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NAVAL UNDERSEA RESEARCH PROGRAM FINAL REPORT: SURFACE AND FLUIDIZATION STUDIES OF COATED ALUMINUM PARTICLES FOR IMPROVED FLOWABILITY

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Naval Undersea Research Program Final Report: Surface and Fluidization Studies of Coated Aluminum Particles for Improved Flowability

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ABSTRACT

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[*] Mawby, L. (2021) Surface Properties of Small Metal Particles, [Doctoral Dissertation], The Pennsylvania State University, University Park, PA. (Will be published on PSU ETDA, https://etda.libraries.psu.edu/catalog upon graduation, December 2021).

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1 Introduction

1.1 Background

The flowability of powders is governed by their bulk properties, surface character, morphology, environmental conditions, equipment used as delivery means, and many more factors. This applies especially to micron-sized powders, as inter-particulate, cohesive forces begin to dictate flow when compared to their larger particle size counterparts. In this work, we study the influence of inter-particulate interactions at the surface and bulk levels to understand their behavior in delivery applications. Specifically, we study the flow of metallic aluminum powder.

Metal powders are used in a wide variety of industrial and engineering applications. Aluminum powder can be used in pyrotechnics, propulsion, and other energy applications. That makes its ubiquitous use ripe with opportunity for new research to customize its flow properties. Many of these applications require transfers or direct delivery through fluidization columns, hoppers or pneumatic conveying to sample a few. All of these rely on ideal powder properties for successful transfer; making powder flow properties tailorable presents an important research opportunity. Herein, we study the delivery of aluminum powder through a fluidizing nozzle and how the surface treatment influences the powder's flow properties.

One common surface modification method is the use of silanes treatments on particles and fillers. The use of silane surface treatments has been shown to improve the flow and fluidization properties of aluminum powders in various applications. [1] [2] [3] [4] [5] Ethoxysilanes are available with multiple functional groups, such as aromatics, alkyl chains, amino-based groups, and carbonyl moieties, which range from hydrophilic to hydrophobic. [1] For the purposes of this work, we selected from both hydrophilic and hydrophobic categories to investigate the nature of the inter-particulate interactions in both cases. Alkyl groups, ranging from a single methyl group to a dodecyl chain were studied in addition to the aromatic phenyl group as the hydrophobic silanes. Amino, di-amino, and carboxy based silanes were selected as their hydrophilic counterparts. The diagram in Figure 1-1 shows example methoxysilanes with the high polarity (strong intermolecular forces) and non-polar (weak intermolecular forces). Sample batches with each silane were prepared and studied as a part of this work.

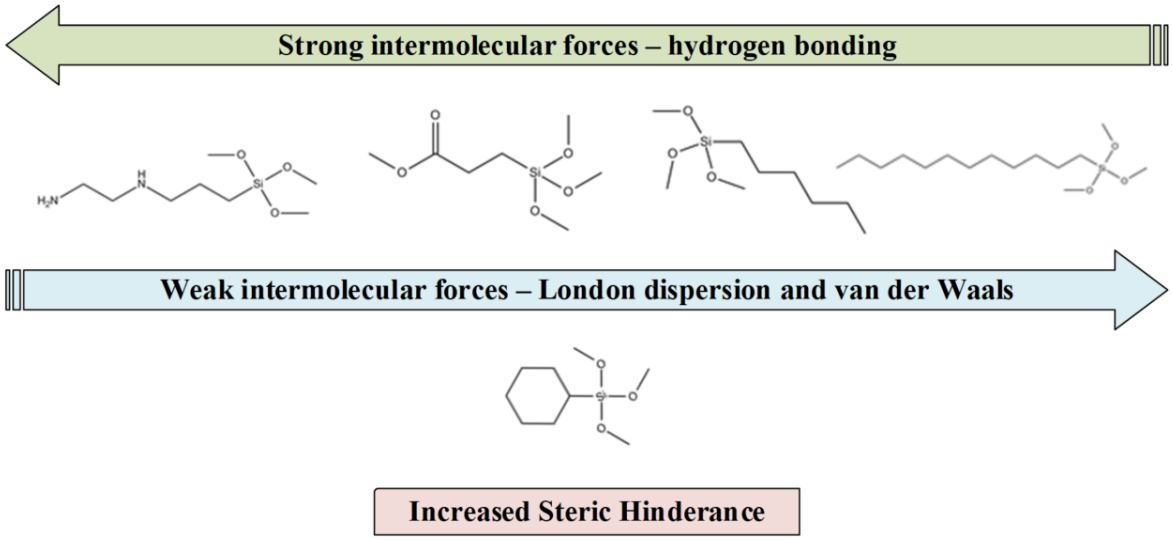


Figure 1-1. Sample methoxysilanes with varying polarity, steric hinderances, and intermolecular forces.

The scale-up of benchtop experiments to the manufacturing scale can be difficult and hard to establish. In this work, we began the down-selection of silane surface treatments at a bench top scale to minimize the amount of waste and reduce the overall experiment time. Although the selection of silane is critical, it is also crucial that the process be scalable to deliver batch sizes large enough for full scale testing. To this end, we used large scale processing equipment at ARL PSU to transfer glassware experiments to a rotary blender. Following the successful preparation of a large scale batch, we were able to proceed with full scale feeder testing, the final goal of our proposed work.

1.2 Objectives and Deliverables

The over-arching goal of this work was to study hydrophilic and hydrophobic silanes as surface treatment agents to improve the flowability of aluminum powder. The project was broken up into three separate research areas:

- Evaluation of hydrophilic and hydrophobic silanes for improved flow properties; selection of silane for further study;
- Fractional factorial experiment of the parameters associated with the solution phase treatment purpose including: humidity, stir rate, and cure time;
- Scale-up from laboratory to pilot scale batch; Flow and fluidization powder testing

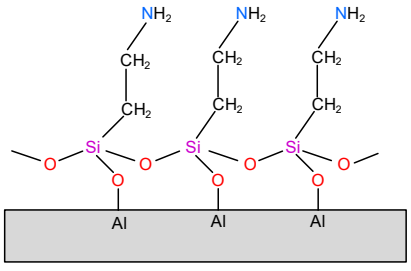
This work was completed as a part of the Naval Undersea Research Program (NURP) from the Office of Naval Research. This program is specifically structured to form a collaboration between a Navy laboratory and a traditional academic setting to educate a graduate student (L. Mawby) in a field of research relevant to the Navy. Following this format, L. Mawby completed the silane evaluation, surface characterization, and fractional factorial study during the first and second years of the project at PSU-main campus. The scale-up of the selected silane, flow property analysis, and fluidization testing were completed during the second and third years of the project at ARL-PSU.

The main hypothesis of this study is that the molecular level properties of the coatings are responsible for the macroscopic fluid properties. In order to investigate this hypothesis, L. Mawby completed a partial factorial experiment screening for the impact of the following parameters: treatment reagent, particle size, age, treatment conditions, and humidity effects. Each of these parameters could influence the final fluid like properties of the particles, as they will affect surface coverage, surface morphology, and coating thickness. Following the results from this work, we scaled to a larger batch scale, capable of full feeder testing.

2 Surface Treatment and Properties: Silane Studies Probing for Interfacial Interactions responsible for Flow Properties

2.1 Introduction

For the work reported herein, we selected hydrophilic and hydrophobic silanes to probe for surface interactions that may be responsible for bulk scale flow properties, both good and bad. The hydrophilic silanes included a diaminosilane and a carbonyl based silane. The amino based silane has the ability to hydrogen-bond with itself and with the particle surface hydroxyls. Research has shown that amino-functional silanes orient themselves such that the hydrolyzed groups react with the surface and the amino groups are left open for hydrogen bonding. This property is useful for dispersion in polar matrices, however, likely influences flow properties by causing agglomeration of the particles. Figure 2-1 shows a simple amino silane, 3-aminopropyltriethoxysilane (γ-APS) with a single amino group, which F. Boetio and J.W. Williams used as a model to study these interactions. [7] For this work, we anticipate that the silane will function in a similar matter. The second hydrophilic silane selected was 2-carbomethoxy-ethyltriethoxysilane (CMES), which has carbonyl functionality. After the initial surface reaction, the carbonyl containing groups also have hydrogen bonding capabilities as shown in Figure 2-1. [8] Due the possible hydrogen bonding capabilities available on these coatings, we anticipate the inter-particulate and hygroscopic properties of the tail groups will cause agglomeration.



