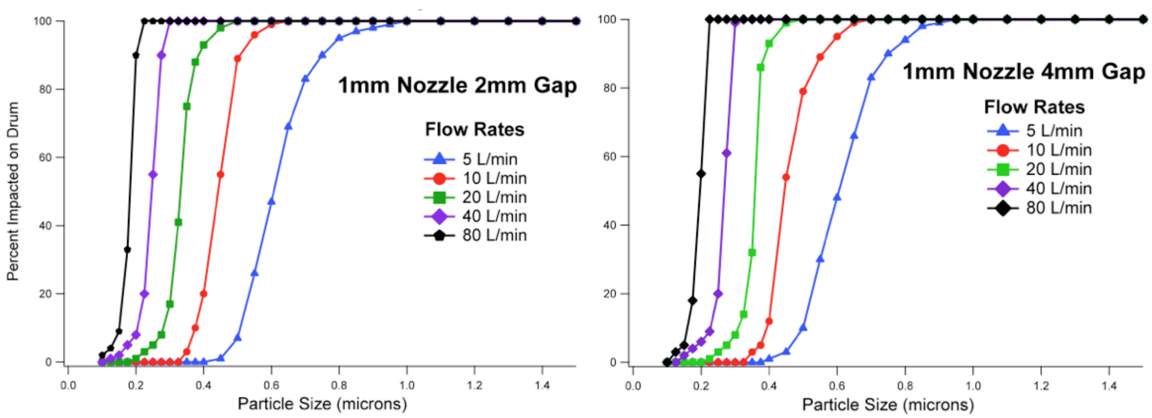

Fig. 4. Simulation results of the 1 mm wide nozzle, having a 2 mm chamber gap (left) and 4 mm chamber gap (right).

Based on this model data, we have determined that for the initial prototype a 1 mm nozzle with a 4mm gap will be employed. This combines the least sensitivity to flow rate variations and machining tolerances for this initial effort. This simplification will allow the initial characterization of particle impaction on a rotating drum made of glassy carbon for comparison with the modeling results. This will allow determination of unknown characteristics of this surface such as particle bounce for subsequent inclusion in the model. The detailed shapes of the impacted particle curve obtained with FLUENT are essential to make the comparisons meaningful. These results also will allow design of a fieldable system with a particle collection range that can be tuned simply by flow rate adjustment to control the collected particle distribution and background particle size cutoff. A complimentary optical method such as laser light scattering could be used to characterize the concentration and size distribution of smaller background particles. It seems clear that in addition to providing starting values for the design of the prototype system, the FLUENT model will enable more sophisticated improvements in subsequent versions, especially when enhanced with the data from our next characterization study. Finally, as designed, the nozzle can be easily replaced if future requirements introduce a significantly different set of sample flow requirements

PROTOTYPE ROTATING DRUM IMPACTOR DESIGN AND FABRICATION

The FLUENT particle flow simulation results were used to develop the RDI mechanical design using SolidWorks 3D CAD software. The rotating drum is formed from a nominal 96 mm diameter by 100 mm length aluminum mandrel as shown in Figure 5 (center). The mandrel surface was machined with a series of 1.59 mm wide channels that serve to retain epoxy during the impactor assembly. A glassy carbon SIGRADUR® G crucible (Figure 5 (left)), having 104 mm diameter, 116 mm height, and 3 mm wall thickness, was obtained from HTW Hochtemperatur-Werkstoffe GmbH to serve as the impactor substrate. Prior LAARS laser vaporization studies, conducted by PNNL, demonstrated excellent performance using glassy carbon planchets of HTW origin. The same glassy carbon, in the form of a crucible, was selected for the cylindrical impaction substrate. While this approach greatly reduces the uncertainty regarding laser vaporization performance, questions remained whether the crucible could be firmly attached to the aluminum mandrel and then be accurately machined. Variation in the glassy carbon outside diameter as a function of position along the rotation axis (i.e., taper) or nonconcentricity of the glassy carbon cylinder and mandrel axis center (i.e., runout) can produce dramatic variation in the energy density of the focused ablation laser beam. This can lead to inconsistent sample atomization that is position dependent. This effect can be difficult to account for during the sample scan, therefore it is worthwhile to minimize it using accurate machining processes and careful optical design.

