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A Spouted Bed Reactor Monitoring System for Particulate Nuclear Fuel

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Abstract – *Conversion and coating of particle nuclear fuel is performed in spouted (fluidized) bed reactors. The reactor must be capable of operating at temperatures up to 2000°C in inert, flammable, and coating gas environments. The spouted bed reactor geometry is defined by a graphite retort with a 2.5 inch inside diameter, conical section with a 60° included angle, and a 4 mm gas inlet orifice diameter through which particles are removed from the reactor at the completion of each run. The particles may range from 200 μm to 2 mm in diameter. Maintaining optimal gas flow rates slightly above the minimum full spouting flow rate throughout the duration of each run is complicated by the variation of particle size and density as conversion and/or coating reactions proceed in addition to gas composition and temperature variations.*

In order to achieve uniform particle coating, prevent agglomeration of the particle bed, and monitor the reaction progress, a spouted bed monitoring system was developed. The monitoring system includes a high-sensitivity, low-response time differential pressure transducer paired with a signal processing, data acquisition, and process control unit which allows for real-time monitoring and control of the spouted bed reactor. The pressure transducer is mounted upstream of the spouted bed reactor gas inlet. The gas flow into the reactor induces motion of the particles in the bed and prevents the particles from draining from the reactor due to gravitational forces.

Pressure fluctuations in the gas inlet stream are generated as the particles in the bed interact with the entering gas stream. The pressure fluctuations are produced by bulk movement of the bed, generation and movement of gas bubbles through the bed, and the individual motion of particles and particle subsets in the bed. The pressure fluctuations propagate upstream to the pressure transducer where they can be monitored. Pressure fluctuation, mean differential pressure, gas flow rate, reactor operating temperature data from the spouted bed monitoring system are used to determine the bed operating regime and monitor the particle characteristics.

Tests have been conducted to determine the sensitivity of the monitoring system to the different operating regimes of the spouted particle bed. The pressure transducer signal response was monitored over a range of particle sizes and gas flow rates while holding bed height constant. During initial testing, the bed monitoring system successfully identified the spouting regime as well as when particles became interlocked and spouting ceased. The particle characterization capabilities of the bed monitoring system are currently being tested and refined.

A feedback control module for the bed monitoring system is currently under development. The feedback control module will correlate changes in the bed response to changes in the particle characteristics and bed spouting regime resulting from the coating and/or conversion process. The feedback control module will then adjust the gas composition, gas flow rate, and run duration accordingly to maintain the bed in the desired spouting regime and produce optimally coated/converted particles.

I. INTRODUCTION

Fluidizing particle beds are used in a wide range of applications including agricultural, pharmaceutical, high tech materials, and advanced nuclear fuel fabrication. Many advanced fuel designs utilize the advantages of small, individual fuel particle forms (Advanced Gas Reactor, dispersed fuels, space propulsion, etc.). Nuclear fuel particle calcining, sintering, and coating processes involve temperatures typically ranging from 1250°C to 1900°C in spouted bed reactors, which are specialized fluidized bed reactors with unique fluidization characteristics and particle handling advantages. Fuel particles for all future reactor designs will require very high quality fuel with low failure fractions during the fuel lifetime in the reactor. Spouted particle beds can cause damage to the circulating particles, adversely impacting particle performance in the nuclear reactors and increasing the fraction of rejected particles that will need to be recycled.

The size, density, and surface morphology of the particles change as a function of time in particle conversion or coating processes. These particle characteristics have a significant effect on the bed spouting regime and, consequently, the conversion or coating performance of the process. Real time changes to the process inlet parameters (inlet gas flow rate, inlet gas composition, and furnace temperature) are necessary to maintain optimal process operation. Changes in the particle characteristics that are not accompanied by appropriate changes in process input parameters may lead to under- or over-spouting of the bed. Under-spouting of the bed may result in particle agglomeration or faceting while over-spouting may result in particle fracture. In addition, as the conversion/coating reactions proceed and the particle characteristics evolve, process input parameter changes may be necessary to maintain the appropriate inlet gas composition and furnace temperature.