(a)

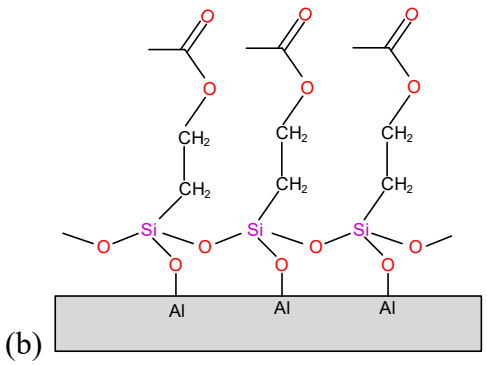


Figure 2-1. Theoretical orientation of (a) simple amino silane showing formation of covalent Al-O-Si bonds from particle surface –OH. (b) Image adapted from reference [7] and [8].

Hydrophobic silanes generally have an alkyl or fluoro group, acting as the non-polar component in non-functional silanes. This allows for dispersion in non-polar mediums, or, they can act as surface coatings to improve flow similarly to our application. Figure 2-2 shows a pictorial which includes the silanes selected for study in our work. This includes arrow indications of the functional components character, from hydrophilic to hydrophobic.

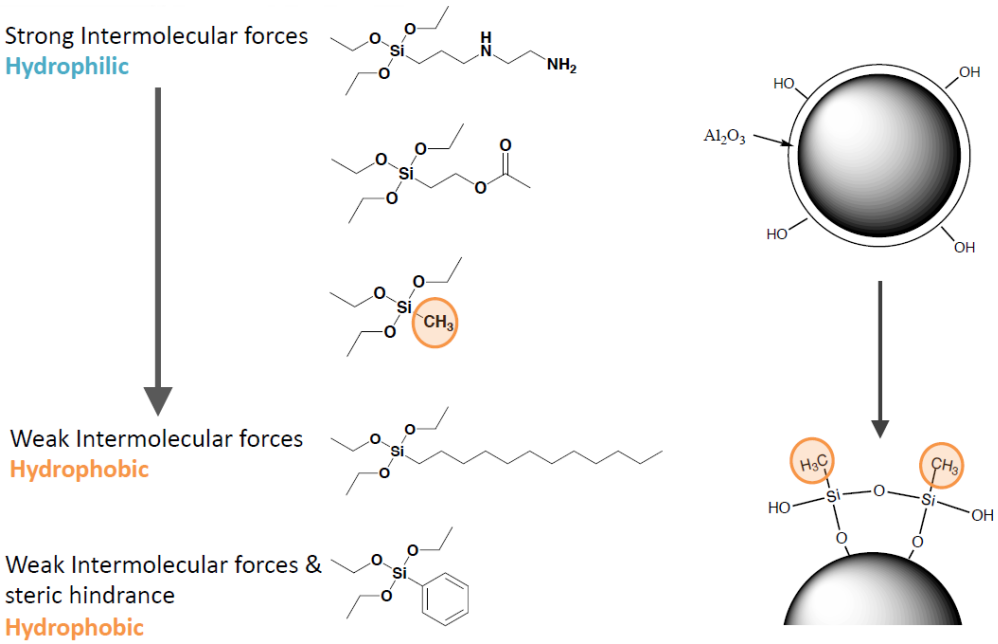


Figure 2-2. Selected silanes, shown with relative polarity scale. Hydrophilic (diamino, carbonyl), Hydrophobic (methyl, dodecyl, phenyl).

2.2 Results of Silane Selection

We used small scale testing to down-select a silane for future, larger scale processing and fluidization testing. All of the silanes were successfully deposited on the aluminum particles in the laboratory. To verify this, we used diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) to study the surface moieties present after treatment. Figure 2-3 shows the IR spectra for the aluminum powder samples treated with each of the selected silanes. In most cases, the presence of the Si-O-Si stretching vibration centered on 1150 cm^{-1} verifies the silane retention as raw powder does not contain any Si-O-Si. In the case of the hydrophobic silanes, aliphatic pieces in the $–CH_3$ deformation (~1265 cm^{-1}) and –CH (3000 – 2800 cm^{-1}) stretch regions also confirm retention.

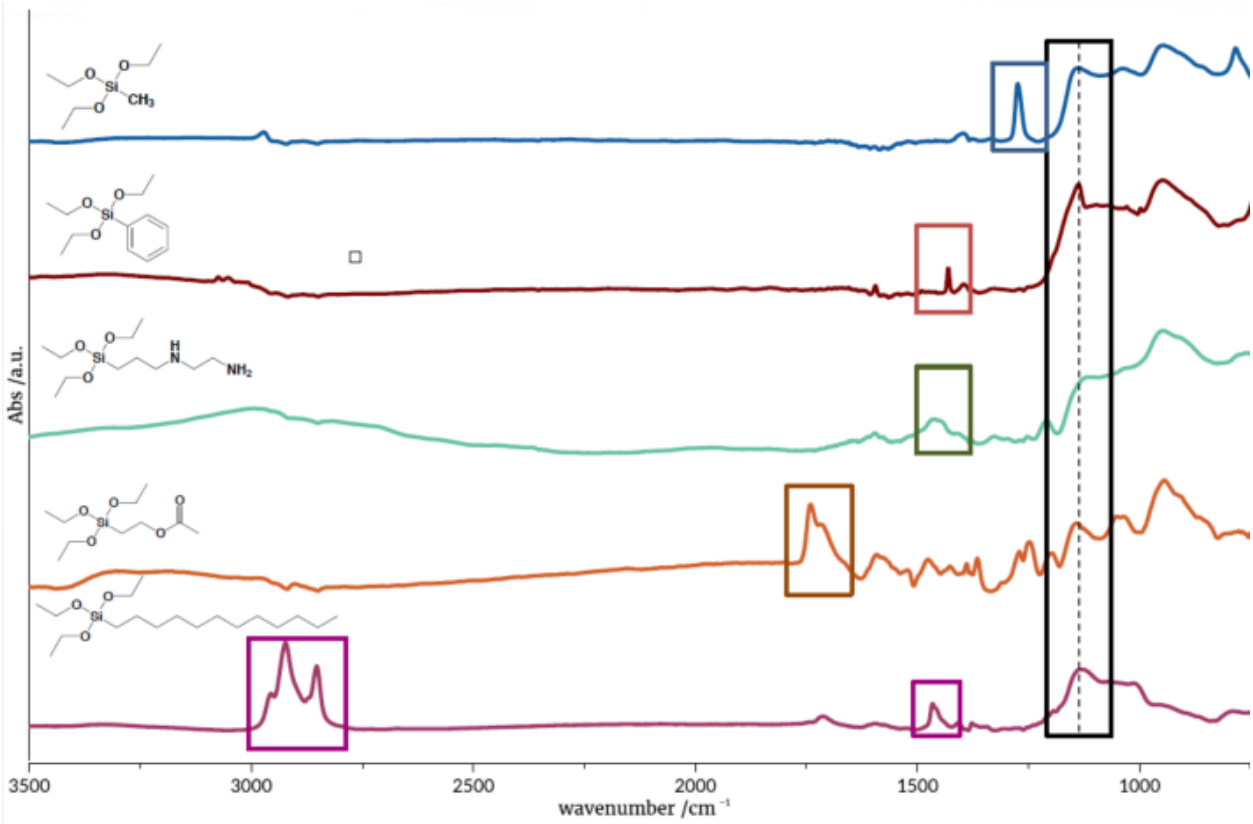


Figure 2-3. Sample spectra for successful deposition of various silanes; The Si-O-Si stretching frequency was shown in each sample verifying siloxane like layer retention.

Based on L. Mawby's work during the academic year, she selected PTES to use on a scaled batch for further flow studies. Her studies showed, that of the silanes we investigated, PTES showed the best properties for scale-up. A full report of this work is currently in preparation for publication and can be found in L. Mawby's doctoral dissertation.