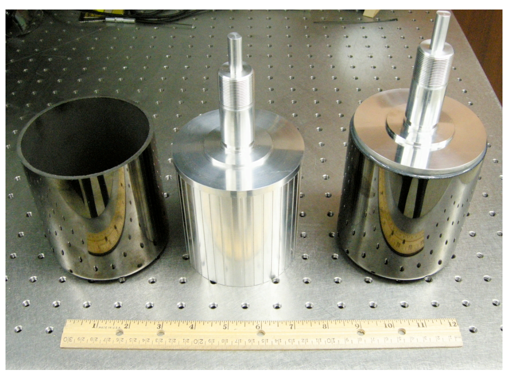

Fig. 5. Glassy carbon crucible (left). Aluminum RDI mandrel with machined epoxy channels (center). Assembled and epoxied mandrel/glassy carbon crucible assembly (right).

The crucible was next attached to the aluminum mandrel using a two-component epoxy adhesive (Master Bond EP21D) as shown in Figure 5 (right). The room-temperature curing property of this epoxy minimizes potential thermal expansion mismatch issues and adhesive property provides a robust bond between the aluminum mandrel and the glassy carbon. The glassy carbon crucible was then precision ground using a glass lathe. This machining process successfully produced an impactor drum with runout and taper within ±25 microns. This exceeded the required accuracy, given the much larger depth of focus (~850 microns) of the focused ablation laser beam. The surface resulted in the desired matte finish, which maximizes the laser atomization efficiency. The final step in the rotating drum fabrication process was to remove the bottom of the crucible.

The rotating drum impactor mechanical design is shown in Figure 6. The assembly consists of a chamber to house the rotating drum. A particle inlet is shown on the left. This inlet receives a particle stream collected from an environment aerosol sampling system located near a likely UF_6 emission source within the GCEP, such as a feed and withdraw station. Particles entering the inlet experience a laminar flow region then pass through the impactor nozzle. Particles with enough inertia impact the glassy carbon drum surface and stick. All other particles are swept around the drum to be removed through the evacuation ports. The rotating drum remains stationary for a prescribed sample integration period then the drum is rotated to expose a new impaction area. Rotation is controlled using a small stepper motor shown in the inset of Figure 6. The design features expansion ports to allow future integration with LAARS for direct uranium particle enrichment analysis. These ports include the argon gas inlet (bottom of chamber), vacuum port (top of chamber), and the ablation window port (far right on chamber). These ports are currently blanks, but will undergo the final machining steps once the feasibility studies are complete. Not shown are pressure and flow sensors, argon gas lines for the LAARS measurement, and a small vacuum pump.

