

# DEVELOPMENT OF A MOLECULAR TAGGING VELOCIMETRY TECHNIQUE FOR NON-INTRUSIVE VELOCITY MEASUREMENTS IN LOW-SPEED GAS FLOWS

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## ABSTRACT

N<sub>2</sub>O molecular tagging velocimetry (N<sub>2</sub>O-MTV) is developed for use in very-high-temperature reactor environments. Tests were carried out to determine the optimum excitation wavelength, tracer concentration, and timing parameters for the laser system. Using NO tracers obtained from photo-dissociation of N<sub>2</sub>O, velocity profiles are successfully obtained in air, nitrogen, and helium for a large range of parameters: temperature from 295 to 781 K, pressure from 1 to 3 bars, with a velocity precision of 0.01 m/s. Furthermore, by using two read pulses at adjustable time delays, the velocity dynamic range can be increased. An unprecedented dynamic range of 5,000 has been obtained to successfully resolve the flow during a helium blowdown from 1000 m/s down to 0.2 m/s. This technique is also applied to the high-temperature test facility (HTTF) at Oregon State University (OSU) during a depressurized condition cooldown (DCC) event. Details of these measurements are presented in a companion paper. This technique shows a strong potential for fundamental understanding of gas flows in nuclear reactors and to provide benchmark experimental data to validate numerical simulations.

## KEYWORDS

Molecular tagging velocimetry, validation data, gas cooled reactors

## 1. INTRODUCTION

Molecular tagging velocimetry (MTV) is a time-of-flight velocity measurement technique that relies on locally creating and tracking molecular tracers [1]. A first laser pulse (or write pulse) creates these tracers with a predetermined spatial pattern, and then a second laser pulse (read pulse) or pulses excite a cross-section of the flow with a controlled time interval. The location of the displaced tracers is recorded for each read pulse with a camera, ultimately leading to velocity profiles. In gas flows, radicals are typically created through photo-dissociation of specific seed molecules and are tracked with planar laser-induced fluorescence. MTV has been demonstrated in gas with a variety of tracers. Examples of seed gas include N<sub>2</sub>O [3], NO<sub>2</sub> [4], Kr [5], acetone [6], biacetyl [7], O<sub>2</sub> [8, 9], N<sub>2</sub> [10], and H<sub>2</sub>O [2,9]. This non-intrusive technique is applicable to a wide variety of flows ranging from stagnant to hypersonic, from cryogenic to flame temperature, and over a large range of pressure.

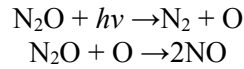
These characteristics make MTV attractive for experimental studies of very-high-temperature gas-cooled nuclear reactor (VHTR). In both the stratified and natural circulation flows that occur in these facilities, which typically have low velocities (below 1 m/s), MTV requires tracers that persist for a long duration to obtain resolvable displacements. This is typically not possible with particle-based methods because settling would be an issue, in addition to contamination of the facility.

The present work focuses on NO tracers obtained from N<sub>2</sub>O (nitrous oxide) seed gas and demonstrates their use in thermodynamic conditions similar to that of VHTR. The velocity measurement precision, dynamic range and other aspect aspects of this technique are discussed.

## 2. EXPERIMENTAL TECHNIQUE

### 2.1. N<sub>2</sub>O Photo-chemistry

The NO tracers are created from the photo-dissociation of the N<sub>2</sub>O seed gas according to the following reactions:



where  $h$  is Planck's constant and  $\nu$  the photon frequency. An excimer laser with a 193 nm wavelength is used for the photo-dissociation, as detailed in the following section. The first reaction relates to the photo-dissociation ratio, given by the equation:

$$\frac{n_d}{n_i} = \frac{\sigma E}{h\nu A}$$

$\sigma$  is the photo-dissociation cross section, equal to  $8.95 \times 10^{-20}$  cm<sup>2</sup> for N<sub>2</sub>O. The cross section slightly increases with temperature.  $E$  and  $A$  are the beam energy and cross sectional area, respectively. NO is stable in inert environment such as helium or nitrogen, which is useful for increasing the probe time to reach high velocity measurement precision. Molecular diffusion of the tracers limits the probe time by spreading the tag line. The latter increases proportionally to  $dt$  to the first order.

