PNNL-29061

Final Report on the Viability of Acoustic Techniques for Density and Mass Flow in Enrichment Plants

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Executive Summary

The primary purpose of this research is to study the viability of acoustic signatures and sensors that could support accurate, noninvasive, and unattended measurement of uranium hexafluoride (UF_6) gas density and mass flow rate in situations representative of gaseous centrifuge enrichment plants (GCEP) under safeguards by the International Atomic Energy Agency. The measurement method under development can be readily retrofitted to the exterior of piping in existing systems, potentially facilitating acceptance by facility operators. The findings from the demonstration of a preliminary prototype design and first-generation analysis algorithms in this project will inform the safeguards community as to whether continued acoustic instrument and methods development are warranted.

This project conducted two types of measurements, depending on the flow conditions of the gas. For static measurements, the focus is on understanding the acoustic properties at low pressure for non-flowing UF_6. For flow measurements, the focus is on understanding the performance of the acoustic system in a simulation of a GCEP pipe using a surrogate gas under fully developed flow conditions. Testing with flowing UF_6 was not considered because the cost of building a flowing UF_6 system with flow similar to a GCEP was considerably greater than allocated project resources.

This document summarizes the results of research conducted over the course of the project. Based on the results to date, the approach is seen to be promising with non-invasive acoustic measurements shown to be possible at the desired pressures. The project has demonstrated

- 5% uncertainty of the UF_6 gas density for static measurements at pressures relevant to GCEP operations over a 5-minute measurement window with a clear path forward to achieving considerably lower uncertainties over a two-hour period,
- Measurements of flowing surrogate gases at pressures relevant to GCEP operations.

However, additional research remains to be done to further develop the measurement approach and transition the technology for field use. These development needs include:

- Understand the impact of flowing gas on gas density measurements
- Adding sensors and improving the measurement procedure
- Augmenting the data analysis methods by leveraging machine learning approaches
- Reevaluating uncertainty requirements for density and mass flow given observed Online Enrichment Monitor (OLEM) measurement uncertainties
- Designing a robust instrumentation package for field-testing
- Evaluating measurement results to understand the potential impact on mass balance calculations within GCEP facilities.

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1.0 Introduction

The primary purpose of this research is to study the viability of acoustic signatures and sensors that could support accurate, noninvasive, and unattended measurement of uranium hexafluoride (UF_6) gas density and mass flow rate in situations representative of gaseous centrifuge enrichment plants (GCEP) under safeguards by the International Atomic Energy Agency (IAEA). The measurement method under development can be readily retrofitted to the exterior of piping in existing systems, potentially facilitating acceptance by facility operators. The findings from the demonstration of a preliminary prototype design and first-generation analysis algorithms in this project will inform the safeguards community as to whether continued acoustic instrument and methods development are warranted.

The scope of this project is to perform laboratory measurements to determine if acoustic measurements are viable for non-invasively determining gas density and flow rate in GCEP unit header pipes under conditions that are encountered at an enrichment plant. This document provides a high-level summary of the project achievements as well as the remaining challenges.

This project conducted two types of measurements, depending on the flow conditions of the gas. For static measurements, the focus is on understanding the acoustic properties at low pressure for non-flowing UF_6. For flow measurements, the focus is on understanding the performance of the acoustic system in a simulation of a GCEP pipe using a surrogate gas under fully developed flow conditions. Testing with flowing UF_6 was not considered because the cost of building a flowing UF_6 system with flow similar to a GCEP was considerably greater than the allocated project resources.

The original uncertainty goal stated in the project proposal was 1% to match the target uncertainty of Online Enrichment Monitor (OLEM). During the project, that number was refined to 1% over the reporting period of the OLEM, which can be as short as two hours. In practice, the OLEM achieves uncertainties of a few percent over four or more hours[1]. Since this system is envisioned to support the OLEM, it is reasonable to increase the maximum uncertainty goal to match the operational OLEM performance.

This report contains three primary sections. The first section describes the achievements accomplished by the project. The next section describes the technical challenges with the implemented approach for the envisioned application, together with suggested ways to address those challenges. And finally, the main body of the report concludes with a brief summary. In addition to these primary sections, there are two appendices on the static measurements and signal analysis.