A secondary component of this work was to investigate the addition of a dipodal silane to improve retention and stability of the MTES treated powders. Dipodal silanes have two siloxy ends, increasing the potential for more surface linkages as shown in Figure 2-4. They have also been shown to have an influence on the hydrolytic stability of the surface coating, decreasing the possibility for breakthrough hydrolysis reactions. Due to the increased number of hydrolysable groups, there is also an increased possibility for denser cross-linking, forming a stable interphase. Gelest, one vendor of dipodal silanes, boosts a 100,000x increase in stability when compared to traditional silanes. [9]

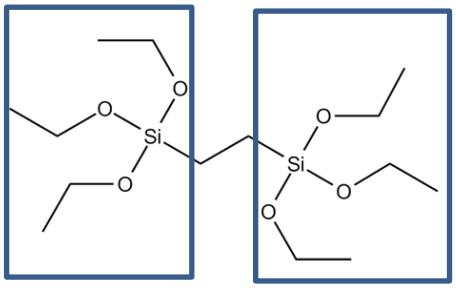


Figure 2-4. Structure of 1,2-bis(triethoxysilyl)ethane with 6 total ethoxy groups for hydrolysis (shown in blue squares)

We investigated two different ratios of regular to dipodal, including 100:1 and 10:1. The powder resulting from the 100:1 mixture showed little to no retention of the dipodal silane as evidenced by DRIFTS. The 10:1 mixture produced a powder with flow properties similar to MTES treated powders, in addition to $-CH_2-$ stretches apparent in the IR spectrum from the retention of BTSE.

2.3 Summary

The surface level work in this section served as the foundation for the bulk-level testing with a full feeder system. By using a wide variety of silanes to down-select from, we covered both hydrophilic and hydrophobic silanes, and found that hydrophobic silanes had the most improved flow properties. This finding was consistent with other work on surface treated aluminum powders

previously reported. Of the hydrophobic silanes tested, PTES had better flow properties when compared to long-chain hydrocarbon silane. This was likely due to the reduced size of the tail; although a phenyl group, the alkyl chain could have retained different tail conformations, potentially hindering further surface reactions with other molecules. The deposition of MTES with BTSE was successful, as ethane components were observed in the DRIFTS data. Of the surface treatments studied herein, PTES and MTES/BTSE were selected for use in full system testing.

3 Flow and Visualization Results from Full Scale Testing

3.1 Introduction

Powder transport using fluidization can be classified as either a lean or dense phase being pneumatically conveyed using a gas phase medium. Though there are many configurations of pneumatic conveying, here we will consider delivery using pressurization through a feed line where the powder moves from high to low pressure. The diameter of the feed line considered in this work is 1.5 mm, which is within the order of magnitude for large agglomerates that can form bridges, blocking the flow. Using the surface treatments discussed, we investigated the influence of surface treatment, not only on flow rate, but also on the ability to produce a steady-state, continuous powder flow. A pictorial of this phenomena is shown in Figure 3-1.

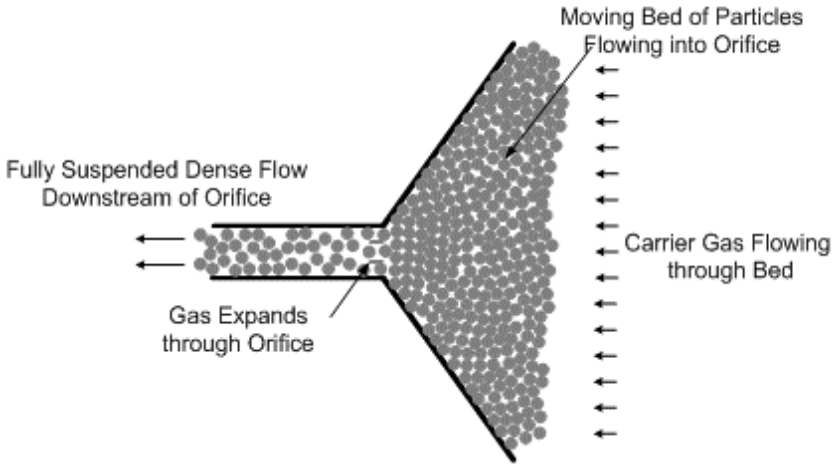


Figure 3-1. Pictorial representation of particles in a gas-fluidized bed moving downstream through an orifice.

3.2 Test Batch Properties

A total of five batches of aluminum powder with different treatment protocols and physical properties were investigated. By picking batches with variable surface treatments and PSD, we hoped to elucidate small differences in the fluidization properties. To provide a control experiment,

we selected the raw, as-received material for testing, in additional to a baseline, MTES treated powder with well understood flow properties. Two of the treated powders were selected based on the results found in section 2.2. Recent protocol development has shifted the treatment process from a fully slurry deposition to using a spray treatment at a larger scale. Although few discernable differences have become apparent from the tested materials, the spray batches were also included for additional insight. We also investigated the influence of PSD, which will be discussed in a subsequent section. The names, treatment, silane, and weight percent (wt.%) silane used are all reported in Table 3-1 for reader reference.

Table 3-1. Sample designations for powders used in full flow testing.

Batch	Treatment	Silane	wt.%
AR (As-received)	N/A	N/A	N/A
Ph-Al	Solution	PTES	1
M-Al	Spray	MTES	1
MB-Al	Solution	MTES+BTSE	1 (MTES), 0.1 (BTSE)
M-Al-01 (low fines)	Spray	MTES	1
M-Al-02 (high fines)	Spray	MTES	1

Prior to powder testing, batches remain under ambient conditions for storage which can last several months depending on the testing schedules. All large-scale flow testing was completed in the final year of this three year effort, although batches were prepared in prior years. Upon inspection, the PTES treated batch (Ph-Al) appeared agglomerated with visible chunks as shown in Figure 3-2a, likely due to long term storage. These chunks broke apart with slight manual force. To remove the potential for less visible agglomerates, the powder was sieved again before flow testing commenced. For comparison, a typical MTES silane treated powder is shown in Figure 3-2b. This finding is important when considering PTES as a silane for future use since any excess powder agglomeration in long term storage may be require additional processing, which is less than ideal.

(a)  (b) 

Figure 3-2. Agglomerated PTES batch (a) compared with MTES treated powder (b) after long term storage.

As mentioned in the introduction, PSD has an influence on a powder's flow and fluidization properties. Although all powders used in this testing were purchased from the same vendor at the same designation, the particle size distribution can vary somewhat between lots. The particle size distributions for all the batches tested are displayed in Figure 3-3. Testing batches AR, Ph-Al and MB-Al were all from the same lot of aluminum powder and thus had a very similar PSD. The baseline MTES batch (M-Al-01) was from a different lot, however, has a similar PSD to AR, Ph-Al, and MB-Al. To investigate the influence of fines, M-Al-02 has a lower average particle size, and thus a higher percentage of fines.

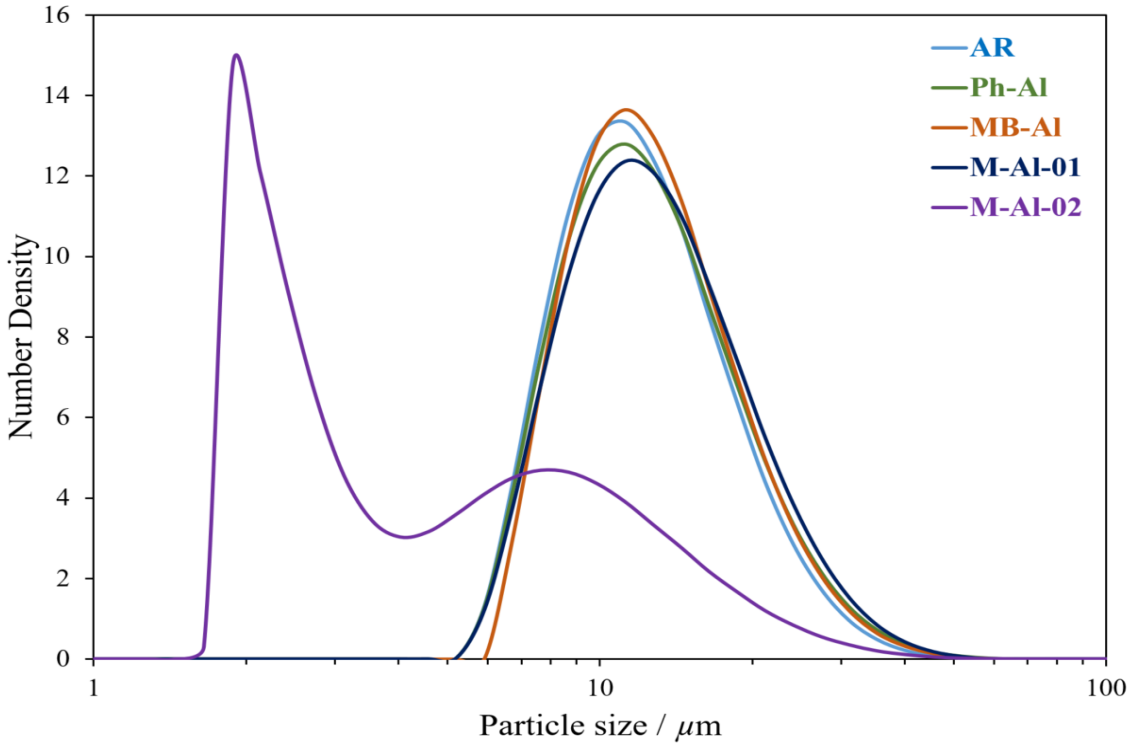


Figure 3-3. PSDs for all batches tested in this work, M-Al-02 shows a large peak in the fines area, suggesting a higher percentage of fines in terms of number density.