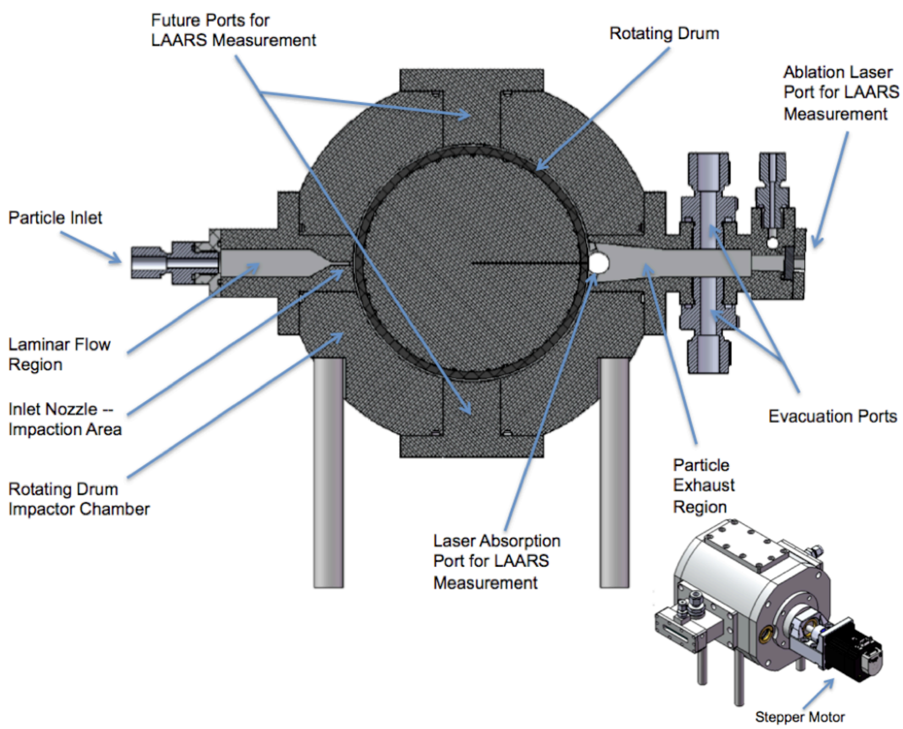

Fig. 6. Aerosol collection system based on a RDI design

The nozzle width is 1 mm wide by 100 mm long (i.e., the drum width and the particle impaction strip will have about the same dimensions as the nozzle. The circumference of the drum is about 320 mm. If each impaction strip is separated by 1 mm, then the drum has about 160 discrete sampling impaction strips. The duration of the particle integration is dependent on the deployment scenario and will require a careful balance between the number of target and non-target particles generated per unit time. In one hypothetical example, integration would capture particles for one week. This could be equivalent to six UF_6 product cylinder withdrawals[1] where uranyl fluoride particles could be generated and then captured by the environmental sampling system. During the entire integration period, background aerosol particles are also collected onto the impaction strip. While the background particles do not generate a false alarm signal, a large build up of these particles, over a long integration period could obscure target particles collected earlier in the integration period. Nevertheless background particle layers can be removed using multiple pulses at from the ablation laser at the same location. This can be repeated until the surface of the glassy carbon drum is reached, as determined by a sharp increase in plasma emission intensity. In this mode the LAARS scanning system can generate enrichment data

[1] Note: The number of product cylinder withdrawals is dependent on the size of the GCEP and is estimated between 6 and 40 per week based on current GCEP production capacity.

spatially in XY as a function of Z depth. In this scenario the drum is capable of collecting environmental particle sample for a period of 160 weeks or about 3 years without intervention.

AEROSOL COLLECTION SYSTEM MODELING

Design and analysis of the environmental aerosol collection system was also conducted using FLUENT software. In this case the aerosol collection system is simply a length of tubing between the given UF_6 leak source and the RDI. It is also assumed that the RDI provides the pumping source to maintain the required system flow rate. The aerosol transmission efficiencies were analyzed as a function of particle size, transmission length, transmission bends, and flowrate. The leak emission and sampling arrangement is shown in Figure 7.

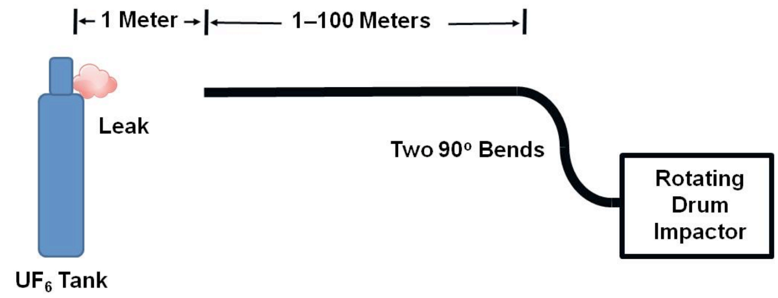

Fig 7. Geometry used for modeling aerosol collection and RDI efficiency

Tubing diameter was maintained as a constant 0.5" I.D., while the flow rate was varied from 5-80 L/min and the straight tube length varied between 1 to 100 meters. This flow range was also used for the RDI modeling, as discuss in the previous section. The tube was oriented parallel to the earth which maximizes the effect of gravity. Although not critical in the RDI design, gravity is the primary loss mechanism for particles in the long sampling tubes due to longer residence time and the lower linear gas and particle velocities. Figures 8 a)-c) show the results from the particle transmission modeling through the section of straight tubing as described above.

a) b) c)

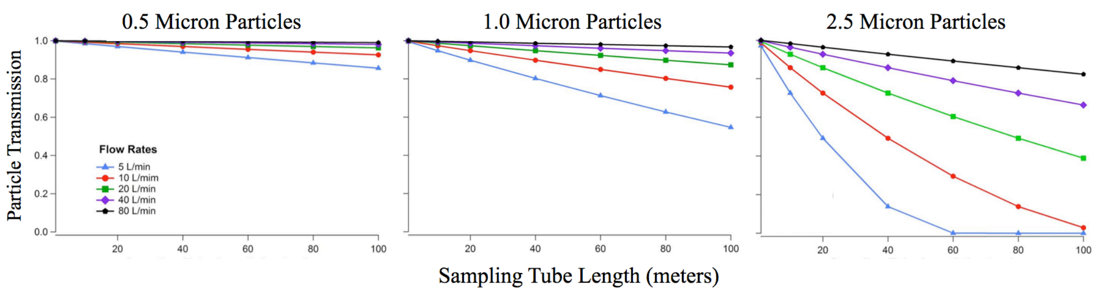

Fig. 8 a-c FLUENT calculations for particle transmission efficiency though .5" I.D. straight tube from 1-100 meters in length at flow rates from 5-80 liters per minute.

