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# RPPR Final Report
as of 18-Mar-2019

Agency Code:

Proposal Number: 72342EGII **Agreement Number: W911NF-18-1-0147**

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Phone: (608) 890-2271
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**Major Goals:** The project had three primary objectives/goals as proposed. These three goals were:
1. Determine the feasibility and implementation of the co-doped ratio method to be used to obtain improved fractional sensitivity for aerosol phosphor thermometry (APT).
2. Determine the feasibility of the Mie ratio method to provide improved accuracy for APT measurements.
3. Demonstrate improvements using the two methods in a heated air jet.

**Accomplishments:** Detailed accomplishments for the project are contained in the attached PDF document.

**Training Opportunities:** The project was staffed by a post doc. The post doc for the project was able to continue their training as a researcher and in particular to develop their ability to independently perform research.

**Results Dissemination:** Results from the work were disseminated via five conference presentations, a conference abstract, two conference papers, and two journal articles.

Results for the co-doped aerosol phosphor thermometry (APT) method were presented at the 37th International Symposium on Combustion in Dublin, Ireland (July 29-August 3, 2018), followed by publication in the biennial Proceedings of the Combustion Institute (PROCI) journal.

Results for the scattering-referenced aerosol phosphor thermometry (SRAPT) technique were disseminated in a conference paper and presentation at the 2018 Spring Technical Meeting for the Central States Section of the Combustion Institute (Minneapolis, MN, May 20-22, 2018) and in a presentation at the Inaugural International Conference on Phosphor Thermometry (ICPT - Glasgow, UK, July 25-27, 2018). Following the ICPT conference, we were invited to submit a journal article for a special issue of Measurement and Science Technology, which was recently accepted for publication.

Results for the combined co-doped APT and SRAPT techniques have recently been accepted for presentation at the 11th U.S. National Combustion Meeting (Pasadena, California, March 24-27, 2019) and the associated conference paper has been submitted.

The principal investigator also presented results from the co-doped APT and SRAPT work in an invited presentation at the AIAA 2018 AVIATION Forum (Atlanta, GA, June 25-29, 2018)

**Honors and Awards:** The principal investigator also presented results from the work in an invited presentation at the AIAA 2018 AVIATION Forum (Atlanta, GA, June 25-29, 2018).

**Protocol Activity Status:**

**Technology Transfer:** Due to the short duration of the project their was limited time for interaction with external entities such as DoD laboratories. As the technique is further developed in the future we hope to interact of DoD laboratories to aid in implementation of the techniques studied.

**PARTICIPANTS:**

**Participant Type:** Postdoctoral (scholar, fellow or other postdoctoral position)
**Participant:** Dustin Witkowski
**Person Months Worked:** 6.00 **Funding Support:**
Project Contribution:
International Collaboration:
International Travel:
National Academy Member: N
Other Collaborators:

**CONFERENCE PAPERS:**

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**Conference Name:** 11th U.S. National Combustion Meeting
Date Received: 17-Mar-2019 Conference Date: 24-Mar-2019 Date Published: 24-Mar-2019
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**Authors:** Dustin Witkowski, Joshua Herzog, David A. Rothamer
Acknowledged Federal Support: **Y**

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Date Received: 17-Mar-2019 Conference Date: 20-May-2018 Date Published: 20-May-2018
Conference Location: Minneapolis, MN
**Paper Title:** Scattering Referenced Aerosol Phosphor Thermometry
**Authors:** Dustin Witkowski, David A. Rothamer
Acknowledged Federal Support: **Y**

# Novel Approaches to Improve the Sensitivity of Aerosol Phosphor Thermometry Approaches

Final Report Submitted to:

US Army Research Office under contract number W911NF-18-1-0147

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Organization:

Regents of Univ of Wisconsin
Madison, 53715-1218, USA

Principal Investigator:
Dr. David A. Rothamer

## Summary:

Aerosol phosphor thermometry (APT) is a thermographic phosphor particle-based technique to perform gas temperature measurements. Upon excitation with a laser source, the phosphor particles emit temperature-dependent radiation that is imaged with cameras to provide two-dimensional temperature measurements. The elastic laser scattering from phosphor particles can be used for particle image velocimetry (PIV), allowing for minimally-intrusive simultaneous temperature and velocity field measurements (APT-PIV). Unfortunately, precise APT has previously been limited to below 800 K due to limitations in the most commonly used APT approach, the spectral luminescence intensity ratio (LIR) method. The three major goals for the work stated in the proposal were:

1. Determine the feasibility and implementation of the co-doped ratio method to be used to obtain improved fractional sensitivity for aerosol phosphor thermometry (APT).
2. Determine the feasibility of the Mie ratio method to provide improved accuracy for APT measurements.
3. Demonstrate improvements using the two methods in a heated air jet.