2.0 Achievements

At the end of FY2018, research activities in the project have resulted in the following major accomplishments:

- Non-invasive acoustic measurements in UF_6 under static (non-flowing) conditions (Appendix A) showed the existence of distinguishable acoustic energy at lower pressures (down to about 10 Torr). The project team has demonstrated that there is an observable gas-coupled acoustic signal in UF_6 that is sensitive to gas density at GCEP-relevant pressures, even allowing for the various sources of noise in the measurement system. There measurements were performed at 55°C to prevent sublimation of the UF_6 on the instrument walls. This temperature is elevated by about 20°C compared to operations

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[1] J. Ely, private communication, Sept. 2018.

at GCEP; however, the acoustics measurement technique performance will not be dramatically impacted by absolute temperature. This is a considerable technical achievement. While there is clearly sensitivity in the measurements to the UF_6 gas density, those sensitivities are subtler than observed for low pressure air. To take advantage of these sensitivities, the project team is exploring machine learning algorithms to quantify gas density from the measurements. An overview of the analysis methodology to date is discussed in Appendix B. The project has achieved a 5% uncertainty in the estimates in density at 10 Torr and 50 Torr when considering multiple measurements conducted in approximately 5 minutes. The project team believes that given an hour to measure the density, uncertainties of approximately 1% are achievable. The project was not able to confirm this experimentally due to limited resources.

- Non-invasive acoustic measurements of flowing surrogate gases at low pressures was demonstrated. The project team designed, fabricated, and completed measurements using a flowing surrogate gas (dry air) at pressures that were similar to the GCEP safeguards scenario. The team was able to demonstrate repeatable measurements at these pressures and flow rates, even allowing for the various sources of acoustic noise in the flow system.

These accomplishments required overcoming a series of technical challenges over the course of the project. These challenges are briefly described below, along with the approaches that were investigated and eventually implemented for overcoming them.

3.0 Technical Challenges and Solutions

The project has identified six critical risks in the successful development of this technology:

- Inadequate signal-to-noise ratio (SNR) of the acoustic signal in low pressure UF_6,
- Unknown boundary conditions for acoustic coupling, e.g. deposits on the inner pipe wall,
- Inadequate acoustic coupling between the wedge and the pipe leading to lower SNR,
- Sensitivity of the observed acoustic signals to gas temperature,
- Acoustical noise in the GCEP environment interfering with the measurement,
- Data Analysis and Prediction Capabilities.

Each of these risks are discussed below.

Low SNR at Low Gas Pressures

The project was designed from its inception to mitigate the risks for low SNR using a staged approach with a critical decision point to terminate the effort if appropriate. The project initially confirmed that there was an observable signal in low pressure air (significantly below current commercially available capabilities) without the presence of structural-borne noise. The second step was the successful demonstration of an observable signal in the presence of structure-borne noise. A review of the progress was conducted at this point by DNN R&D to evaluate if the project should proceed. The third step was an attempt to observe signals in the same setup using SF_6. It was determined that SF_6 is not a representative surrogate due to high acoustical attenuation in SF_6 compared to air and UF_6, so that the SNR was unacceptably low. The fourth step was the measurement of the acoustic signals in UF_6 at representative gas pressures of a GCEP, 10-50 Torr.

Acoustic attenuation dictates the amount of energy available for measurement after the wave-gas interaction and contributes to further lowering the SNR. While acoustic attenuation in UF_6 increases as the pressure decreases, acoustic attenuation in UF_6 does not appear to be a significant contributor here, based on values documented in the literature (Cravens et al. 1979; Bass and Rogers 1984; Bass et al. 1983) as well as measurements from this project. Further, experimental data for UF_6 (both in measurements from this project and other published work (Cravens et al. 1979)) also do not appear to show absorption resonant behavior, as in SF_6, that may limit the applicability of acoustic measurements in the frequency and pressure ranges of interest.

Unknown boundary conditions

The project team acknowledges that wall deposits may hinder acoustic measurements. The project has focused on determining if there is an adequate SNR to have a reliably accurate measure of gas properties without the presence of wall deposits and deferred the issue of wall deposits to a later time. It is conceivable to conduct an initial investigation on the impact of wall deposits with the current static UF_6 test setup.