### 2.2. Molecular tagging velocimetry system

The diagnostics rely on several lasers and a camera synchronized together. The seed gas is initially dispersed in the flow being probed. A first laser beam (the write pulse) is from an excimer laser (GAM Laser EX5) at 193 nm. This beam is gently focused by a 1.5 m focal length lens and photo-dissociates the N<sub>2</sub>O in its path. The NO molecules are created nearly instantaneously (<10 ns) and form a line, the location of which is to be measured subsequently using fluorescence. Two laser pulses shaped in a 3-mm

thick sheet (read pulses) are then emitted from a tunable dye laser (Sirah Cobra-Stretch) at a wavelength of 226.1929 nm to illuminate the NO tracers and induce fluorescence. An intensified camera (QImaging QIClick CCD camera coupled with a LaVision IRO intensifier) records the fluorescent signal and rejects the laser light with a long pass filter (280 nm cut-off), informing on the location of the tracers at two different instants in time. The velocity is then obtained by measuring the displacement using cross-correlation techniques and knowing the delay between the two images. Since only a single tag line was used in these studies, only a single component of velocity can be resolved.

### 2.3. Test Section

A stainless steel pressure vessel was built to investigate the performances of the diagnostics in conditions as encountered in a VHTR, namely high pressure and high temperature. This test section is fitted with two UV-transparent fused-silica viewports to allow the lasers beam to enter it and the fluorescent signal to be observed.

A 9-mm diameter jet is located vertically in the center of the test section to generate a well-controlled flow of nitrogen or helium containing a small fraction of N<sub>2</sub>O that can be probed with N<sub>2</sub>O-MTV. A National Instruments data acquisition system controls and monitors pressure and temperature in the chamber, as well as jet flow rate and N<sub>2</sub>O molar fraction.

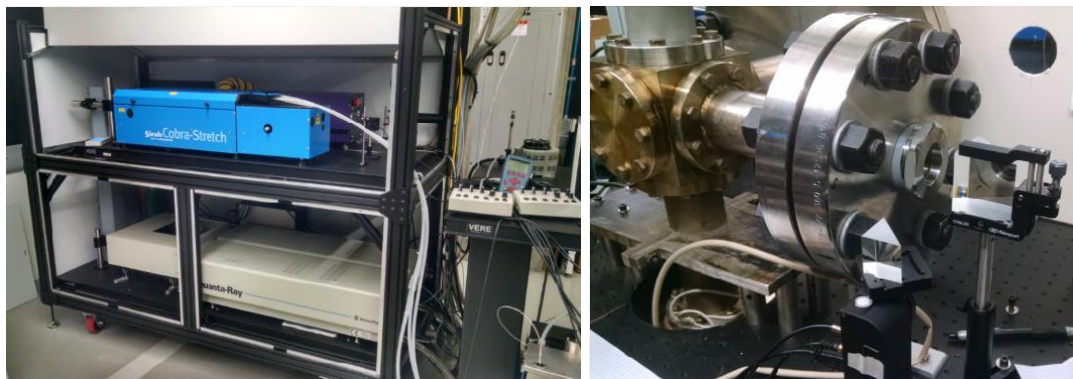
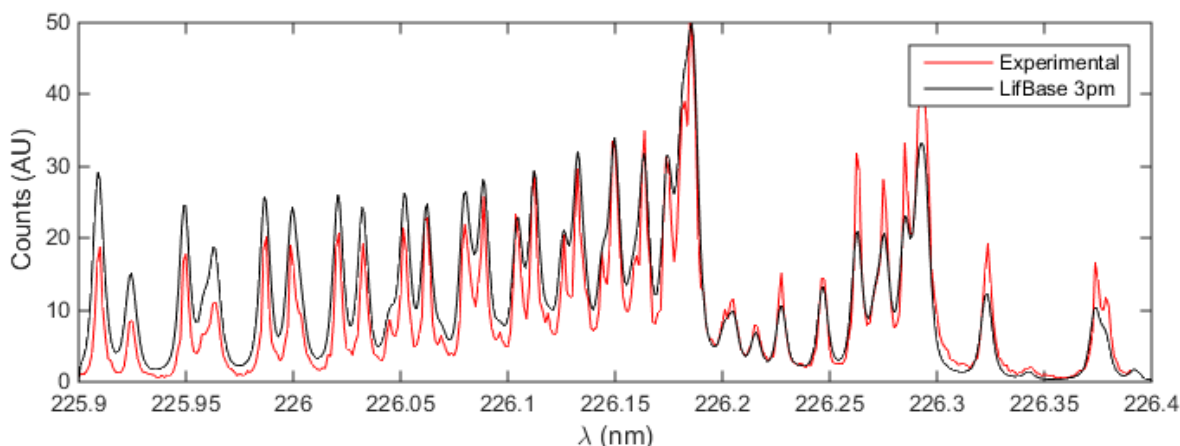