As indicated in the goals/objectives for this work, the project investigated two new strategies, the co-doped-ratio method and the Mie-ratio method, to improve the precision of APT at high temperatures by exploiting the temperature-sensitive signal levels characteristic of thermographic phosphors upon thermal quenching.

The first technique investigated was the co-doped-ratio method termed co-doped APT. The method utilizes a host material doped with two rare-earth ions. The ratio of emission from each ion is measured and calibrated versus temperature. To maximize the temperature sensitivity, ions are selected such that the signal for one of the ions is constant or increasing in the temperature range of interest while the other ion is selected to exhibit strongly decreasing signal with increasing temperature due to thermal quenching. This strategy was investigated for trivalent cerium ($Ce^{3+}$) and praseodymium ($Pr^{3+}$) co-doped into yttrium aluminum garnet (Ce,Pr:YAG). Temperature imaging was performed in a heated jet, and single-shot precision comparable to the state of the art for APT measurements was demonstrated from 300 – 450 K. Additionally, it was shown that high-precision temperature measurements could be performed in adjustable temperature ranges by doping $Ce^{3+}$ and $Pr^{3+}$ into different host materials. These results meet the goal set forth in goal 1 from the proposal and have been reported in an archival journal publication [1].

The second technique, the Mie-ratio method, referred to here as scattering-referenced aerosol phosphor thermometry (SRAPT), uses the ratio of a temperature-insensitive reference signal from elastic laser scattering and the temperature-dependent luminescence intensity from phosphor particles to determine temperature. Similar to the co-doped method, the temperature range where precise measurements are achievable can be changed by utilizing different phosphor materials. For the SRAPT technique, choosing an appropriate phosphor for a specific temperature range is simpler than for the co-doping method as only the quenching characteristics of a single dopant ion need to be considered. Demonstration of the SRAPT technique was performed for Ce:LuAG in a seeded air jet at elevated temperatures. Single-shot temperature precisions better than 2% (<15 K) were achieved with seeding densities less than 150 $mm^{-3}$ for temperatures of 750 K and 820 K, providing a significant improvement to the current state of the art for APT measurements at these temperatures. These results meet the objective set forth in goal 2 and have been reported in a second archival journal publication [2].

For both the co-doped APT method and SRAPT the performance of the methods were demonstrated with imaging measurements in a seeded heated jet. The results of these heated jet measurements demonstrated the performance comparable to the state of the art for the co-doped method at low temperatures and for SRAPT demonstrated performance exceeding the state of the art in terms of precision at high temperatures, exceeding 800 K. The heated jet imaging results for the two techniques meet the objective set forth in goal 3 of the proposal.

After meeting the 3 proposed goals, preliminary work was performed on integrating the two measurement methods to extend the temperature measurement range for APT. Both co-doped APT and SRAPT strategies are typically limited to a measurement range of ~300-400 K for precise measurements before the signal levels becomes too weak due to thermal quenching. To overcome this, simultaneous application of the co-doped and SRAPT methods was investigated using Ce,Pr:LuAG. For the combined co-doped/SRAPT technique, single-shot precision better than 20 K was estimated over a range from 400 – 1000 K with a seeding density of 300 $mm^{-3}$. Temperature imaging in an air jet heated to temperatures of 600 K and 800 K demonstrated single-shot precision better than 15 K at these conditions for seeding densities below 100 $mm^{-3}$.

The nine month project resulted in five conference presentations, two conference papers [3, 4], two journal articles [1, 5], and has extended the capabilities of simultaneous APT-PIV to a temperature range applicable for studying turbulence-chemistry interactions in igniting flows. Future work is focused on further characterizing these newly developed techniques and applying them to study low temperature ignition phenomena at compression-ignition engine relevant conditions.