Inadequate Coupling

The coupling of acoustic energy from the wedge to the pipe can be problematic at elevated temperatures (temperatures in excess of about 30° C). In most applications, a layer of gel acoustic couplant is used to improve the coupling of energy from the wedge to the pipe. However, gel couplants tend to degrade at higher temperatures and over time can dry out, decreasing coupling efficiency. We have examined a number of options in the laboratory for addressing this acoustic coupling problem at both high temperature and room temperature:

- High temperature epoxy (HARDMAN® Machinable Epoxy DOUBLE/BUBBLE® Yellow Package #04002) was examined for use at the higher temperature experiments in the laboratory but was ultimately eliminated from consideration given the difficulty in maintaining coupling between metal and plastic at high temperatures. The difficulty arises from the differential thermal expansion between the metal pipe and the plastic wedge material and can result in the wedge becoming uncoupled from the pipe.
- High temperature silicone-based couplant (HiTempco by Echo Ultrasonics) proved to be significantly more efficient. Laboratory tests have indicated that this couplant does not dry out as easily as other materials and is able to maintain efficient acoustic coupling between the wedge and the pipe at high temperatures over the course of several weeks.
- Epoxy (HARDMAN® Extra-Fast Setting Epoxy, DOUBLE/BUBBLE® Red Package #04009) was found to be effective at room temperature, maintaining acoustic coupling effectiveness over the course of the project (3 years).

Dry couplant membranes are typically made from thin layers of elastomers and are a proven industry solution for maintaining the effectiveness of acoustic coupling over long periods. While we did not evaluate this option in the laboratory on this project, data from past projects indicate long-term stability of dry acoustic coupling as long as the test equipment is kept in a reasonably climate-controlled area and the temperature does not exceed about 30° C. Alternative dry acoustic couplant materials are also available through manufacturers such as Innovation Polymers[2] that appear to be stable at temperatures up to 100° C. These may be a viable option as the acoustic density and gas flow measurement technology is further matured and evaluated in representative environments.

At GCEP conditions, the nominal UF_6 temperature range is much lower than those examined in this project. The expected temperature range in GCEPs is between ~22° C and ~30° C. Given the expected

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[2] http://www.innovationpolymers.ca/, last accessed September 2018.

environmental conditions, and the findings from the project, we expect that standard epoxy or commercially available dry couplant material will work well in a fielded system.

Sensitivity to Temperature

Temperature variations in the GCEP environment are a possible source of uncertainty in the measurements, given the changes in sound speed and density with pressure and temperature. Pressure, density and temperature are related through the ideal gas law, and sound speed increases with the temperature of the gas. If the temperature is varied during the course of the measurement, the changes in density and sound speed will result in measurement uncertainties through:

- Acoustic impedance (product of density and sound speed) changes that influence the SNR through changes in amount of acoustic energy coupled in and out of the gas,
- Changes in the arrival time of the gas-coupled signal.

Data taken over the course of this project have indicated that the changes in signal attenuation and arrival time are small (less than 0.5%) as long as the temperature variations are small (5° C or less, typical of a GCEP). To further reduce uncertainty, we developed compensation algorithms that monitor the temperature on the outer wall of the pipe (as would be done in the field, as penetrations in the pipe to monitor the gas directly will be not permitted), use UF_6 thermophysical data, and compute correction factors for the amplitude and arrival time of acoustic energy. Analysis indicate that the correction factors are able to account for changes in temperature, *further reducing uncertainty in arrival time to less than 0.1%.* Calculations based on UF_6 thermophysical data also suggest that the impedance mismatch variation with temperature will be limited (~1%) if the temperature variation is small (~1%), as the sound speed and density variations are small over these temperature ranges. As a result, we believe that the temperature issue will not be a significant factor in any fielded system. If necessary, the average of the arrival times with and against flow, which is sensitive to the speed of sound in the gas, may provide an additional correction related to the temperature.