Due to the opacity of the high-temperature retorts and process gases, optical monitoring of the progress of desired reactions and controlling process parameters to compensate for changing particle properties is not possible. However, bed activity may be directly measured using back pressure data from the retort inlet gas flow. Pressure fluctuations in the gas flow are caused by specific particle motions in the particle bed during spouting. Pressure fluctuations within the reactor reveal information regarding the particle size and spouting regime, which can be used to determine optimal coating/conversion process parameters and adjust input parameters accordingly, leading to reductions in product variability and defect fractions. In order to measure these pressure fluctuations in the gas flow, a real time, high-frequency monitoring system was developed. Pressure fluctuation data was then correlated to visualization data obtained from an optically clear "mockup" retort with geometry identical to that of the high-temperature retort.

II. EXPERIMENTAL

II.A. High-Temperature Fluidizing Furnace

The spouted bed reactor retort consists of three vertically-oriented, concentric, radially-symmetric segments machined from a single piece of UCAR Carbon Company, Inc. grade CS graphite. The three segments of the retort, listed in order from bottom to top, are the inlet, base, and main chamber. The inlet has a constant internal diameter of 4 mm and a height of 78 mm (3.07 inches). The internal diameter of the retort increases as a function of height in the base, with several diameter versus height profiles possible. To date, research at the INL has been performed using a conical profile base with an included angle of 60° and a height of 51.5 mm (2.03 inches). The main chamber is a cylindrical section with a constant internal diameter of 63.5 mm (2.5 inches) and a height of 310 mm (12.2 inches).

The retort is mounted in a high temperature graphite furnace capable of operating in a temperature range from room temperature to 2300°C, depending on gas composition. The flow path through the inside of the retort is isolated from that outside of the retort to prevent particulate contamination of the furnace internals. Gas flow (composition and flow rates) inside the retort and the furnace chamber outside the retort is controlled by a combustible gas safety system. The combustible gas safety system accommodates use of flammable process gases and maintains a reducing environment in the graphite furnace by removing oxygen from the system via vacuum and inert gas purges, regulating the introduction of flammable gases into the system, and preventing diffusion of oxygen into the system by maintaining a system pressure greater than ambient pressure. The combustible gas safety system also performs controlled oxidation of process exhaust gases using an igniter.

A valve operated gravity-feed particle loading system is positioned above the retort on the outside of the furnace. The outlet of the particle loading system is directed through a port in the furnace wall and into the top of the retort main chamber.

Gas flow into the retort is controlled by up to four mass flow controllers (MKS Instruments, Inc. 1559 series) operating in parallel to provide control of inlet gas composition. The mass flow controllers are configured in series with the combustible gas safety system solenoid valves to ensure that combustible gases are not allowed to enter the system unless a successful purge sequence has been completed. Exhaust gas from the retort and furnace is cooled and filtered before passing through back flow prevention check valves, flash arrestors, and the controlled oxidation igniter.

Once a gas flow rate sufficient to retain the particles in the retort reaction zone (conical base and cylindrical main segments) has been set, the particles in the loading system are dumped directly into the retort. Particles are removed from the retort by decreasing the gas flow rate below the terminal velocity value, which allows the particles to fall through the retort inlet, past the horizontally-oriented inlet gas supply port, and into a particle collection vessel positioned directly underneath the retort outside of the furnace.

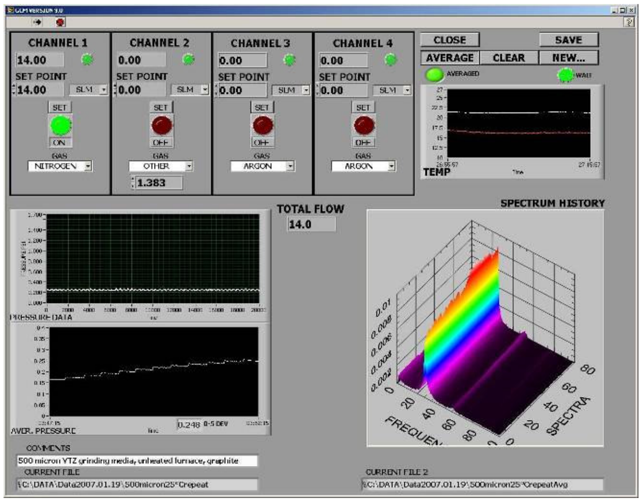
II.B. Data Acquisition and Control System