The modeling results reinforce an intuitive conclusion to minimize the tubing interconnection length and indicate that higher flow rate is desirable to reduce transit time and therefore gravitational loss. It is clear that even a modest sampling tube length of 20 meters there are substantial losses of 2.5 micron particles and to a lesser extent the 1 micron particles. Smaller losses are observed in general for 0.5-micron particles, which are less subject to gravitational settling. Before concluding that the highest sampling flow rate is most desirable however, it is important to consider losses to inertial impaction at tubing bends likely to be required to interface the RDI. Particle transmission through 2 right angle bends as a function of particle size and flow rate is shown in Figure 9.

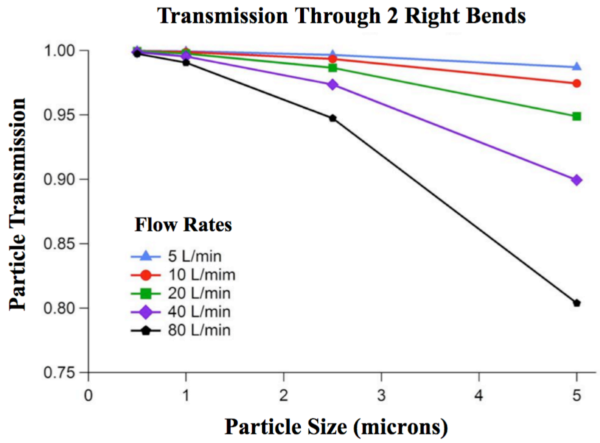

Fig. 9 FLUENT modeling of inertial impaction losses through two consecutive right-angle bends

These results show the importance of considering impaction losses in tubing bends as well as gravitational settling. Larger particle diameters were included in this study to emphasize the difficulty in sampling larger particles and the importance of the high sensitivity of the LAARS approach that allows isotopic abundance determination at trace levels. Particles at 5 micron suffer ~20% loss, while the 2.5 micron particles experience ~5 % loss at 80 L/min through two bends. In a real installation scenario it is likely that more than two bends will be required, so a careful balance between tubing interconnection length and flow rate must be maintained.

It is also important to point out that losses are not only important in terms of LAARS sensitivity, but also cross-contamination between subsequent release events due to large particle disposition within the tubing. A cyclone or similar inertial impaction filter may be required to prevent larger particles from entering the sampling system. Using this approach, we can estimate that particles from 0.5-1.0 micron can be delivered to the RDI using flow rates between 40-80 L/min at around 98% efficiencies, even at 100-meter distances. Shorter sampling distances will increase those efficiencies still further.

It is instructive at this point to use the FLUENT modeling result to estimate target aerosol collection efficiency from a typical UF_6 release scenario, such as a release during UF_6 cylinder feed and withdraw. Figure 8 indicates such a scenario, with a proposed 1-meter distance between the release point and the sampling inlet. Actual GCEP facility design information varies from plant to plant, so an approximate scenario was developed for this modeling study. If details of the ventilation system such as inlet and outlet locations and flow rates were available, a very accurate facility-specific collection system can be designed and evaluated. The maximum OSHA air exchange standard (OSHA 1910.1450 App A) was used (12 room air exchanges/hour) and 1 mg UF_6 release into air was assumed. It is assumed that 100% of the UF_6 is hydrolyzed into uranyl fluoride particles. The volume expansion rate was taken at 1 meter/min to calculate a time-dependent concentration as shown in figure 10.

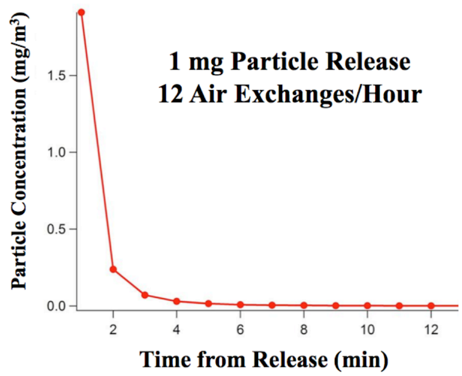

Fig. 10 Particle concentration vs. time for a particle cloud expanding at 1 meter/min, roughly corresponding to 12 room exchanges per hour.