3.3 Flow Testing and Fluidization Modeling/Visualization

Using full feeder testing, the batches discussed above were compared through a series of flow tests to investigate fluidization differences. One hypothesis of this work is that if powders have optimal flow properties measured outside of the feeder environment, then they will have high-performance when used in system testing. Each batch of powder was tested using a full feeder system and specific parameters were measured, including density, flow rate, and delivery pressure. To aide in the data interpretation, powder plumes were imaged as well and processed for comparison.

A piston-driven, cylindrical powder feed system was used to dispense the various batches of aluminum powder into a collection device where the expelled powder weight was measured to infer the mass flow rate. The results of these tests are displayed in Figure 3-4. For comparison, the delivery pressure was kept constant within tolerances for most of the powder batches. Minor differences (± 200 kPa) in powder delivery pressure can account for some variation in the mass flow rate. Due to slight testing protocol differences, the results for MB-Al were completed at a

lower delivery pressure. However, these results are also included as the results were still similar to the other batches despite this difference.

Figure 3-4 shows the flow rate plots measured over time for each of the trial batches. Due to the noise associated with this measurement, a rolling average is collected and reported as a single numerical value in Table 3-2. The rolling averages for each powder were around 9 – 11 g/s, indicating little variation in the flow rate as a result of the various surface treatments and PSD. It should be noted that the accuracy of the scale used when handling large quantities of powders precludes deciphering subtle differences down to the milligram level.

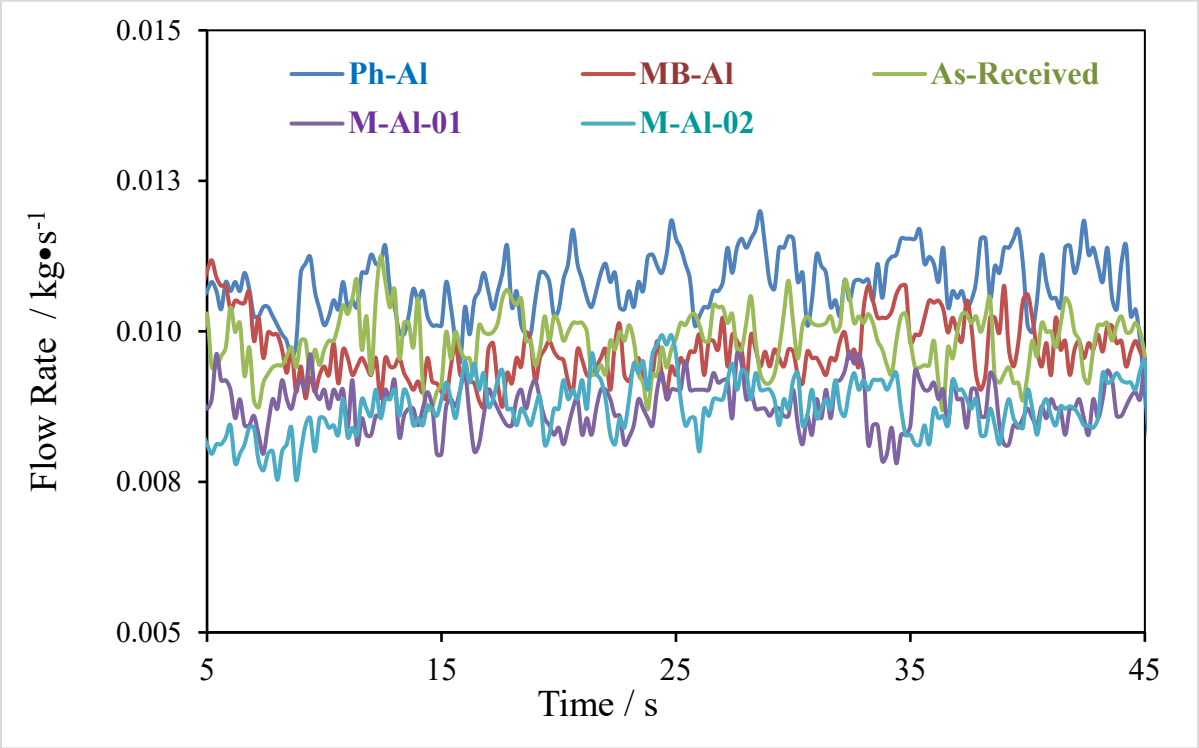


Figure 3-4. Flow rates for each of the test batches over a period of 45 seconds; the results show limited differences between all five batches.

Table 3-3 also includes the delivery pressure and density values collected from each test. Most batches were run using similar conditions at delivery pressures of ~2200 - 2400 kPa. Overall, all five of the powders flowed well and were within normal operating parameters. Additionally, there was no evidence of flow sputtering or clogging anywhere in the powder feed system. No discernable differences were noted with regards to different surface treatment processes or particle size distribution.

Table 3-2. Flow test powder properties, including flow rates and delivery pressures.

Property	AR	MB-Al	Ph-Al	M-Al-01	M-Al-02
Flow Rate (g/s)	9.98	9.98	10.66	9.07	9.07
Delivery Pressure (kPa)	2316.64	1703.01	2364.9	2261.48	2309.74
Tap Density (g/mL)	1.56	1.72	1.67	1.72	1.69
Feeder Density (g/mL)	1.59	1.52	1.63	1.70	1.70

The powder density is an important property that dictates the amount of feasible storage. Here, we report the tap density and the feeder density in Table 3-2. Tap density is measured using a calibrated instrument *ex situ*, whereas the feeder density is directly calculated based on the dimensions of the feeder and amount of material loaded. The packing density is a function of the loading process as well as the powder's properties. By treating the surface of the powder, it allows for the particles to settle inside the feed system, making it pack efficiently and thus increasing the packing density. Typically, a treated batch of powder will have a higher packing density than the as-received material, as observed in this testing effort. The packing density of the feed system is also subject to factors of the loading process and therefore is not the most reliable measurement. Due to the mechanism of the measurement, the tap density should likely be higher than the feeder density, which was observed in most cases. The exception was the raw material; in this instance, the sheer amount of material loaded, 5 kg vs. 100 grams, likely has a higher influence with a more cohesive powder simply due to the weight of material closing air gaps within the powder bed.

Beyond physical measurements, all batches were also investigated through a flow visualization technique. The same piston cylinder powder feed system was used to dispense powder through an injection nozzle into open atmosphere to form a plume. An example image of a sample powder plume is displayed in Figure 3-5.

Figure 3-5. Sample unprocessed powder plume image captured with visualization equipment.

The flow imaging cart is shown in Figure 3-6 and utilizes a LED light source that is used to illuminate the powder plume. A lens is positioned in front of a camera, which is used to capture a series of images of the backlit plume. The images are then processed to produce a plot of number density. This is accomplished by first using Mie Scattering Theory to determine the theoretical light scattering due to a cloud of particles with a given particle size distribution. The images of the powder plume are compared to images of the light source to determine the amount of light passing through the plume. By then comparing this result to Mie Scattering Theory, the number density of particles in the plume is calculated and graphical representations are produced. The analysis also divides the image into two halves, from which the expansion angle of the plume can be determined. The same conditions, including delivery pressure and injector, were used for all flow visualizations.