The spouted bed monitoring system provides real-time monitoring and control of the operating regime of the spouted bed. Spouted bed temperature monitoring and control is achieved through use of a Eurotherm 2404 temperature controller. Temperature readings are provided through use of a Type C thermocouple attached at the top of the furnace with the probe inserted through a port into the retort. This configuration allows positioning of the thermocouple junction in the conical base segment of the retort where direct temperature readings of the particle bed temperature may be made.

Differential pressure across the particle bed is achieved through a Sensor System Solutions 5200-B1-0005-2-3 differential pressure transducer. The high pressure side of the transducer is plumbed into the gas inlet tubing as close as possible to the retort inlet as permitted by the furnace cooling and structural support hardware. The low pressure side of the transducer is plumbed into the retort exhaust gas outlet tubing.

A National Instruments PCI-6259 high-speed multifunction M Series data acquisition board is used for data capture and process control. Signal processing and data analysis is accomplished with a software user interface written at the Idaho National Laboratory using National Instruments LabVIEW 8.0 software. A National Instruments CA-1000 shielded enclosure containing one SCB-68 and one CB-68 LPR connector block, both supplied by National Instruments, serves as the interface between the temperature, pressure, and flow monitoring and control hardware and the data acquisition board.

The monitoring system user interface includes set point controls and flow rate readings for each of the mass flow controllers supplying gas to the retort, temperature data for various areas of interest in the system, instantaneous and average differential pressure drop data, and real-time differential pressure fluctuation spectral analysis data. The data acquisition system samples at 10,000 samples per second for a two second loop duration. The high data sampling rate provides high frequency pressure fluctuation data while the long loop time provides sufficient data for low frequency pressure fluctuation analysis.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CHANNEL 4: 0.00; SET POINT 0.00 SLM; OFF; GAS ARGON
TOTAL FLOW 14.0
0.248 0-5 DEV
COMMENTS: 500 micron YTZ grinding media, unheated furnace, graphite
CURRENT FILE: C:\DATA\Data2007.01.19\500micron25°Crepeat
CURRENT FILE 2: C:\DATA\Data2007.01.19\500micron25°CrepeatAvg

Fig. 1. Spouted Bed Reactor Monitoring System Screen Capture.

II.C. Room-Temperature Bench Top Spouted Bed Experiment

An acrylic retort with dimensions identical to those of the high-temperature graphite retort was fabricated for use in making visual observations of bed spouting characteristics and to correlate these visual observations with data features/characteristics/events. The acrylic retort is mounted in a portable, bench-top configuration which can be positioned in close proximity to the spouted bed reactor monitoring system for simultaneous observation of the spouted bed and real-time data presented.

There is no temperature or pressure control capability inherent to the bench-top configuration. Therefore, operation is constrained to ambient temperature and pressure outlet conditions. Differential pressure between the spouted bed inlet and outlet are measured using a Sensor System Solutions 5200-B1-0005-2-3 differential pressure transducer with a location and orientation identical to that of high-temperature system pressure transducer; The high pressure side of the transducer is plumbed into the acrylic retort gas inlet and the low pressure side is open to atmospheric pressure conditions just as is the acrylic retort outlet. The output signal from the differential pressure transducer is wired to the spouted bed reactor monitoring system CA-1000 interface.

Gas flow control for the bench top system is achieved via the mass flow controller interface of the spouted bed reactor monitoring system. Particles are loaded directly into the top of the acrylic retort and are removed from the retort through a path leading downward through the retort gas inlet port and valve positioned physically below the gas inlet in the retort support structure.

II.D. Experimental Parameters

Two experiments were performed to illustrate the capabilities of the data acquisition and control system. The particles used for the experiments were spherical YTZ grinding media with particle diameters of 300 μm, 400 μm, and 500 μm supplied by Tosoh Corporation.