## *1. Introduction*

The coupling between turbulent mixing and thermal energy transfer significantly impacts the behavior of many important processes, such as turbulence-chemistry interactions [6, 7] and low-temperature ignition behavior of different fuels [8, 9]. Often these processes occur in environments inaccessible to conventional measurement techniques. Therefore, the development of minimally intrusive simultaneous temperature and velocity diagnostics are required to gain deeper insight into these processes. Unfortunately, the development of high-precision spatially-resolved temperature diagnostics suitable for harsh combusting environments has proven elusive. The goal of this project was to evaluate two new concepts to improve the temperature-sensitivity and measurement precision of a spatially-resolved single-shot gas temperature measurement strategy referred to as aerosol phosphor thermometry (APT).

APT utilizes temperature-dependent emission properties of thermographic phosphors, which are often composed of rare-earth ions doped into crystalline host structures and ground or formed into particles (~0.1 to 10 μm in diameter). In practice, they are seeded into the flow under investigation, and upon excitation by a laser, the rare earth ion dopants emit fast temperature dependent $4f^{N-1}5d \rightarrow 4f^{N}$ emission that is usually insensitive to pressure and chemical environment. The benefits of APT compared to other diagnostics are readily apparent; upon seeding appropriate levels of phosphor particles into the flow of interest, minimally intrusive simultaneous planar measurements of velocity and temperature fields in high-speed flows are achievable [10, 11].

Reliable APT measurements had previously been limited to below 800 K due to inherent limitations in the most commonly used approach, the spectral luminescence intensity ratio (LIR) method. The spectral LIR method utilizes temperature dependence of the emission spectra of thermographic phosphors. Emission is collected from two different wavelength bands ($S_1$ and $S_2$), and a temperature-dependent intensity ratio ($R = S_2/S_1$) is formed to determine temperature via a calibration. The temperature precision index for the technique ($s_T$) is equal to the product of the fractional ratio precision index ($s_R/R$) and the fractional temperature sensitivity ($\xi_T$), i.e.,

$$s_T = \frac{\partial T}{\partial R} s_R = \frac{(s_R/R)}{\xi_T} \tag{1}$$

where

$$\xi_T = \frac{1}{R}\left(\frac{\partial R}{\partial T}\right). \tag{2}$$

Precise temperature measurements with the spectral LIR method are limited by decreasing signal intensity due to increasing nonradiative transition rates at elevated temperatures (thermal quenching) and limited fractional temperature sensitivity ($\xi_T$) that typically is decreasing with increasing temperature [12]. In contrast to the emission spectra of thermographic phosphors, the absolute signal level is highly temperature-sensitive when thermal quenching is occurring, as shown in Figure 1, but has been difficult to utilize for flow thermometry due to spatial and temporal variations in particle seeding density and laser energy. The two new strategies explored in this project were aimed at exploiting this absolute signal temperature sensitivity while minimizing dependencies on particle seeding density and laser energy. This report discusses each new strategy, as well as the key outcomes and accomplishments of this project. Finally, some conclusions will be drawn and the future direction of this research will be briefly discussed.

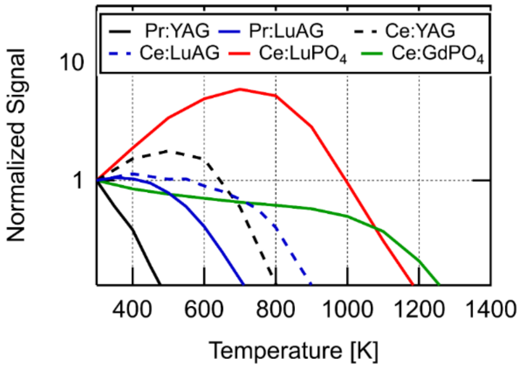

Figure 1. Luminescence intensity vs temperature for six thermographic phosphors measured in a tube furnace.

## 2. *Co-Doped APT*

The first technique, termed co-doped APT is performed by co-doping a host material with two rare-earth ions. The ratio of emission from each ion ($R_{co-doped} = S_{ion,2}/S_{ion,1}$) is measured and calibrated versus temperature. Ions are selected such that the signal for one of the ions exhibits minimal temperature dependence, while the other is selected to exhibit strong signal temperature dependence over the range of interest, therefore maximizing the fractional temperature sensitivity of the technique. The ions chosen for the co-doped APT method are $Ce^{3+}$ and $Pr^{3+}$ for several reasons. First, their quenching temperatures are often significantly different, as shown in Figure 1 for $Ce^{3+}$ and $Pr^{3+}$ in yttrium aluminum garnet (YAG) and lutetium aluminum garnet (LuAG). This is due to the fact that they often quench through different mechanisms [13]. Second, each ion's 5d levels are often accessible with conventional pulsed laser sources, and many times they can be excited with the same source. Finally, for a given host $Ce^{3+}$ and $Pr^{3+}$ will emit in distinct bands separated by ~12000 $cm^{-1}$ [14], making signal collection straightforward.