Figure 1. Laser system (left). Test section with laser viewport (right)

## 3. RESULTS

### 3.1. Tracer Spectrum and wavelength selection

The fluorescence excitation spectrum of NO is first measured and analyzed to identify the optimum wavelength for the dye laser (read pulse). The MTV signal is recorded while the dye laser scans the wavelength, all other parameters (laser power, N<sub>2</sub>O concentration, probe time) being held constant. The scan is run between 225.9 and 226.4 nm and results are presented in Figure 2 for ambient temperature and pressure. The emission spectrum of NO computed by the software LIFBASE [11] is also plotted on this figure and shows a good agreement with the experiment. The strongest signal (highest peak) is obtained at a wavelength of 226.1929 nm which is the Q-branch band head.



**Figure 2. NO excitation spectrum at ambient pressure and temperature. Experiment versus simulation. A laser linewidth of 3 pm was assumed in the simulation.**

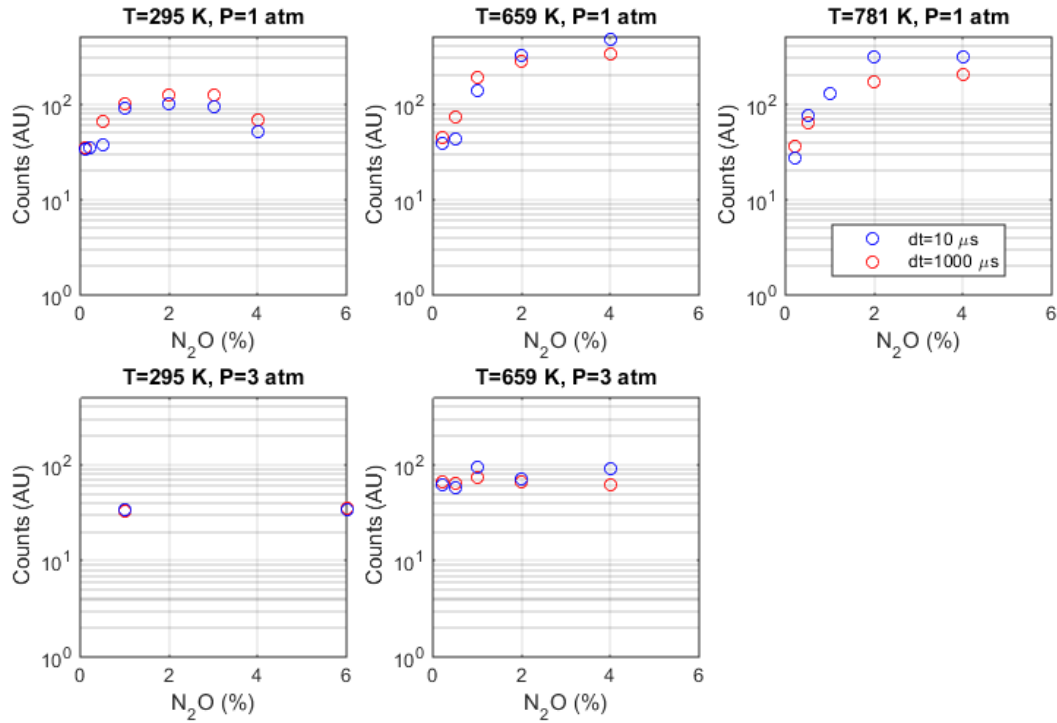
Tests were also performed at higher pressure (up to 3 bars) and temperature (up to 500 °C). While pressure and temperature affects the peak height and width through collisional quenching and broadening, the best signal was still obtained at the same wavelength, which ensured the dye laser would not need to be tuned for each pressure or temperature condition. Therefore the rest of the study was done with the read pulses at 226.1929 nm.