TABLE I

Variable Particle Diameter Experiment Parameters: Vary flow rate at constant temperature

Variable	Range Tested
retort	acrylic
particle diameter	300 μm, 400 μm, 500 μm
temperature	~20°C (room temperature)
inlet gas	nitrogen
flow rate	$0.75 \cdot Q_{mfs}$, $1.00 \cdot Q_{mfs}$, $1.25 \cdot Q_{mfs}$
bed height	52 mm (2.1 inches)
bed mass	255±10 grams

TABLE II

Variable Temperature Experiment Parameters: Vary flow rate with constant particle diameter

Variable	Range Tested
retort	graphite
particle diameter	500 μm
temperature	~15°C, 400°C, 800°C
inlet gas	nitrogen
flow rate	Vary from minimum flow rate required to keep particles suspended in bed to flow rate substantially above Q_{mfs}. Actual ranges vary with particle diameter.
bed mass	75 grams

In the Variable Particle Diameter Experiment, the acrylic bench top spouted bed experiment was used to generate differential pressure fluctuation data for three particle diameters. The gas flow rate was varied while holding the temperature constant. The spouted bed reactor monitoring system was used to perform real-time spectral analysis of the differential pressure fluctuation data. In the Variable Temperature Experiment, the high-temperature fluidizing furnace was used to generate differential pressure drop and differential pressure fluctuation data at three temperatures. Testing was performed by varying the gas flow rate through a particle bed of constant mass and particle diameter at each of the temperatures.

In each of the experiments the spouted bed reactor monitoring system was used to perform real-time spectral analysis of the differential pressure fluctuation data. A listing of the test parameters for each of the experiments is provided in Tables I and II.

III. RESULTS AND DISCUSSION

Several significant differences between the test procedures for the Variable Particle Diameter Experiment and the Variable Temperature Experiment existed. The first difference was the retort construction material. The Variable Particle Diameter Experiment was performed in the acrylic retort and the Variable Temperature Experiment was performed in the graphite retort. The second difference was the particle bed mass that was placed in each retort. Thirdly, the two experiments were performed under significantly different ambient pressure and temperature conditions due to use of the combustible gas safety system with the graphite retort for the Variable Temperature Experiment. The discrepancies between testing procedures of the Variable Particle Diameter Experiment and the Variable Temperature Experiment prevent direct comparison of the test data. Data and observations from the Variable Particle Diameter Experiment were used to correlate visual observations of particle bed behavior to data features and trends that were observed while using the spouted bed monitoring system during the Variable Temperature Experiment.

The first trend observed during the Variable Particle Diameter Experiment was the difference in the spectral analysis peak amplitude and frequency for equal bed heights of differently sized particles spouted at their minimum full spouting flow rate (the flow rate at which continuous bed spouting is maintained). The smaller diameter particle bed spectral analyses are characterized by lower frequency, higher amplitude peaks as compared to the larger diameter particle bed spectral analyses, which are characterized by higher frequency, lower amplitude peaks. The higher amplitude, higher energy pressure fluctuation events associated with the smaller diameter particle beds are attributed to the low interstitial space and highly tortuous flow path through the particle bed leading to the buildup of higher backpressures between pressure fluctuation events. Additionally, the greater total surface area of the smaller diameter particle bed results in increased internal bed friction, again leading to increased backpressure and higher energy pressure fluctuations. The small particle diameter frequency peak most likely occurs at a lower frequency than the larger particle diameter frequency peak due to the greater amount of time required to build pressure sufficient to overcome the internal friction associated with the small diameter particle bed. The spectral analyses illustrating these data features are presented in Figure 2.