As a proof of concept of the co-doped APT strategy, trivalent cerium ($Ce^{3+}$) and praseodymium ($Pr^{3+}$) co-doped into YAG (Ce,Pr:YAG) was chosen for an imaging demonstration in a heated jet. High temperature-sensitivity and single-shot precision comparable to the state of the art for APT measurements [15] was demonstrated from 300 – 450 K. Figure 2 displays selected single-shot images and Figure 3 shows the single-shot precision results. The results of this work were recently presented at the 37th International Symposium on Combustion in Dublin, Ireland in August of 2018, followed by publication in the biennial Proceedings of the Combustion Institute (PROCI) journal [1].

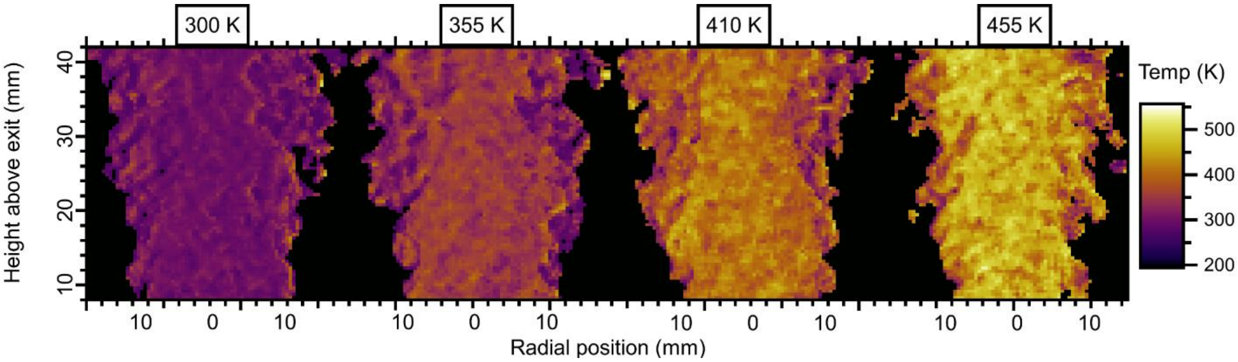

Figure 2. Single-shot temperature fields for different jet exit temperatures using co-doped APT.

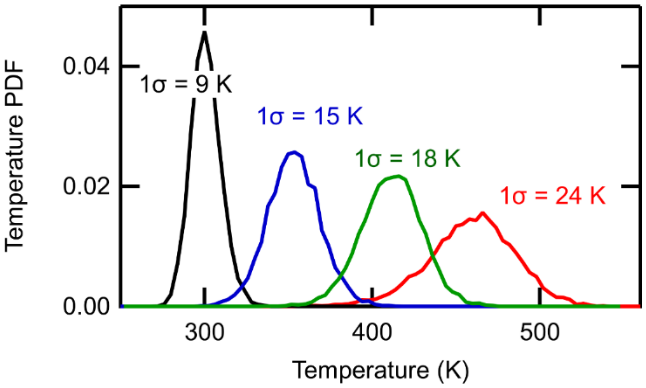

Figure 3. PDFs of the single-shot temperature measurements near the jet exit for each jet temperature.

One of the most promising aspects of the co-doped APT technique is that phosphors for specific temperature ranges can be made by merely changing the host material. For example, the YAG host was able to provide precise measurements in the temperature range from 300–500 K. By replacing the YAG host with lutetium aluminum garnet (LuAG), precise measurements from 400 K – 700 K are achievable (see Figure 1 and section 4). Finally, a recently developed search methodology [13] to identify high quenching temperature host materials has resulted in the identification of the orthophosphate host family ($REPO_4$, RE=Gd, Lu, Y), and specifically a $Ce:GdPO_4$ phosphor with a quenching temperature of 1000 K and 10% of its room temperature signal remaining at 1300 K (see Figure 1). Previous analysis has estimated that $Pr:GdPO_4$ may quench at even higher temperatures than reported for $Ce:GdPO_4$ [13]. Therefore, there is evidence that both $Ce^{3+}$ and $Pr^{3+}$

will maintain high signal levels at temperatures above 1000 K when doped in orthophosphate hosts, making them promising candidates to further extend the high temperature limit of co-doped APT.