### 3.2. Effect of temperature, pressure, probe time, and N<sub>2</sub>O concentration

For the data described in this section the NO signal is measured by keeping the wavelength of the read pulse constant (where the signal is maximum) and the temperature, pressure, and N<sub>2</sub>O concentration are varied. The tracers are probed at two different times of 10 and 1000 μs after being created by the write pulse. The tracers are probed non-consecutively, with only one probe pulse. The results are summarized in the plots of figure 3. Each sub figure presents a different combination of pressure/temperature with the first row at ambient pressure, and the first column at ambient temperature. Maximum temperature and pressure in this study are 781 K (508 °C) and 3 bars, respectively. The N<sub>2</sub>O concentration is varied between 0.1 and 6% by volume. Note that the only previous study on N<sub>2</sub>O-MTV was done at 4% [3].

The main findings are:

- Signal increases with temperature, which is convenient for VHTR studies since the flow can be up to 900 °C.
- Signal decreases with an increase in pressure. This can be problematic at low temperature (lower left subfigure has the lowest signal of all), but this detrimental factor can be offset by the gain obtained from temperature increase, as shown for the 659 K, 3 atm plot.
- Signal intensity is similar between 10 and 1000 μs, which confirms the chemical stability of NO over this duration. Further study of the lifetime showed successful probing of NO for at least 20 ms with molecular diffusion being the limiting factor. The effect of the probe time delay on the velocity measurement is discussed in the following section (3.3).
- Signal increases with N<sub>2</sub>O concentration for low concentration (<2%). Above 2%, the signal either plateaus or decreases. This is discussed in more details in section (3.3).
- Although the only N<sub>2</sub>O measurements reported in literature used 4% N<sub>2</sub>O, these results show that data can still be obtained at sub-one percent concentration.



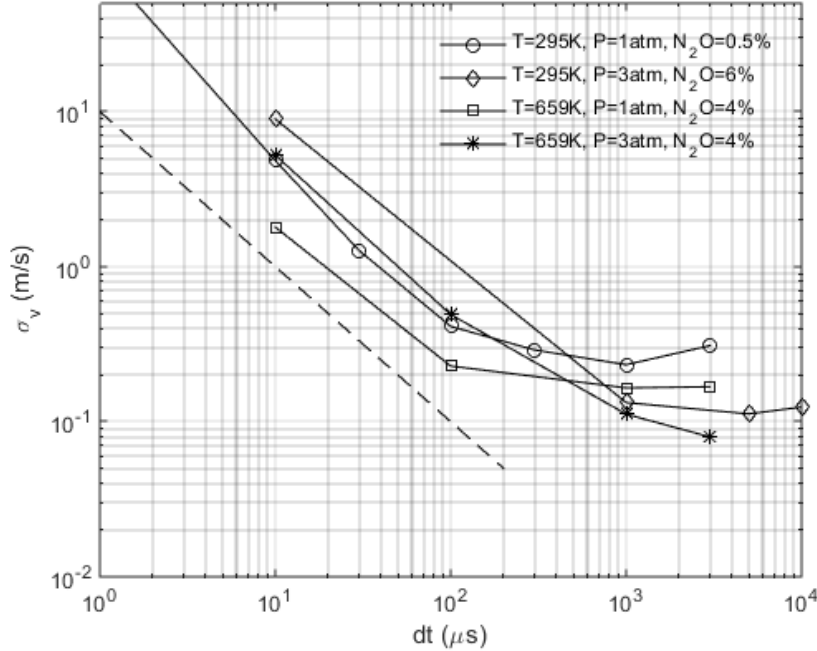
**Figure 3. NO signal for various temperature, pressure, and N<sub>2</sub>O seed gas concentration**