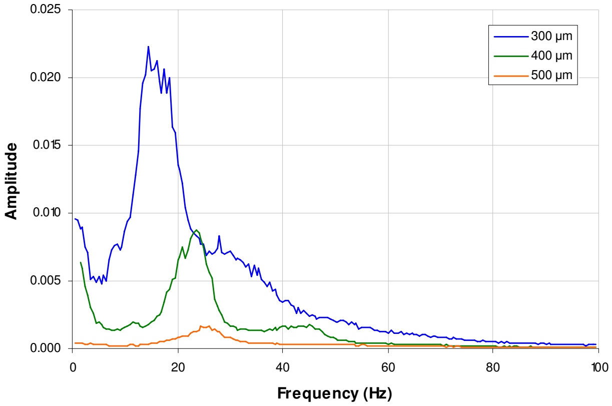

Fig. 2. Spectral analysis of 300, 400, and 500 µm constant-height particle beds operated at minimum full spouting flow rate.

Tests performed using the acrylic retort indicated that, in addition to particle size, the spectral analysis signal amplitude is correlated to the particle bed spouting regime. Visual observation of acrylic bed spouting indicated that the signal amplitude decreases markedly upon “bridging”, or the termination of particle bed circulation associated with a decrease in inlet gas flow. Similarly, the signal amplitude increases greatly when spouting commences as a result of inlet gas flow rate increases. This data trend was also observed during the Variable Temperature Experiment, despite the previously mentioned differences in retort construction material, temperature, and pressure.

Spectral analyses at selected gas flow rates for the 15°C, 400°C, and 800°C Variable Temperature Experiment tests are presented in Figures 3, 4, and 5, respectively, while spectral analyses for the full range of gas flow rates tested are presented in Figures 6, 7, and 8. Differences in the spectral analyses recorded during the Variable Particle Diameter Experiment versus the Variable Temperature Experiment included the frequency and amplitude of the dominant spectral analysis peaks. The dominant spectral analysis peak of the 500 µm particle bed test in the Variable Particle Diameter Experiment (500 µm curve in Figure 2) occupies a broader frequency band with a smaller amplitude than the dominant spectral analysis peak recorded in the minimum full spouting flow rate regime of the 15°C Variable Temperature Experiment test (8.0 slpm curve in Figure 3). It is speculated that the broadened dominant frequency peak observed during the Variable Particle Diameter Experiment was a consequence of vibration damping resulting from high velocity particles transferring energy to surrounding particles in the larger, more extensive particle bed.

Both the Variable Particle Diameter Experiment and the Variable Temperature Experiment low temperature, 500 µm particle bed test spectral analyses display a single dominant frequency peak at all flow rates tested. However, the Variable Temperature Experiment elevated temperature tests indicate the appearance of a second, lower frequency peak in the flow rate regime associated with the onset of bed spouting. The appearance of the second peak is indicative of the emergence of additional particle vibration modes or particle bed circulation associated with the thermal expansion of the inlet gas stream as it is heated to the bed temperature. The second frequency peak increases in amplitude with increasing bed temperature and broadens as a function of increasing gas flow rate.

The onset of continuous particle bed spouting in the 15°C Variable Temperature Experiment test occurs at an inlet gas flow rate of approximately 8 slpm. This event coincides with a “knee” in the 15°C pressure drop versus flow rate data trace included in Figure 9 and an increase in the amplitude of the spectral analysis dominant frequency peak included in Figures 3 and 6.

Continuous particle bed spouting in the 400°C test occurs at a flow rate of approximately 8 slpm. As in the case of the 15°C test, this event coincides with a knee in the pressure drop versus flow rate curve included in Figure 9. However, the knee follows a segment of relatively constant pressure drop as a function of gas flow rate that corresponds to a spectral analysis with lower signal amplitude than the surrounding flow regimes. A second frequency peak appears when the minimum full spouting flow rate of 8 slpm is achieved. Further increases in flow rate are accompanied by increases in the second frequency peak amplitude and width.

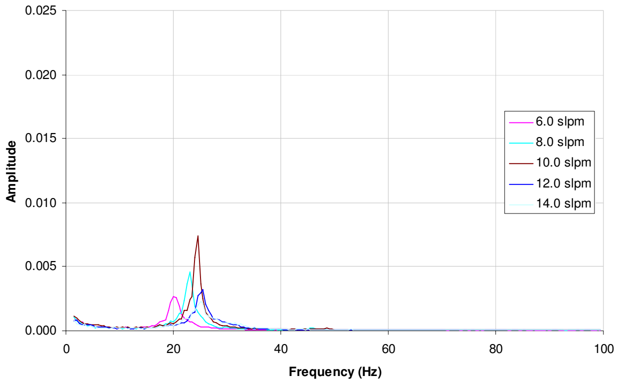

Fig. 3. Spectral analyses of 500 μm diameter spherical YTZ particle bed at selected gas flow rates during 15°C test.