Acoustical Interference

A common concern about this approach is the potential interference of acoustical noise in the GCEP environment with the observed gas-coupled signal. Principle sources of such noise are likely to be vacuum pumps and centrifuges. The project team acknowledges that these issues exist but believes that they will have minimal impact on the acoustic measurements for several reasons. First, the frequency of the noise, below ~100 kHz, is well below the frequency of the acoustic signal used for measurements in this project. Second, the sources of noise are likely to be a long way away from the acoustic measurement system, attenuating the acoustic noise. Third, it is possible to clamp the pipe upstream and downstream of the acoustic system to attenuate this acoustic noise. Fourth, the acoustic system itself relies on significant damping of the acoustic signals in the pipe wall, which will also attenuate the majority of external sources of the acoustical noise. As a proof-of-concept, tests on the low-pressure air flow system were conducted with and without the vacuum pump running and the resulting observation was that the SNR was not influenced by equipment noise.

Data Analysis and Prediction Capabilities

The analysis of signals below pressures of 100 Torr of UF_6 indicate a weak dependency between integral quantities such as signal energy (L_2-norm) and pressure. This situation is expected due to reduced acoustic coupling between the signal and the fluid at these low pressures. Currently, we are investigating a number of signal processing techniques and machine learning algorithms to overcome this challenge. The development of a convolutional neural network (CNN) model for pressure and mass flow rate prediction is recommended; despite the proven performance of CNN algorithms as predictive models, we recognize that risks exist in terms of accuracy and discrimination of low-pressure levels.

4.0 Summary

This document summarizes the results of research conducted over the course of the project to assess the viability of using acoustic measurements in low-pressure environments for measuring density and flow rate to calculate UF_6 mass flow rates in GCEP. Based on the results to date, the approach is seen to be promising with non-invasive acoustic measurements shown to be possible at the desired pressures. However, additional research remains to be done to further develop the measurement approach and transition the technology for field use. These development needs include:

- Understand the impact of flowing gas on gas density measurements
- Adding sensors and improving the measurement procedure
- Augmenting the data analysis methods by leveraging machine learning methods
- Reevaluating uncertainty requirements for density and mass flow given observed OLEM measurement uncertainties
- Designing a robust instrumentation package for field-testing
- Evaluating measurement results to understand the potential impact on mass balance calculations within GCEP facilities.

5.0 References

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Appendix A

Acoustic Measurements in Static UF_6

A custom measurement setup, shown in Figure A-1, was constructed for demonstrating density quantification of UF_6 at desired pressures using acoustic techniques. UF_6 contained in a Hoke tube is rapidly sublimed into the test cell by heating the supply tube. UF_6 gas in the test cell may also be condensed back into the Hoke tube; rapid condensation is achieved by cooling the tube with dry ice. This system is capable of achieving internal pressures ranging from a few milli-Torr to about 400 Torr, with the UF_6 maintained in gas phase at higher pressures by means of heating jackets. Acoustic transducers (hidden behind the heating jackets in figure) are mounted on the outside of the test cell. Pressure transmitters are also located at either end of the test cell to monitor the internal pressure. The test system is evacuated by means of a high efficiency vacuum pump connected to filters that capture most residual contamination.

Figure A-1. Custom test setup for acoustic measurements of UF_6 density

Initial analysis of the data taken at a number of pressures indicates a trend between actual and estimated density. The actual density was determined from the measured temperature and pressure. A preliminary support vector analysis was performed for predicting the UF_6 density from the acoustics data, as shown in Figure A-2. The results suggest an uncertainty of ~25% at 10 Torr and ~5% at 50 Torr *for a single signal*. At 10 Torr and 50 Torr, the density of UF_6 is approximately 0.17 and 0.85 kg/m^3. The data points indicate the mean and standard deviation of the estimated density from a collection of acoustic signals at a specific density. These results motivate a more detailed analysis that is currently underway.

The reported uncertainty values are based on a set of data (30 separate signals) obtained at a single pressure. The uncertainty is the standard deviation of the distribution of the extracted densities from those 30 signals. Thus, the uncertainties are interpreted as the uncertainty from a single signal. Each signal takes approximately 5 seconds to collect, including buffer time between measurements. Given more time, it is possible to conduct more measurements and combine the results. As the results from multiple signals are averaged and random noise removed, we observed that the uncertainty is reduced. For instance, if we average of the 30 signals, we achieve uncertainties of 5% at 10 Torr. We expect that obtaining data over the course of two hours, under static conditions, will reduce uncertainty to below 1% for all GCEP-relevant pressures.