## *3. Scattering-Referenced APT*

The second technique, referred to as scattering-referenced aerosol phosphor thermometry (SRAPT), uses the ratio of a temperature-insensitive reference signal from elastic laser scattering and the temperature-dependent luminescence intensity ($R_{SRAPT} = S_{sca}/S_{ion}$) from phosphor particles to determine temperature. Similar to the co-doped method, the temperature range where the sensitivity peaks can be changed by utilizing different phosphor materials. For the SRAPT technique, choosing an appropriate phosphor for a specific temperature range is simpler than for the co-doping method as only the quenching characteristics of a single dopant ion need to be considered. However, differing dependencies of the scattering and luminescence signals on particle size and laser fluence can lead to measurement bias and decreased temperature precision. To evaluate the feasibility of SRAPT, experimental measurements were performed for Ce:LuAG in a seeded air jet at elevated temperatures. Single-shot temperature precisions better than 2% (<15 K) were achieved with seeding densities less than 150 mm$^{-3}$ for temperatures of 750 K and 820 K. Figure 4 shows selected single-shot measurements and Figure 5 displays the single-shot precision of this technique as a function of seeding density.

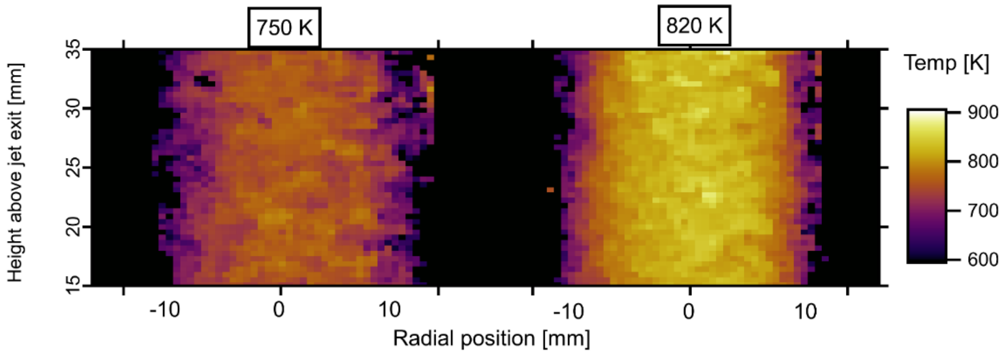

Figure 4. Single-shot temperature fields for different jet exit temperatures using the SRAPT method.

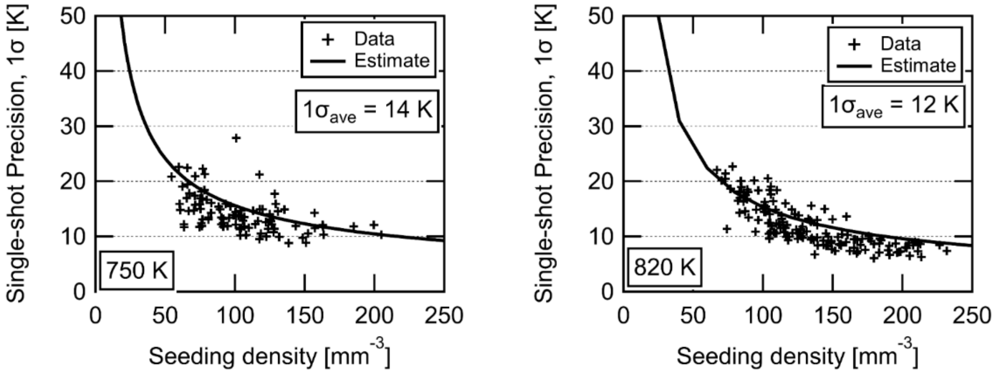

Figure 5. Single-shot precision vs. seeding density for jet temperatures of (Left) 750 K and (Right) 820 K using SRAPT.