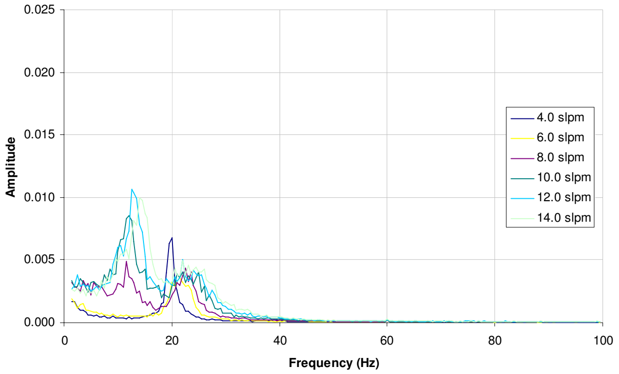

Fig. 4. Spectral analyses of 500 μm diameter spherical YTZ particle bed at selected gas flow rates during 400°C test.

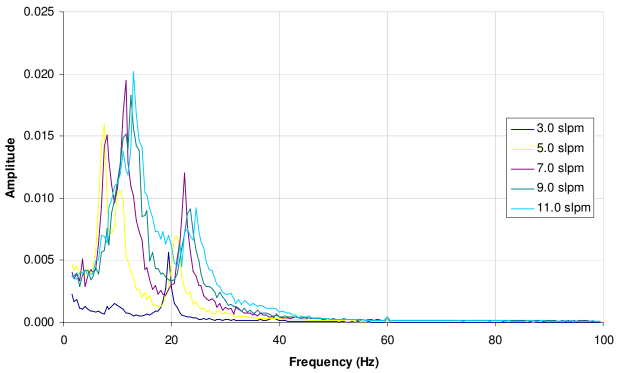

Fig. 5. Spectral analyses of 500 μm diameter spherical YTZ particle bed at selected gas flow rates during 800°C test.

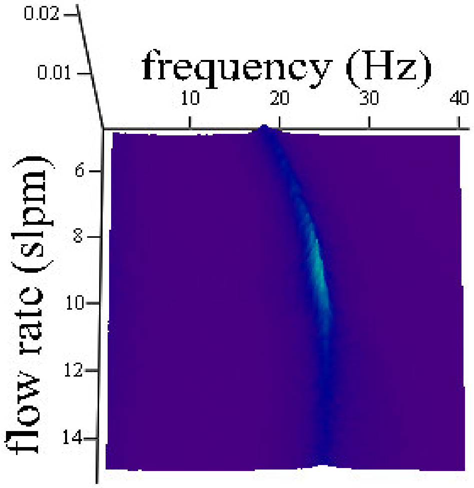

Fig. 6. Spectral analysis, 500 μm particle diameter, 15°C.

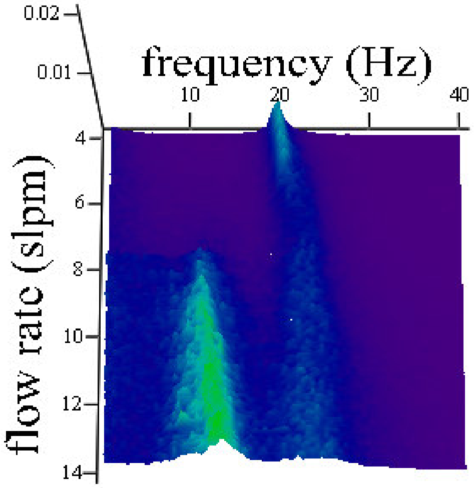

Fig. 7. Spectral analysis, 500 μm particle diameter, 400°C.