### 3.3. Velocity measurement precision

MTV is a time-of-flight technique, where the velocity is obtained by measuring a displacement over a period of time according to the equation  $V = \Delta x / dt$  with  $\Delta x$  the tracer displacement, and  $dt$  the probe delay time. Since the resolution of the displacement measurement is limited by the camera resolution (discretized in pixels), larger  $dt$  are required to probe slower flows for a given magnification (magnification being set by the size of the field of view, which is largely determined by the lens used, distance of the lens from the CCD, working distance and CCD sensor size). The precision of the velocity measurement is then expressed as

$$\sigma_v = \sigma_x / dt$$

The precision on the measured displacement is a function of the camera, optics, and processing algorithm, as well as signal quality (typically characterized by the signal-to-noise ratio –SNR). The following figure presents the measured precision for 4 cases with different pressure, temperature and N<sub>2</sub>O concentration as a function of probe time.



**Figure 4. Velocity measurement precision versus probe time.**

The dashed line on Figure 4 has an arbitrary height and a slope of -1 (in log-log plot) to represent the aforementioned theoretical relationship between velocity precision and probe time. At low probe time ( $<100 \mu\text{s}$ ), the SNR is high and diffusion is negligible, thus the precision of the displacement is constant, and the experimental data follow the -1 slope trend line. As  $dt$  increases further, the precision keeps decreasing (i.e. improves), but departs from the theoretical trend. This is because the jet in this test was convecting the tracers out of the probe sheet. Precision down to 0.1 m/s are obtained for this set of experiments. Improvement of the setup (reducing jet velocity and enlarging probe laser sheet height) yielded precision down to 0.01 m/s at probe time of 10 ms. This precision is for a given magnification and field of view, which limits the maximum displacement that can be measured. This corresponds to about 1 m/s, and thus the technique has a velocity dynamic range of 100:1, which is similar to that of particle image velocimetry (PIV).

### 3.4. Optimization and Outlook

To be deployable to VHTR experiments, additional challenges must be addressed concerning the  $\text{N}_2\text{O}$ -MTV technique.

#### 3.4.1. Seed Gas Concentration

Because  $\text{N}_2\text{O}$  has a relatively large absorption cross section, it is easily photo-dissociated by the excimer laser. However, this also means that the excimer beam will be attenuated as it travels through the gas mixture (nitrogen+ $\text{N}_2\text{O}$ ). The transmission efficiency is:

$$\frac{E}{E_0} = e^{-\sigma n_i L}$$

with  $L$  the path-length of the beam in a gas containing a concentration  $n_i$  of  $\text{N}_2\text{O}$ . The exponential nature of this equation indicates that the beam will be quickly absorbed if the quantity  $\sigma n_i L$  is large.  $\sigma$  is a physical constant, and  $L$  is geometrical constraint which corresponds to the distance the laser beam has to

travel in the experiment between the viewport and the measurement region. For integral effect test facility, this distance can easily be on the order of meters, which put strong limitations on the concentration of  $\text{N}_2\text{O}$  to avoid beam attenuation. Other considerations in wanting to reduce the  $\text{N}_2\text{O}$  concentration include cost, safety and the desire to minimally perturb the flow system.

By combining the equation for the photo-dissociation efficiency and beam attenuation, the optimal  $\text{N}_2\text{O}$  concentration to maximize the amount of dissociated molecules for a given path-length is  $n_{opt} = \sigma L$ . For  $L=30$  cm, the optimum  $\text{N}_2\text{O}$  concentration is 1.5%. For  $L=100$  cm, it is 0.45%. The previous results showed that data can be obtained with concentration as low as 0.2% with the present setup, which corresponds to a path-length of 225 cm. Longer path lengths could still be acceptable with lasers of higher power. Alternatively, the beam could also be enclosed in a  $\text{N}_2\text{O}$ -free environment within the experiment until it reaches the measurement section (provided this enclosure does not disturb the flow).