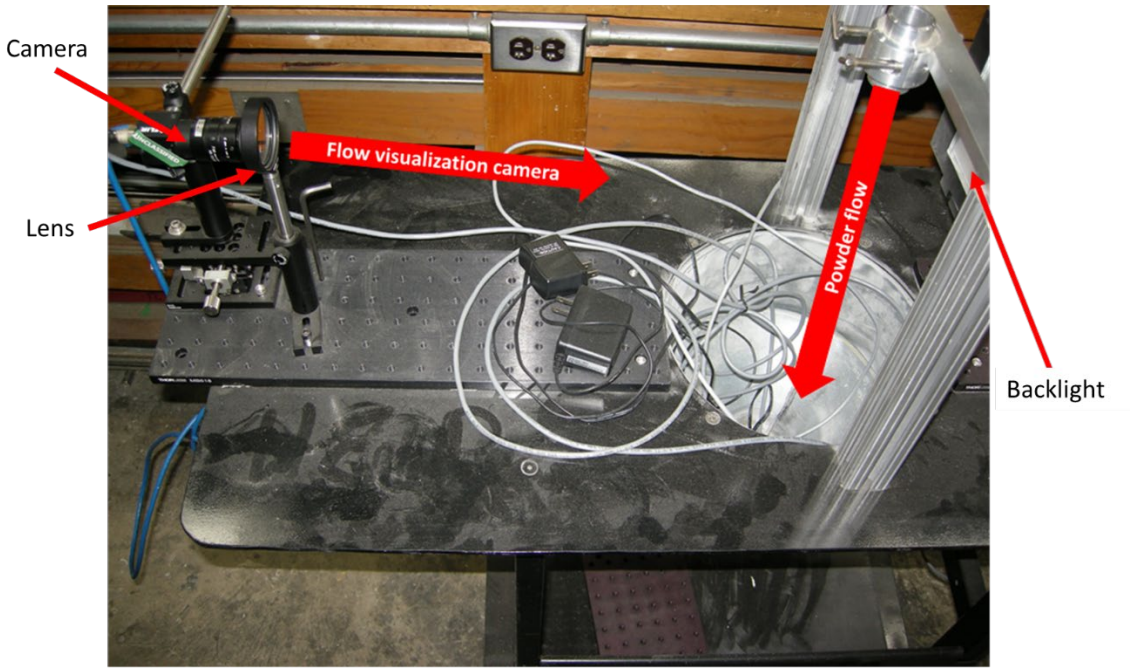


Figure 3-6. Flow visualization cart showing angle of powder flow relative to camera angle and back light.

The four batches with similar PSD (AR, MB-Al, Ph-Al, M-Al-01), all had similar number density visualization results. Four example "right side" images are shown in Figure 3-7. Upon inspection, there are small lateral differences between the AR, Ph-Al, and M-Al-01, where the density shows small fluctuations along the length of the plume. Batch AR and Ph-Al appeared to have slightly higher density regions that stretched further down the plume when compared to M-Al-01. Batch M-Al-02 showed higher density regions throughout the length of the plume, which were approximately twice that of all the other samples towards the top of the plume (~ 7 x 10^7 vs 3 x 10^7 particles/cm^3). All processed plume images and angle expansion graphs can be found in Appendix A.

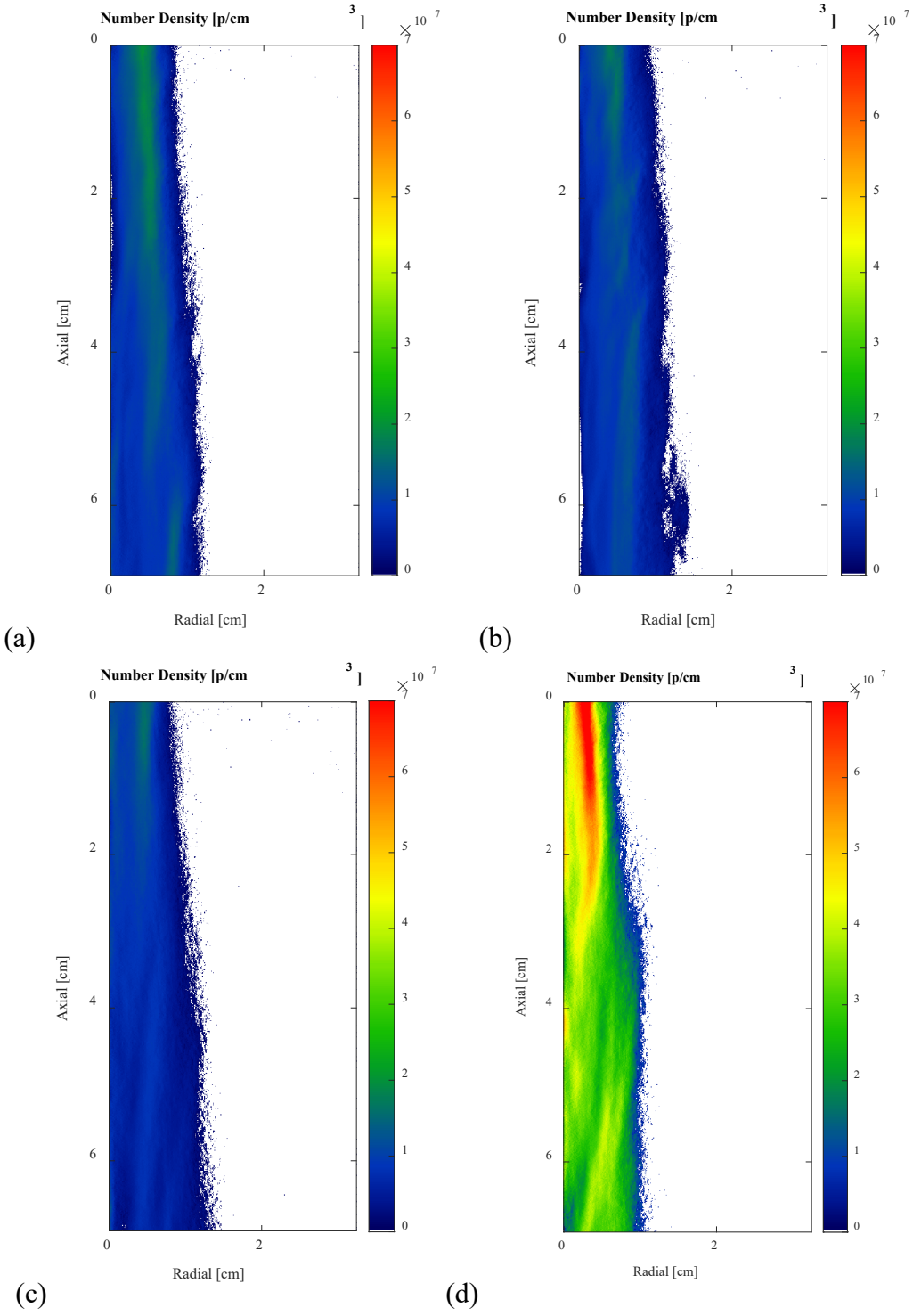


Figure 3-7. Right side particle density images of samples (a) AR, (b) Ph-Al, (c) M-Al-01, and (d) M-Al-02; M-Al-02 showed the highest number density due to the higher percentage of fine particles in the powder.

The angle of expansion, summarized in Table 3-3, is mainly a function of the injector and operating pressure, but is influenced by the powder flow and its properties. A half angle, with the style of injector used, can range anywhere from 2° to 12°, and is nominally around 3°-5°. Anecdotally, batch Ph-Al had a noticeably smaller half angle, which suggests that this powder does not expand as well as the others during flow testing. It is possible, due to the observed agglomeration after storage, the material may be more agglomerated at the micron level as well, reducing the "dustiness" of the material. The PSD distribution measurements collected herein were prepared in solution, which would not account for micron sized agglomerates present in the dry state. It may be possible to further understand this result by using direct imaging of the particles for a different perspective on the PSD in the dry state.

Table 3-3. Calculated half angles for various test powder reported by plume side.

Plume Side	MB-Al	As Received	Ph-Al	M-Al-01	M-Al-02
Right Side	3.5°	3.7°	2.5°	4.1°	4.3°
Left Side	5.2°	4.5°	2.9°	3.7°	6.5°

3.4 Summary of Flow Testing Results

Five different batches of aluminum powder were successfully flow tested, comparing various attributes, including surface treatment and PSD. These findings showed the robustness of the feeder system used in this work, showing few discernable differences based on treatment and PSD. Untreated, as-received powder was successfully run through the feed system and showed reasonable flow rates and density values. The various surface treatments were also difficult to differentiate based on the results of the large scale flow testing. The most apparent difference came from the powder with the high percentage of fines; the particle density was found to be higher in the plume than in the other powders tested. Although not surprising, this finding warrants further investigation to fundamentally understand powder dispersion within the plume as influenced by PSD. It should be noted that the state of batch Ph-Al was concerning. This powder had clumped during storage, which will limit its shelf life before having to sieve the powder again to break up any agglomerates.