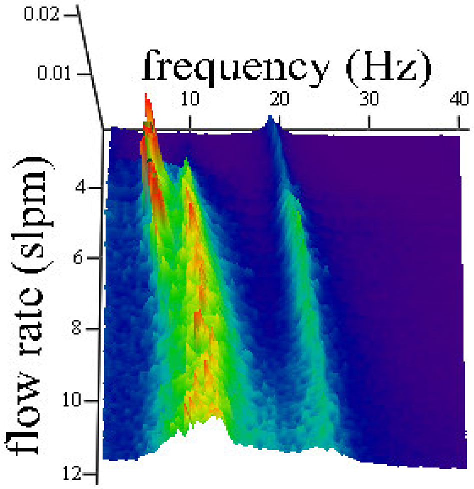

Fig. 8. Spectral analysis, 500 μm particle diameter, 800°C.

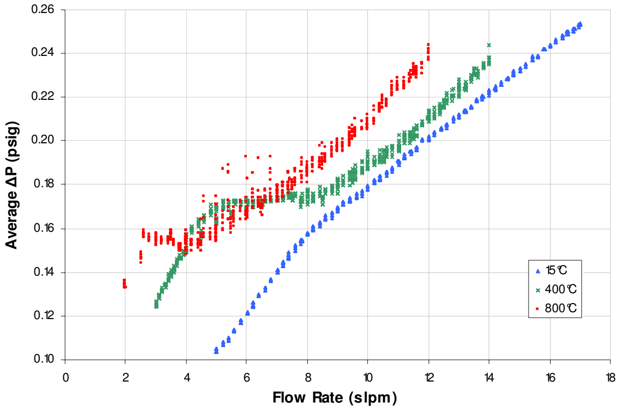

Fig. 9. Nitrogen-spouted 500 µm spherical YTZ.

The 800°C test undergoes a transition to particle bed spouting, as indicated by the pressure drop versus flow rate plot, presented in Figure 9. The transition occurs between 4 and 7 slpm. In the transient zone unsustainable bed spouting exists, and the pressure drop versus flow rate data plot is enveloped by two distinct curves corresponding to bed spouting and bed surface stagnation. In the flow rate range below the transition zone (3.0 slpm curve in Figure 5), the spectral analysis signal amplitude is a minimum. In the transitional flow rate range (5.0 and 7.0 slpm curves in Figure 5), the second frequency peak appears with a significantly greater amplitude than that of the initial peak before decreasing in amplitude and shifting to a higher frequency at the higher flow rates where the bed is fully spouted, (9.0 and 11.0 slpm curves in Figure 5).

Testing performed with an identical particle bed mass in the acrylic and graphite retorts indicated that the minimum fluidization velocity is greater for particles in the acrylic retort. A number of factors may account for this phenomenon. It is likely that the major factor responsible for this discrepancy is the increased gas density present in the graphite retort due to the elevated pressure spouted bed operation imposed by the combustible gas safety system. In addition, during acrylic retort spouting tests static charge buildup in the particle bed and a resulting change in spouting behavior were observed. The electrical conductivity of graphite is superior to that of acrylic, and the static electricity buildup in the graphite retort is expected to be minimal compared to that in the acrylic retort.

IV. CONCLUSIONS

Preliminary spouted bed tests have been performed to validate experimental equipment performance and collect pressure drop versus flow rate and pressure fluctuation spectral analysis data. Visual observations recorded using the acrylic retort experimental setup correlate to specific data features and events, which can in turn be used to describe spouted bed characteristics in the graphite retort experimental setup.

Future testing will focus on collecting data over an increased range of particle diameters, particle densities, bed masses, gas flow rates, gas compositions, and furnace temperatures. The pressure fluctuation spectral analysis and visual observation data collected will be used to develop input parameters for the next version of the spouted bed reactor real-time monitoring and feedback control system to maintain optimum bed spouting, gas composition, and furnace temperature conditions. An additional planned feature of this data acquisition and control system will calculate particle size and control run duration.

NOMENCLATURE

ΔP	pressure drop
Q_{mfs}	minimum full spouting flow rate

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