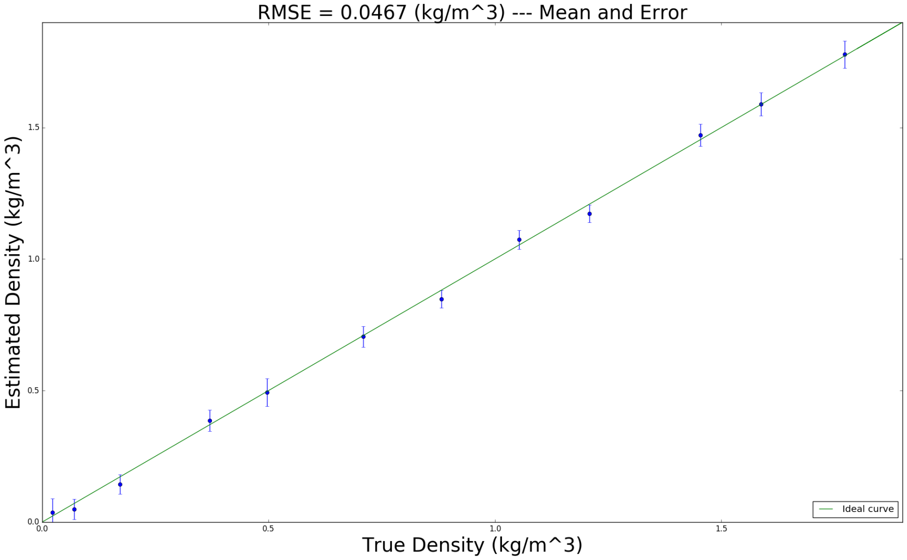

Figure A-2. Preliminary results from support vector analysis of UF_6 data predicting the gas density. The error bars denote the one standard deviation in the collection of data for each density for a single measurement.

Appendix B

Signal Analysis

B.1 Background and Objectives

Initial analysis of the acoustic signals from the UF_6 measurements suggested that the relationship between the acoustic signal and the gas pressure was subtler than for acoustic signals in low pressure air. For low pressure air, a relatively simple analysis of the relevant acoustic signal using the L_2-norm clearly demonstrated a nearly linear relationship between signal size and gas pressure. For UF_6 however, a similar analysis revealed that the acoustic signal strength was roughly constant for pressures below 100 Torr. As a result, it was concluded that an analysis incorporating the time information, not just integral quantities such as the signal size, should be explored.

This appendix presents progress on developing a signal processing methodology and predictive capabilities to accurately measure pressure or density levels as well as mass flow rates. We have studied different techniques to identify the presence of data structures or features in the signals that carry the information relative to the quantities being sought for pressures below 150 Torr.

We explored a number of signal processing techniques to evaluate the presence of distinguishing features in the received signals. In particular, we focused on the development of spectrograms, a 2-D representation of the signal in the time-frequency domain based on the Short-time Fourier Transform (STFT) (Oppenheim et al. 1999). The procedure for computing STFTs is to divide a longer time signal into shorter segments of equal length and then compute the Fourier transform separately on each shorter segment.

We show that spectrograms are capable of extracting a variety of data structures that represent good candidates for the determination of pressure levels using machine learning algorithms. We recommend the use of a convolutional neural network (CNN) framework to analyze the spectrograms. In recent years, CNNs have been proven very effective in image recognition and classification, and similar techniques have been used in speech recognition algorithms (Abdel-Hamid et al. 2014).

B.2 Signal Processing Algorithms

In this study, we used experimental data (acoustic transmission responses through UF_6 using the static measurement setup described in Appendix A); these data sets represent a set of transmitted and received sinusoidal pulses with a central frequency obtained at different pressure levels. The sampling frequency was set to 25 MHz in the experimental setup, a value that is more than adequate to resolve the spectral content in the signal up to a Nyquist frequency of 12.5 MHz.

For each pressure level, the data sets contain a number of experiments corresponding to several realizations of the same input signal being pulsed into the system. The input signal considered in this study is a sinusoidal function.