The uranyl fluoride particle concentration is nearly zero by 5 minutes, consistent with one room ventilation exchange. After ten minutes and two air exchanges, the entire leak has been removed from the facility location. This time-dependent particle concentration data was used with the straight tube with bends total collection efficiency to estimate the total amount of material impacted on the rotating drum. This was studied as a function of straight sampling tube length for flow rates from 5-80 L/min, assuming the 1 mm nozzle and 4 mm gap design shown in figure 4. These results are shown in Figure 11 a)-c).

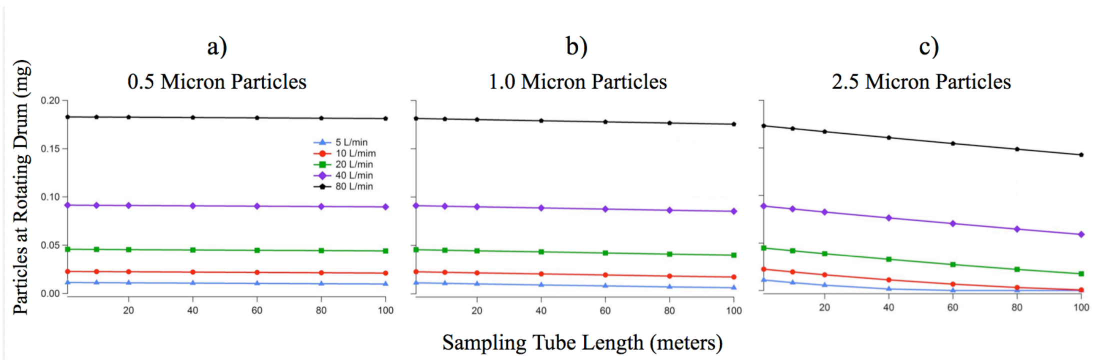

Fig 11 a-c Total particle mass deposited on impactor for the sampling scheme shown in Figure 8

These results emphasize the previous conclusions that particles > 1 micron should be rejected to prevent cross-contamination effects. These estimates also suggest that flow rates between 40-80 L/min are most desirable, mainly because higher flow rates lead to more efficient emission collection before the aerosols are lost to the facility ventilation. This study represents initial steps toward better understanding GCEP environmental aerosol collection dependencies to enable improved collection efficiency and minimize cross-contamination issues.

CONCLUSIONS

We have presented a new approach for detecting misuse within a GCEP that combines continuous facility aerosol collection with onsite uranium particle detection and enrichment analysis. This approach resolves the weaknesses of infrequent sample collection (sample dilution, loss, and timeline uncertainty) and offers the potential to provide timely and cost effective GCEP misuse detection.

CFD modeling of a simplied two-dimensional RDI and aerosol collection design was conducted to study the design interdependencies and the operational characteristics. High flow rates generally were found to provide better particle collection, conveyence, and capture onto the impactor, but are subject to increased tubing bend loss and potential cross-contamination. The aerosol collection system performance is ultimately subject to a careful balance between gravitational and inertia losses. Access to actual GCEP facility design information, such as ventilation inlet and outlet locations and flow rates, would enhance the accuracy of these simulations.

A prototype RDI design was developed and fabricated, based on the modeling results. Future work is planned to evaluate the performance of the LAARS RDI system. The prototype RDI assembly will be exposed to particle streams with known distributions. The impacted particle size distributions will be analyzed by optical and SEM methods for comparison with the model results. This will include the actual spatial extent of the impacted particle area for comparison and incorporation into the model. Real time measurement of transmitted (not impacted) particle numbers and size distributions will be made with a LASAIR II optical particle counter, which measures particles from 0.1 to 5 microns in 10 size bins. The aerodynamic diameter, composition, and density can be further characterized using an aerosol mass spectrometer (AMS) available at PNNL. Data from the LASAIR and the AMS will provide valuable feedback needed to calculate shape factors and to fine tune and optimize the aerosol collection system and rotating drum impactor for morphologies and densities of actual uranium particles found in processing facilities.

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