4 Conclusions and Future Work

The work presented herein shows how as-received aluminum powder can be surface treated and processed to enable liquid like flow and delivery through a fluidized powder feeder. Based on our existing familiarity with silane surface treatments of aluminum powder, we expanded the knowledge base by including new silanes to further understand the fundamental inter-particulate interactions that cause cohesivity. To this end, we studied different silane coupling agents with hydrophobic and hydrophilic functionality. By utilizing surface and bulk scale testing, we were able to down select the best silane to a hydrophobic silane, PTES. This silane was used for large scale processing and testing to study the flow behavior in a fluidized powder feeder.

Large scale feeder testing was conducted using PTES treated powder, in addition to several other variants to investigate potential differences in the flow imaging. Interestingly, significant differences were not observed, even when using powder that had not been surface treated. This speaks to the robustness of the system to produce flow even with a wide variety of powder properties. The batch variants included powders with different PSD; this manifested itself in the particle density images by showing a high concentration of particles in the plume, which is to be expected. The limitations of this testing should be noted. The series of experiments consisted of relatively short test durations, on the order of a minute. It is possible that with longer tests, discrepancies between the powders may arise, especially with comparison of the treated and as-received powders. Over time, the untreated powder may cause gas channeling in the powder bed or clogging in the feed system, which could lead to decreasing mass flow rate or other flowability issues. This testing was also performed with only one specific size of aluminum powder, H15, at one target mass flow rate, and with consistent feeder parameters. It is unknown what effect changing these aspects would have, but it is possible that they could cause differences between the various batches of powder. Future testing efforts could focus on these aspects and their effects on large scale flow testing.

Future work in this area could include a more refined study of the powder properties ex situ, including dry PSD measurements, such as aspirated measurements or by particle imaging. This could give some indication of how the PSD is influenced by the dry state in which is it delivered, rather than using a liquid phase measurement. Additional insight could be drawn from powder

rheology measurements, which could refine the differences between the powders. Regardless, the system was able to deliver all powder samples irrespective of its treatment and PSD.

As is a significant component of NURP, L. Mawby successfully defended and is currently working on publications from her work in the down-selection process and other surface studies which are reported in her dissertation.