In order to reduce the number of experimental data points at each pressure level, we have investigated the use of averaged waveforms over the number of samples at each pressure level. Analysis of the standard deviations (2σ) of the averaged data set were reviewed to determine the following aspects:

- The averaged datasets are a suitable representation of the datasets
- Consistency between datasets
- The magnitude of the statistical uncertainty at each pressure level is acceptable as compared to the magnitude of the signal

Figure B-1 shows the averaged pressure signals with their associated standard deviation (2σ). It shows that the magnitude of the standard deviation for the averaged waveforms is negligible as compared to the magnitude of the signal; therefore, it is possible to use the averaged waveforms to represent each pressure level.

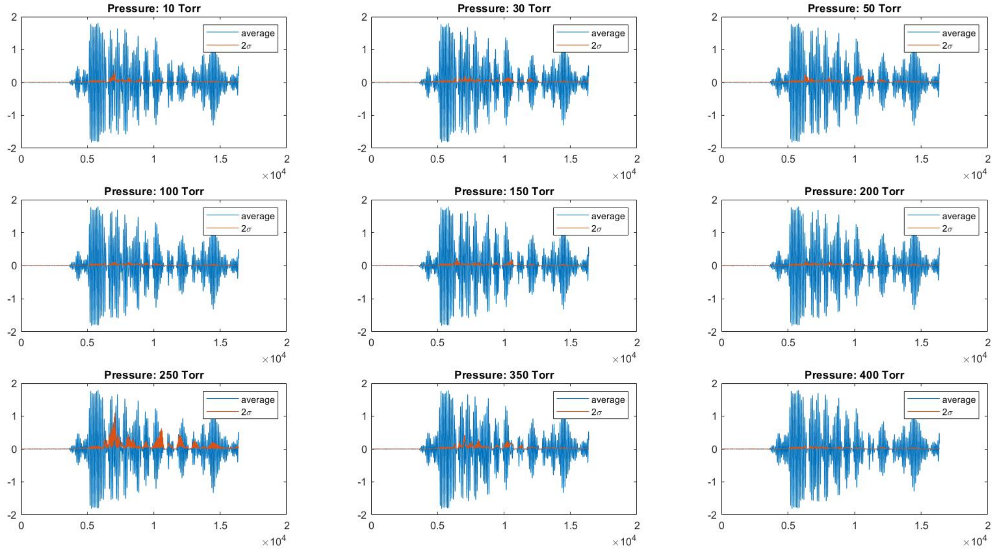

Figure B-1. Averaged time histories and standard deviation at different pressure levels

In our analysis we have also investigated the use of the *envelope function*[1]. The envelope of an oscillating signal is a smooth function outlining the signal extremities. Figure B-2 shows the difference between the averaged signal and its upper envelope at a pressure of 10 Torr.

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[1] https://www.mathworks.com/help/signal/ref/envelope.html

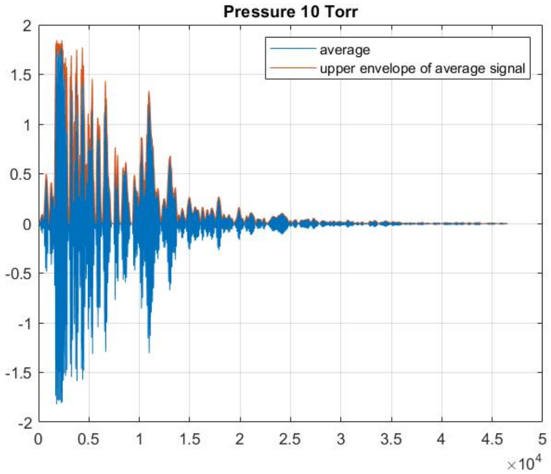

Figure B-2. Averaged and upper envelope at 10 Torr

We discovered that when the envelope signal is transformed in the time-frequency domain via a STFT, more distinguishing data structures can be discerned at different pressure levels. The STFT was applied to both the averaged and averaged-enveloped signals at different pressure levels. Figure B-3 shows the spectrograms for both cases at pressure levels ranging between 10 and 100 Torr. We observe structural vibrations induced by the input signal and propagated throughout the pipe for arrival times below 0.4 ms. These components are dominant in magnitude, but they do not contain information relative to pressure. For arrival times approximately between 0.85 and 1.1 ms, we observe that the enveloped signals (Figure B-3 on the left) present a data structure that is not shown in the purely averaged signals (Figure B-3 on the right). In addition, these data structures appear to be correlated to the pressure level.