### 3.4.2. Velocity Dynamic Range

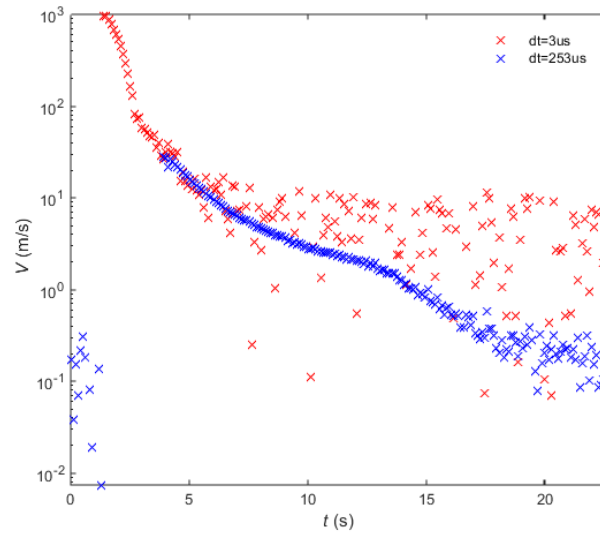
The velocity measurement relies on the displacement of the tracers between two instants in time. In the present work, this is accomplished by two consecutive read pulses. However, most MTV studies to date have used only one read pulse. The initial location of the tracers is recorded in a separate step by firing the read pulse within a few nanoseconds of the write pulse. The assumption that the initial tracer location is constant is usually reasonable when doing measurements in the laboratory, where the environment is well controlled. Factors potentially affecting the position of the write beam are vibrations, beam steering through turbulent flow and thermal expansion. Therefore, it is preferable to rely on two probe beams when doing field measurements where such issues may occur.

In more controlled experiments where the initial tracer location is constant, the two probe pulses can be taken advantage of to increase the velocity dynamic range. For instance, a first pulse can be fired after a short delay, allowing measurement of high speed flows, and then a second pulse is fired after a longer time, enabling the measurement of lower velocities with high precision. But this approach could be susceptible to the previously mentioned error factors (vibration, etc.).

The velocity dynamic range of the MTV system was investigated. This test was performed with HO tracers created from  $\text{H}_2\text{O}$  seed gas. This technique, called hydroxyl tagging velocimetry (HTV) [2], is similar to  $\text{N}_2\text{O}$ -MTV, though HTV generally has a weaker signal at long  $dt$ . Similar tests would work even better with  $\text{N}_2\text{O}$ -MTV. In this test, a small chamber (5.7 liters) is pressurized at 3.5 bars with Helium and a  $\frac{1}{4}$ " diameter valve is then quickly opened to vent the chamber to the atmosphere. Such flow would be similar to a blowdown, as is experienced during the depressurization phase of a DCC. Velocity is measured with MTV at the valve exit. The first read pulse is 3  $\mu\text{s}$  after the write pulse and is used for resolving high speed flow ( $\sim 100$  m/s). For low flow speed (order of 1 m/s), the displacement is very small, and the precision is poor. The lower speed part of the flow is precisely resolved with a longer  $dt$ , of 253  $\mu\text{s}$ . The initial tracer location is obtained from measurements before the opening of the valve, when the flow is quiescent and the line is not convected.

The jet mean velocity as a function of time is plotted in figure 5. The valve opens at  $t=1.5$  s. The flow is initially fast (limited by the speed of sound in the gas,  $\sim 1,000$  m/s for helium), and is correctly captured by the first pulse at  $dt = 3$   $\mu\text{s}$ . The second pulse at  $dt = 253$   $\mu\text{s}$  is not visible because out of the camera FOV. When the flow is slow enough ( $< 20$  m/s), the second pulse becomes visible and is then used to calculate the velocity.