Results from work on the SRAPT technique were disseminated in a conference paper and presentation at the 2018 Spring Technical Meeting for the Central States Section of the Combustion Institute (Minneapolis, MN, May 20-22, 2018) [3] and in a presentation at the Inaugural International Conference on Phosphor Thermometry (ICPT - Glasgow, UK, July 25-27, 2018) [16]. Following the ICPT conference, we were invited to submit a journal article for a special issue of Measurement and Science Technology, which was recently accepted for publication [2]. The principal investigator also presented results from the co-doped APT and SRAPT work in an invited presentation at the AIAA 2018 AVIATION Forum (Atlanta, GA, June 25-29, 2018)

### *4. Combined Co-doped/SRAPT Technique*

Recent work has focused on improving the temperature range that precise measurements can be performed with these techniques. Since both co-doped APT and SRAPT rely on temperature quenching of the phosphor signal, each method is typically limited to a range of ~300-400 K before the signal level becomes too weak [1, 2]. To alleviate this restriction, simultaneous application of the co-doped and SRAPT methods has recently been demonstrated to provide a measurement range from 400 K to 1000 K using $Ce^{3+}$ and $Pr^{3+}$ co-doped into LuAG (Ce,Pr:LuAG). For this technique three cameras are utilized to simultaneously measure two intensity ratios for each data set; a co-doped ratio using the $Ce^{3+}$ and $Pr^{3+}$ emission ($R_{co-doped} = S_{Ce}/S_{Pr}$) and a SRAPT ratio using the elastic scattering and $Ce^{3+}$ emission ($R_{SRAPT} = S_{sca}/S_{Ce}$). The scattering signal acquired with the third camera can also be used for simultaneous velocity field measurements using particle image velocimetry (PIV). Therefore, the setup provides simultaneous temperature and velocity imaging using three cameras.

The single-shot precision for the combined co-doped/SRAPT technique was estimated based on measurements of the ratio precision for each technique in a seeded jet and a furnace calibration to translate each ratio to temperature. The results are displayed below in Table 1. Single-shot precision better than 20 K is estimated over a range from 400 – 1000 K with a seeding density of 300 $mm^{-3}$ for Ce,Pr:LuAG. To validate these estimates, an imaging demonstration was performed in an air jet heated to temperatures of 600 K and 800 K. Selected single-shot images are provided in Figure 6. Figure 7 shows that single-shot precision better than 15 K was demonstrated at these conditions for seeding densities below 100 $mm^{-3}$.

These results have recently been accepted for presentation at the 11th U.S. National Combustion Meeting (Pasadena, California, March 24-27, 2019) and the associated conference paper has been submitted [4]. In addition, a plethora of valuable data has been acquired from these most recent measurements that still needs to be analyzed. With additional time and funding the final anticipated result of this work is multiple journal articles, including an in-depth characterization of the co-doped APT method for Ce,Pr:LuAG, demonstration and characterization of the combined co-doped/SRAPT technique for precise measurements over a wide range of temperatures, and a direct comparison of the co-doped and SRAPT techniques in terms of measurement precision, experimental biases, and high-precision temperature range.

Table 1: Estimated temperature precision for the combined co-doped/SRAPT techniques at different temperatures. Specified object pixel size of 0.19 $mm^3$, laser fluence of 60 $mJ/cm^2$ and seeding density 300 $mm^{-3}$.

| Temperature [K] | APT Method | Ce Intensity [counts] | Pr Intensity [counts] | $\frac{s_R}{R}$ | $\frac{1}{R}\left(\frac{\partial R}{\partial T}\right)$ [%/K] | Temperature Precision [K] |
|---|---|---|---|---|---|---|
| 400 | Co-doped | 37682 | 27243 | 0.04 | 0.24 | 18 |
| 500 | Co-doped | 35868 | 17635 | 0.05 | 0.46 | 11 |
| 600 | Co-doped | 30615 | 7245 | 0.07 | 0.87 | 8 |
| 700 | Co-doped<br>SRAPT | 23292 | 2556 | 0.11<br>0.05 | 0.62<br>0.43 | 18<br>12 |
| 800 | SRAPT | 14344 | -- | 0.06 | 0.76 | 8 |
| 900 | SRAPT | 6736 | -- | 0.08 | 1.0 | 8 |
| 1000 | SRAPT | 2729 | -- | 0.11 | 0.59 | 19 |