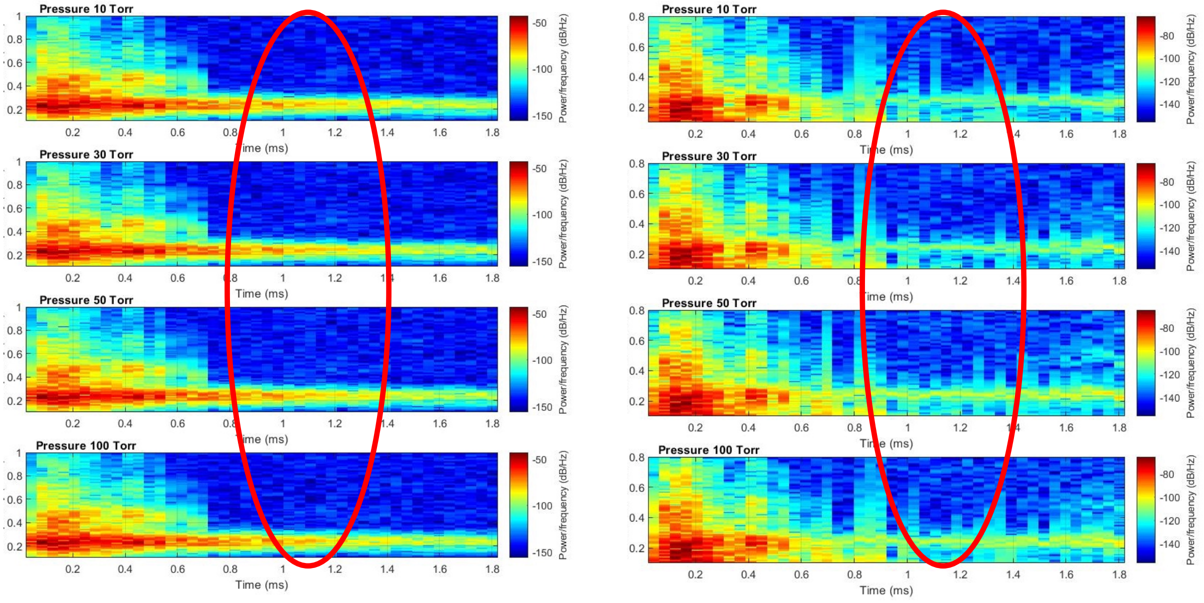

Figure B-3. Spectrograms of processed acoustic signals in frequency versus time for four different pressures. The averaged signals are on the left and the averaged-enveloped signals are on the right. The y-axis is frequency in an arbitrary scale.

In order to expand the number of features that we can extract from the raw signals, we have also applied a stationary wavelet transform (SWT) to both the averaged and enveloped-averaged signals (Nason and Silverman 1995). We use the second order Daubechies wavelet family, and the resulting detail and approximation coefficients have been transformed using a STFT. Figures 4 and 5 show the spectrograms for the approximation and detail coefficients at different pressure levels.

The results from the SWT applied to the enveloped-averaged signals is consistent with what we have observed on the spectrograms of the raw signals. Again, we observe distinctive data structures for times greater than 0.8 ms centered around a central frequency (0.25 on the arbitrary scale).

These techniques can be used to generate several images at various pressure levels and eventually used in a convolutional neural network (CNN) classifier as data input.