The first pulse is still visible, but the very small displacement makes the measured velocity very imprecise (on the order of 10 m/s). The precision on the second pulse measurement is about 0.2 m/s. Overall, the flow is resolved from 1000 m/s down to 0.2 m/s, which corresponds to a dynamic range of 5,000:1. Using a single pulse allow a dynamic range of only 100:1.



**Figure 5. Jet velocity versus time for a helium blowdown**

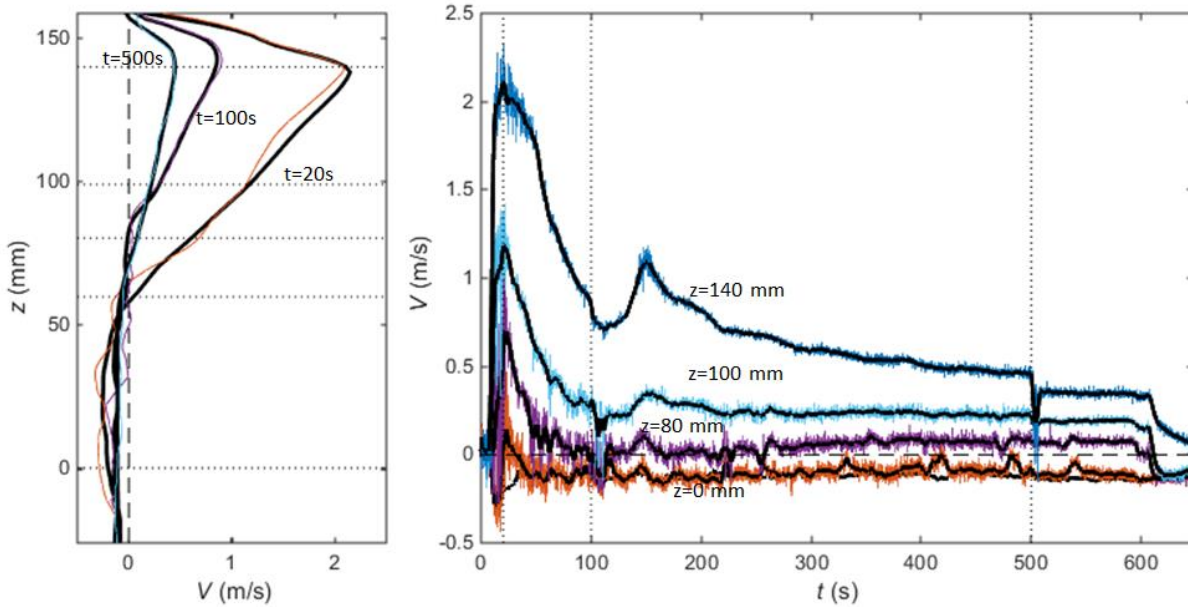
#### **4. DEMONSTRATION OF THE DIAGNOSTICS ON THE HTTF AT OSU**

The diagnostics described were designed to be deployed to the High Temperature Test Facility (HTTF) at Oregon State University (OSU) to perform velocity measurements during a depressurized conduction cooldown (DCC) in this Integral Effect Test (IET) facility. The implementation of this technique to the HTTF and detailed results are presented in a companion paper and presentation. Sample results are presented here to illustrate to capabilities of the  $N_2O$ -MTV method for VHTR investigations.

The HTTF replicates a DCC by opening both hot and cold leg of the primary to a large cavity (RCST) simulating the ambient atmosphere surrounding the reactor. Velocity profiles are measured at the exit of the hot leg at a frequency of 10 Hz for 30 minutes. In a DCC scenario, simulations predict helium to flow out of the reactor, and air (here nitrogen is used a surrogate) to ingress into the reactor, in a buoyancy-driven flow. Figure 6 shows velocity profiles and time evolution of the velocity during an isothermal DCC. These results are the first detailed velocity data obtained from an IET, and they reveal the complexity of the evolution of the flow during such events.