APPENDIX A: FLOW VISUALIZATIONS

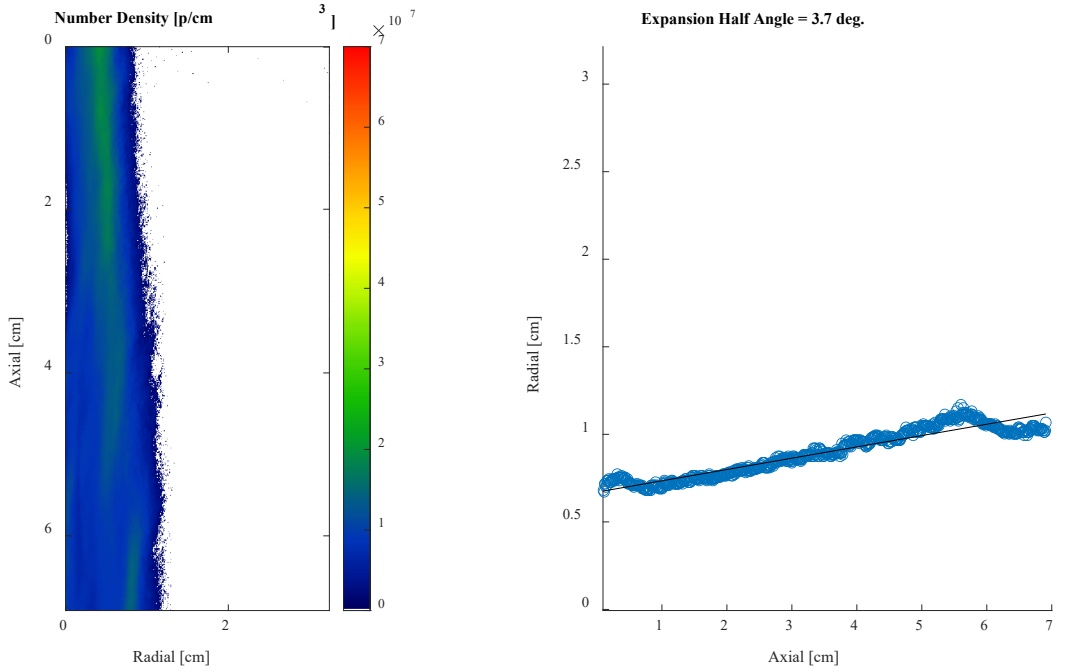


Figure A-1: AR-As Received, Right Side of Plume Image

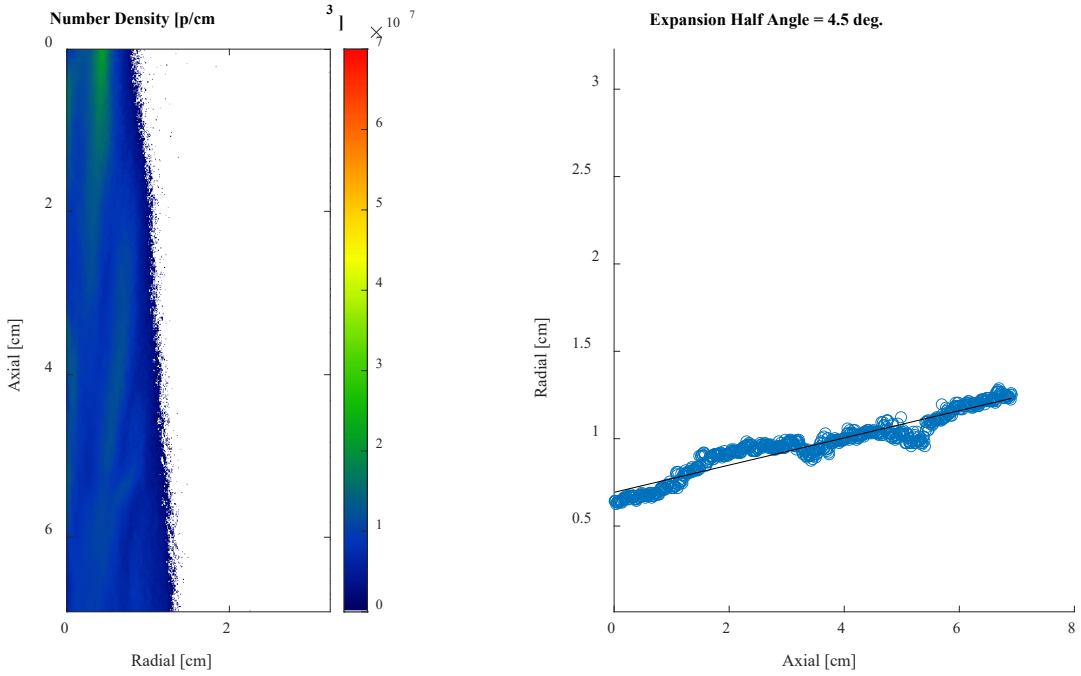


Figure A-2: AR-As Received, Left Side of Plume Image

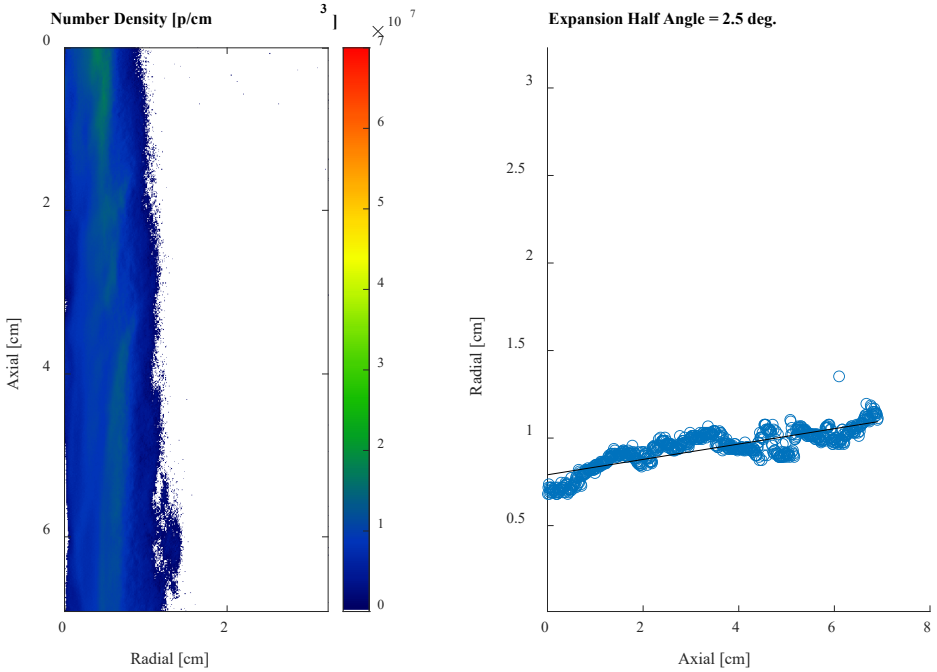


Figure A-3: Ph-Al, Right Side of Plume Image

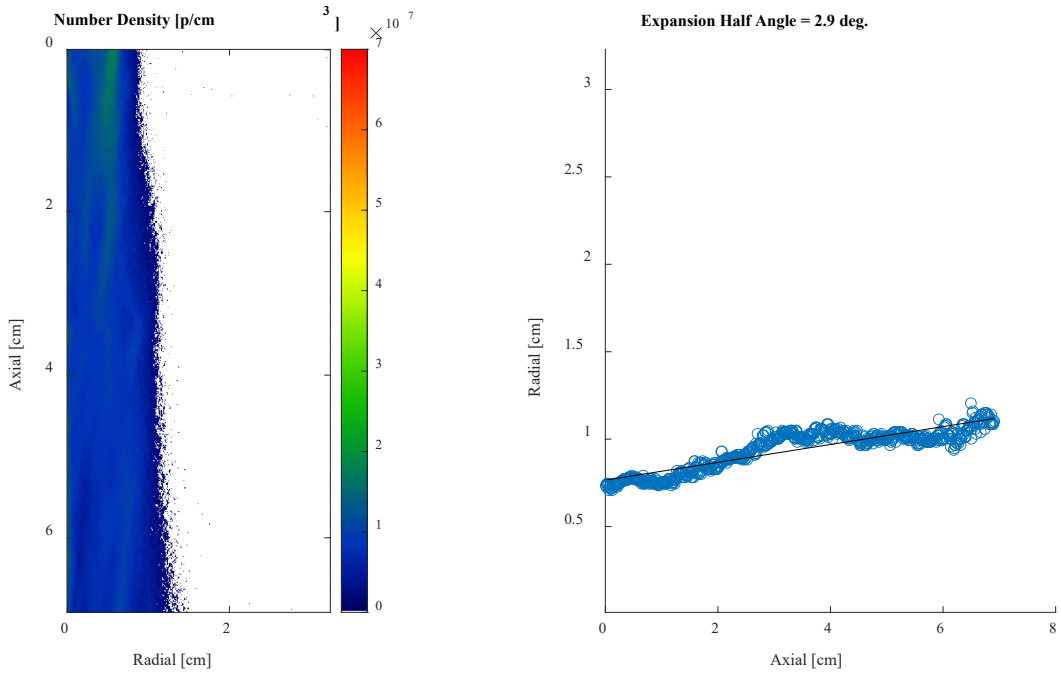


Figure A-4: Ph-Al, Left Side of Plume Image

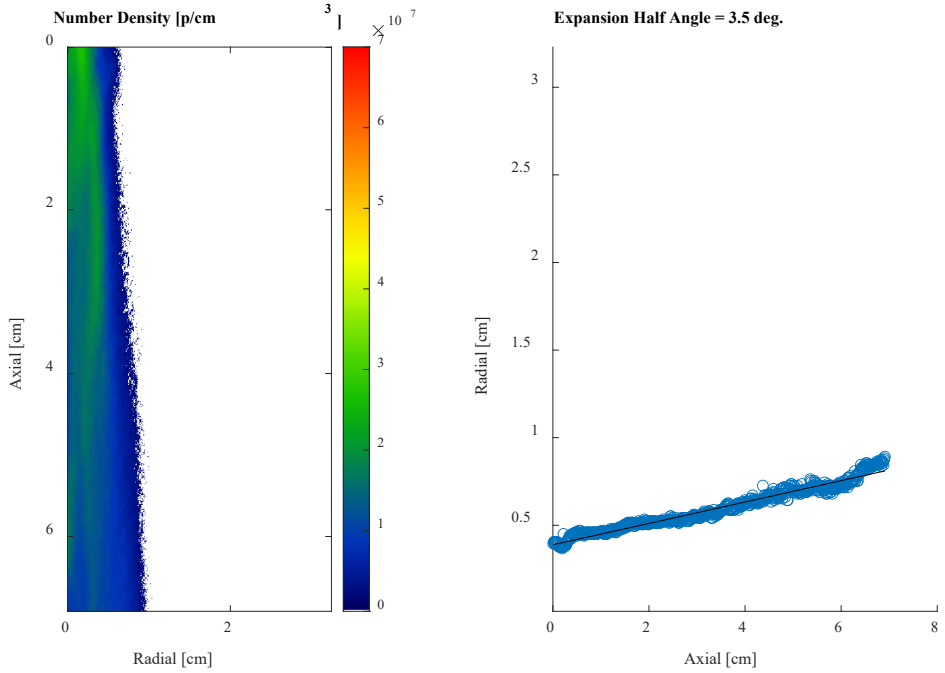


Figure A-5: Mb-Al, Right Side of Plume Image