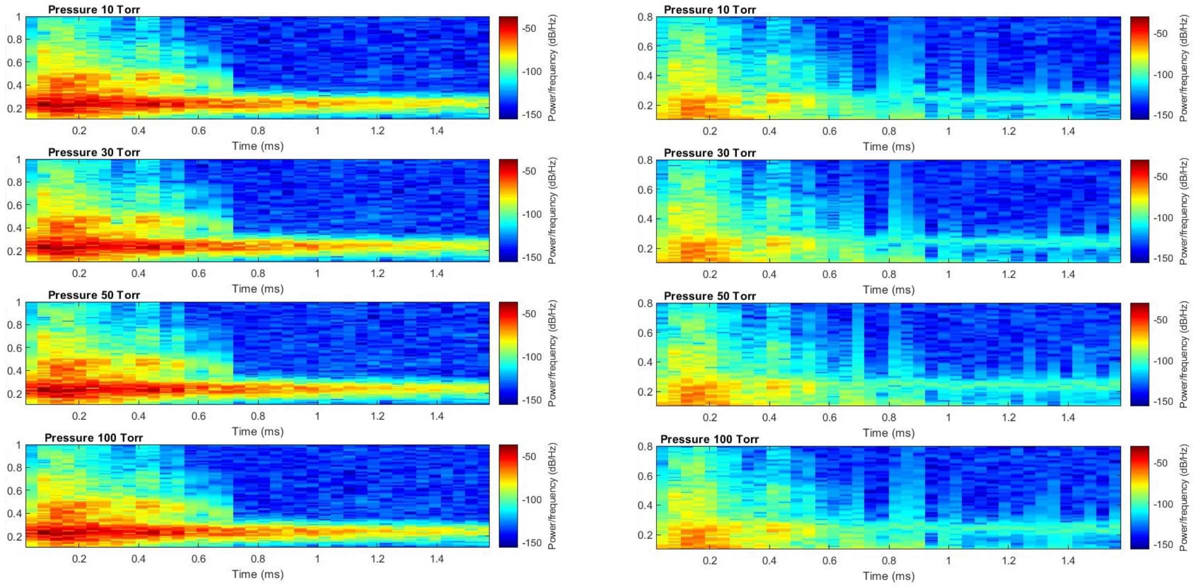

Figure B-4. Spectrograms of the SWT approximation coefficients in frequency versus time for four different pressures. The averaged signals are on the left and the averaged-enveloped signals are on the right. The y-axis is frequency in an arbitrary scale.

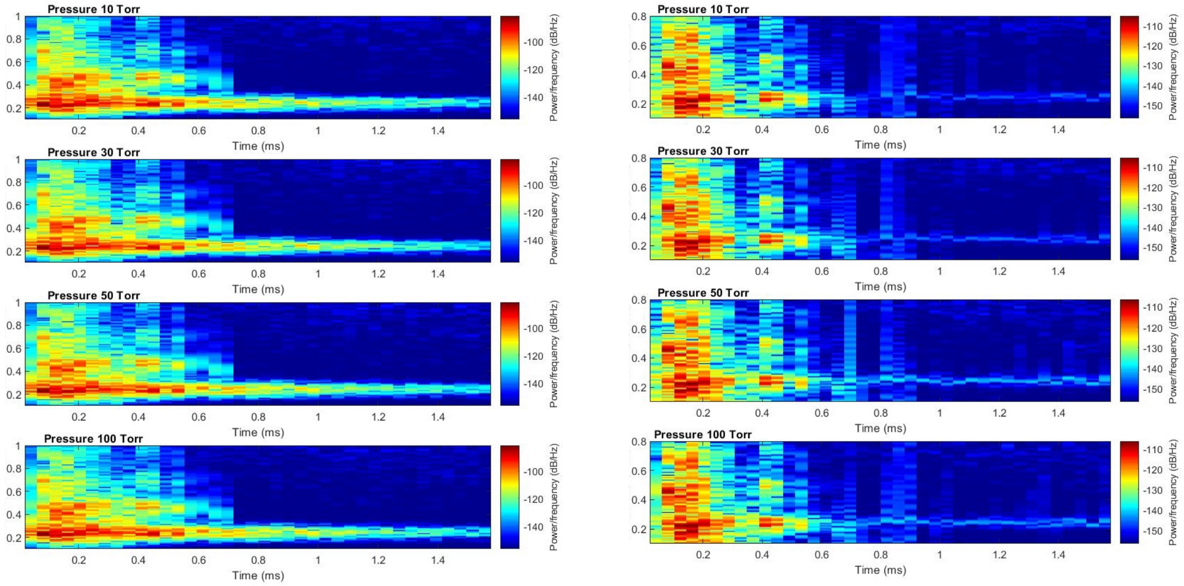

Figure B-5. Spectrograms of the SWT detail coefficients in frequency versus time for four different pressures. The averaged signals are on the left and the averaged-enveloped signals are on the right. The y-axis is frequency in an arbitrary scale.

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