The high measurement precision was crucial in these experiments because of the low velocities experienced in the HTTF ( $< 2\text{m/s}$ ). The laboratory investigation of the effect of temperature/pressure of the NO signal,  $N_2O$  concentration, and timing parameters of the tests that is detailed in this paper made the *in-situ* measurements successful.





**Figure 6. Velocity result for isothermal DCC. Left figure shows velocity versus location (spatial profiles) at the instants marked by a dotted line on the right figure. The right figure shows velocity versus time (temporal profiles) at the location indicated by dotted lines on the left figure. Thick black lines are locally averaged profiles**

## 5. CONCLUSIONS

A non-intrusive velocity measurement technique based on molecular tagging has been investigated to determine its suitability for gas flow measurements in VHTR conditions. NO tracers obtained from N<sub>2</sub>O seed gas allow precise measurements over a large range of temperature (295 to 781K) and pressure (1 to 3 atm). We also demonstrate that the required concentration of seed gas can be significantly lower than in previous studies (0.2% vs 4%), which minimizes the change of the thermo-physical properties of the flow under investigation. The probe time of NO can be at least 10 ms which is much longer than used in previous work. This long delay gives adequate precision (0.02 m/s) when measuring low speed flows (<2 m/s).

On the other hand, high-speed flow such as during a blowdown can also be probed by using a shorter probe time. High and low speed flows can be simultaneously probed by taking advantage of the dual-probe pulse capacity of the current system since vibration, beam steering and thermal expansion do not appear to be significant in the current tests. A dynamic range of 5,000:1 is demonstrated by measured the velocity of a blowdown jet from 1,000 m/s to 0.2 m/s.

The N<sub>2</sub>O-MTV technique is readily implementable on VHTR-type facility. Data have been recorded during a DCC on the HTTF at OSU, and are presented in more details in a companion paper.

## ACKNOWLEDGMENTS

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## REFERENCES

1. Springer Handbook of Experimental Fluid Mechanics, ed. Topea, Yaris, Foss, ch. B.5.4.

2. M Andre, P Bardet, R Burns, and P Danehy. "Development of Hydroxyl Tagging Velocimetry for Low Velocity Flows", 32nd AIAA AVIATION Forum, (AIAA 2016-3247)
3. AM ElBaz and RW Pitz. N<sub>2</sub>O molecular tagging velocimetry. *Applied Physics B*, 106(4):961-969, 2012.
4. PM Danehy, S O'Byrne, AF Houwing, JS Fox, and DR Smith. Flow-tagging velocimetry for hypersonic flows using fluorescence of nitric oxide. *AIAA journal*, 41(2):263-271, 2003.
5. NJ Parziale, MS Smith, and EC Marineau. Krypton tagging velocimetry of an underexpanded jet. *Applied optics*, 54(16):5094-5101, 2015.
6. WR Lempert, N Jiang, S Sethuram, and M Samimy. Molecular tagging velocimetry measurements in supersonic microjets. *AIAA journal*, 40(6):1065-1070, 2002.
7. B Stier and MM Koochesfahani. Molecular tagging velocimetry (MTV) measurements in gas phase flows. *Experiments in Fluids*, 26(4):297-304, 1999.
8. A Noullez, G Wallace, W Lempert, RB Miles, and U Frisch. Transverse velocity increments in turbulent flow using the RELIEF technique. *Journal of Fluid Mechanics*, 339:287-307, 1997.
9. RW Pitz, JA Wehrmeyer, LA Ribarov, DA Oguss, F Batliwala, PA DeBarber, S Deusch, and PE Dimotakis. Unseeded molecular flow tagging in cold and hot flows using ozone and hydroxyl tagging velocimetry. *Measurement Science and technology*, 11(9):1259, 2000.
10. PM Danehy, BF Bathel, N Calvert, A Dogariu, and RP Miles. Three-component velocity and acceleration measurement using FLEET. In 30th AIAA Aerodynamic Measurement Technology and Ground Testing Conference, page 2228, 2014.
11. J Luque and DR Crosley. LIFBASE: database and spectral simulation program (version 1.5). SRI international report MP, 99(009), 1999.