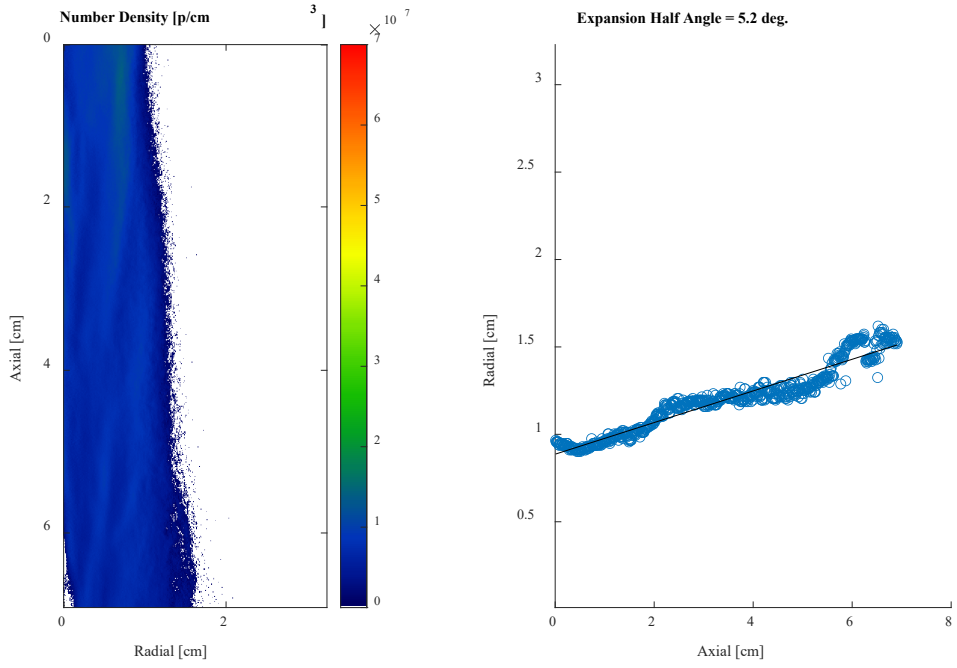


Figure A-6: Mb-Al, Left Side of Plume Image

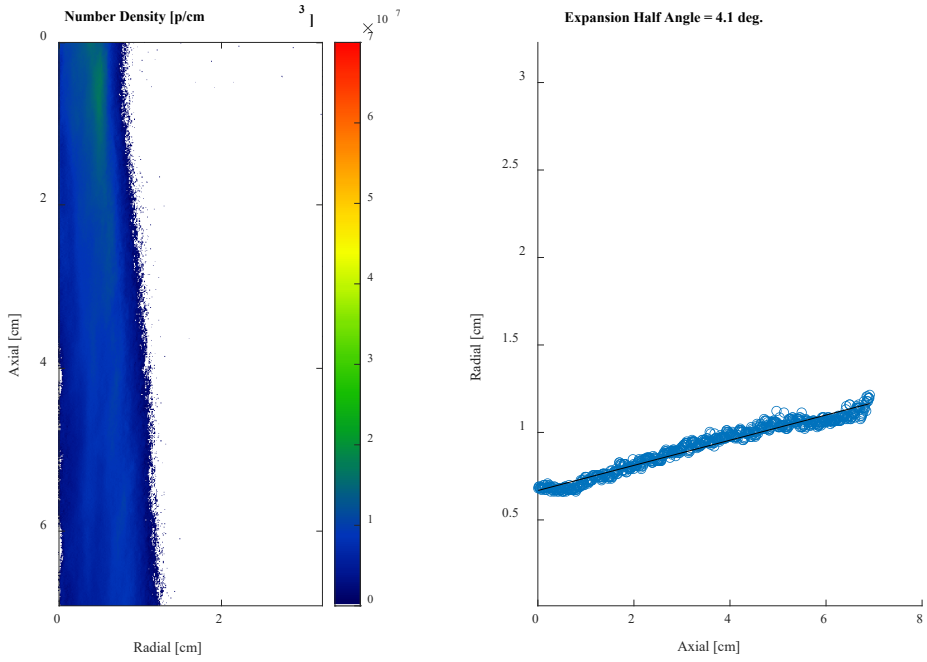


Figure A-7: M-Al-01, Right Side of Plume Image

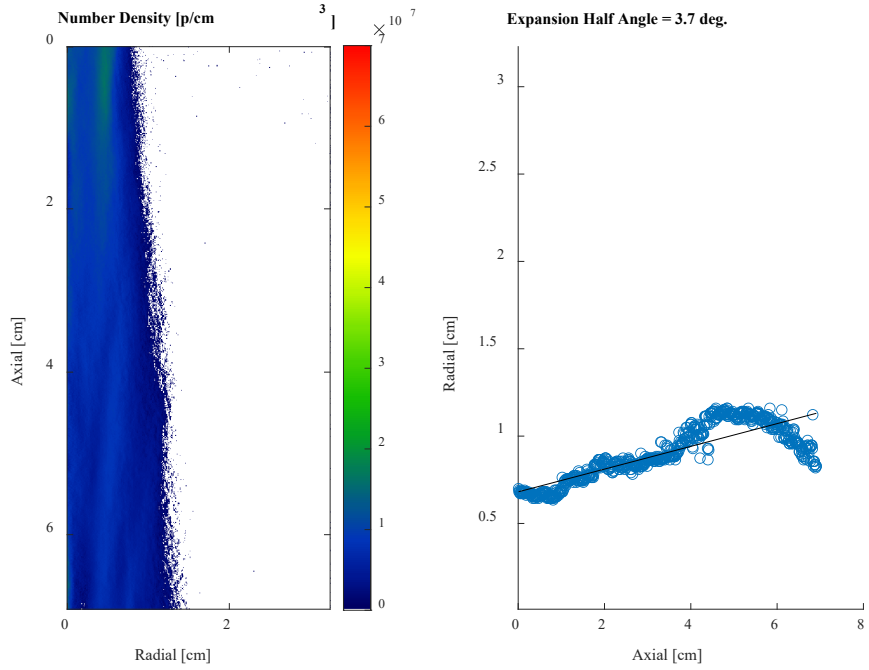


Figure A-8: M-Al-01, Left Side of Plume Image

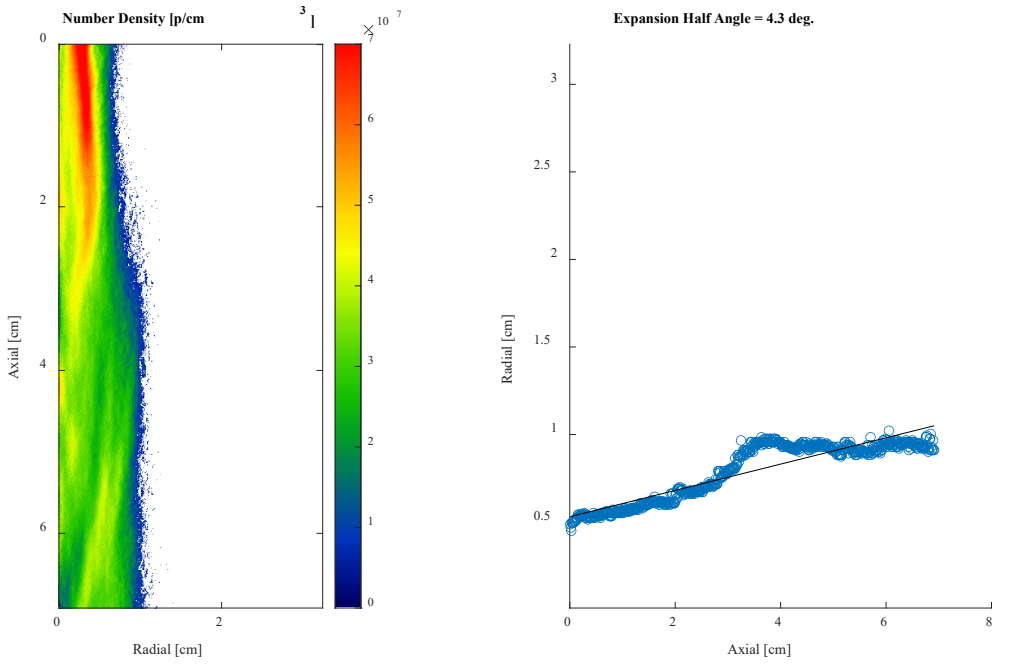


Figure A-9: M-Al-02, Right Side of Plume Image

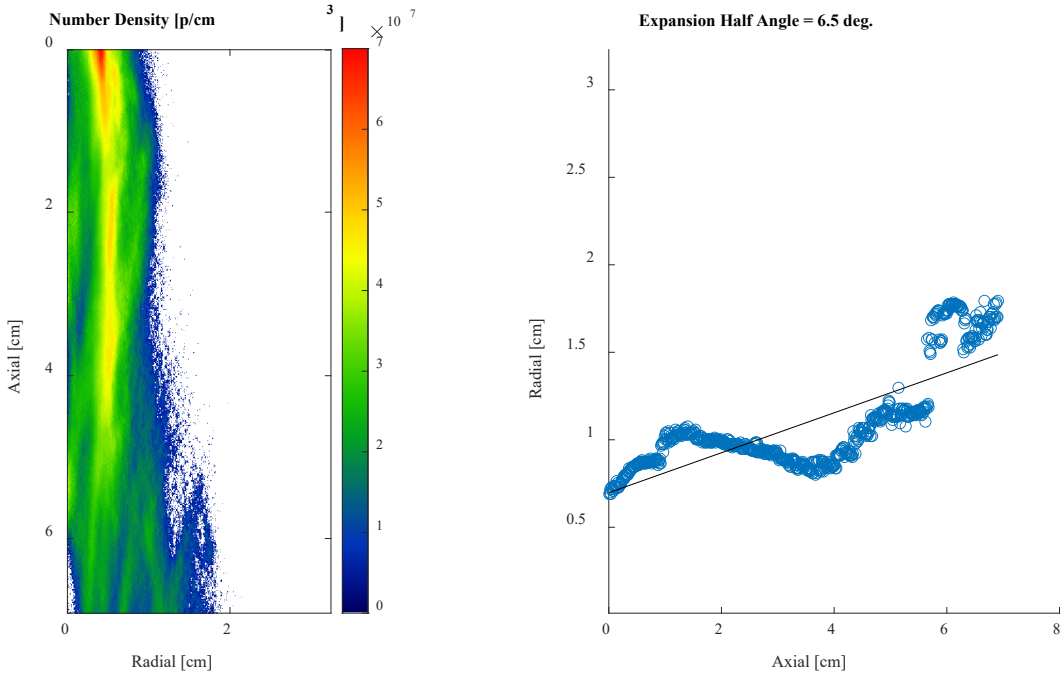


Figure A-10: M-Al-02, Left Side of Plume Image

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