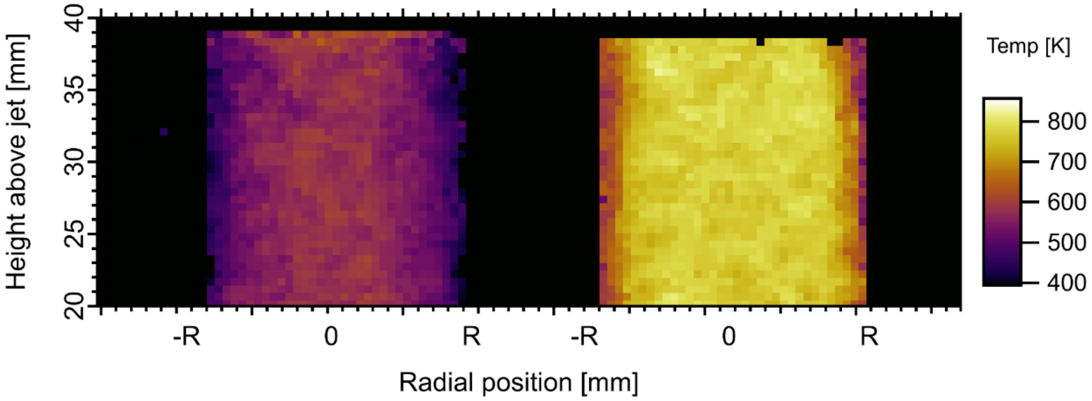

Figure 6. Single-shot temperature fields for different jet exit temperatures using the combined co-doped/SRAPT method.

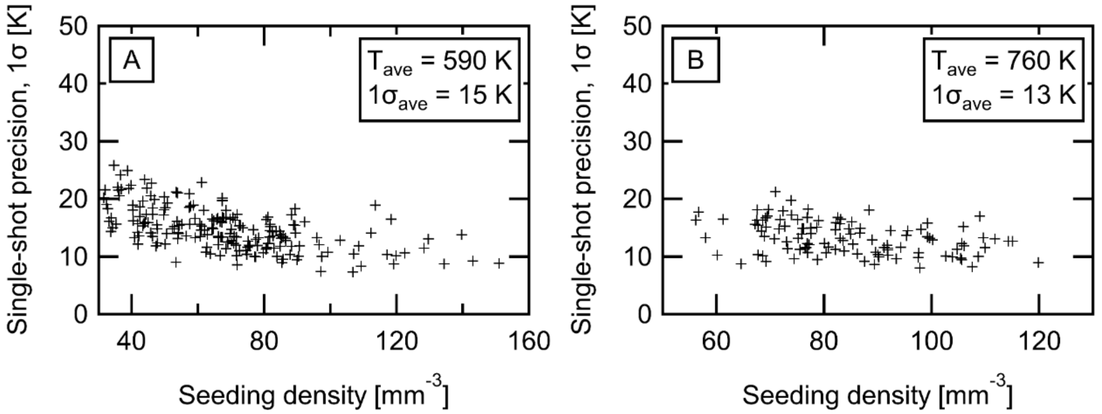

Figure 7. Single-shot precision as a function of seeding density for APT measured jet exit temperatures of (Left) 590 K and (Right) 760 K using the co-doped/SRAPT method.

## 5. *Conclusions*

The goal of this project was to evaluate two new concepts aimed at improving the measurement precision of aerosol phosphor thermometry by utilizing the high temperature sensitivity of thermographic phosphor signal levels. For all phosphors tested, both the co-doped and SRAPT techniques demonstrated measurement precision on the order of 10-20 K with moderate seeding densities applicable for simultaneous particle image velocimetry (PIV) measurements. Additionally, whereas precise measurements with APT have previously been restricted to well below 800 K due to the combination of low signal levels and poor temperature sensitivity of the commonly used spectral LIR approach, this work has demonstrated measurement precision better than 15 K at temperatures exceeding 800 K with seeding densities as low as 100 mm$^{-3}$. Finally by combining the co-doped/SRAPT techniques, measurement precision better than 20 K was estimated over a wide range of temperatures, from 400 K – 1000 K with the phosphor Ce,Pr:LuAG. These estimates were partially validated with imaging demonstrations at 600 K and 800 K. The results of this work extend the capabilities of simultaneous APT-PIV to a temperature range applicable for studying turbulence-chemistry interactions in igniting flows.

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