11TH INTERNATIONAL SYMPOSIUM ON FLOW VISUALIZATION
August 9-12, 2004, University of Notre Dame, Notre Dame, Indiana, USA

ON THE USE OF PHOSPHORS FOR FLOW MEASUREMENTS

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ABSTRACT

Phosphor based thermometry usually involves applying a phosphor coating to a surface of interest and determining temperature from the phosphor's fluorescence characteristics. This method is well established and th[i]e applications for it are increasing. An area of application that is less well explored involves injecting phosphor particles themselves into a liquid or gas phase fluid flow for diagnostic purposes. We will present some of our results for flow temperature and transport measurement. Most recently this concerns infusion measurements in a brain phantom. Key issues which related to particle size and excitation source will be discussed. We will also discuss the potential for transient measurements for gas phase flows. The availability and the growing properties database of nano-sized phosphor materials like those used in our study should enable a number of new flow visualization applications. In addition, the availability of high brightness light emitting diodes and arrays of them that emit not only in the visible but in the near ultraviolet should also have positive implications for this field.

1 INTRODUCTION

Phosphor measurement techniques have addressed a variety of noncontact applications such as in turbine engines, combustion engines, steel manufacturing, and electric motors (1,2). Contact applications have also proliferated for which phosphor is applied to the end of a fiber or for which the fiber itself is fluorescent(3). The accumulated knowledge and experience base in conjunction with advances in fiberoptics, lasers, electronic signal processing, and personal computing continue to improve the utility of the approach. At the same time, the state of the art in using light scattering particles for laser doppler velocimetry (LDV) and related flow diagnostic parameters has advanced in a similar way with the aid of the same technologies and, in addition, computational fluid dynamics. It would seem that there is much potential in merging phosphor and particle flow diagnostics. Described here are some aspects of applying the materials and methods of phosphor thermometry to flow measurements.

2 PREVIOUS WORK

Nakatoni et al (4-6) introduced phosphor particles as flow tracers in liquids as reported at the 2nd International Flow Visualization Symposium. They used various combinations of ZnS, CdS, and CaS materials. The long decay times (~ 1.5 s) were well suited for visualizing relatively slow moving fluids. Streak photographs tracked the seeded fluids, yielding distance versus time relationships that made it possible to resolve flow velocity and direction. No efforts to measure temperature were described.

In the 1990's, as described in his dissertation and a subsequent publication at ORNL, Miller seeded La_2O_2S:Eu phosphor into a fluid for temperature measurement (7,8). For this, a thin film of liquid lithium bromide (LiBr) flowed on the inner surface of a transparent tube as part of a program related to heat pump development. Temperature in the film ranged from 35 to 50 C with a resolution of 1.5 C. The results showed that a specific approximation used in such studies is valid and can be scaled with absorber tube length and diameter.

The most extensive investigation in this area to date to the author's knowledge is reported by Guerrier et. al. (9). They demonstrated a method for visualizing mixing of two different flow streams within an internal combustion engine cylinder by injecting different phosphors into the respective flow paths. They illuminated YVO_4:Eu (emission 620 nm) and YVO_4:Dy (emission 575 nm) with a nitrogen laser at 337 nm. They used a scientific grade gated intensified charge-coupled device camera to successfully image separate flow streams. They indicate it is feasible to infer direction and bulk flow velocity by the smearing during 5 ms gated detection periods. They did not seek to exploit the decay temperature dependence, though with such equipment it would have been possible. It may be noted here that, with the decay time aproach, this Dy phosphor is an excellent temperature indicator between 300 and 450 C (1). The Eu phosphor temperature range is from about 450 to 800 C (1).

3 FEATURES OF PHOSPHOR FLOW TRACERS

The following is a list of features and advantages of phosphor particles used as flow tracers. Phosphor particles

1. May yield temperature information as well as function in the usual way as a light scatterers for LDV.
2. Are commonly of ceramic material and therefore quite robust in surviving and functioning in harsh, high temperature environments.
3. May be encapsulated. It may be possible to control density and thus buoyancy for liquid applications.
4. Can be adjusted in size during the manufacturing process.
5. May indicate flow patterns by the streak method. The luminescence will have a characteristic duration ranging from nanoseconds to hours, depending on the particular material. The speed of the flow will dictate phosphor choice.
6. Exhibit an emission wavelength or spectral distribution that is distinct from the illumination source.
7. Unless encapsulated, may have irregular shape. In an inviscid fluid, particle affinity may lead to agglomeration. Also, some phosphors tend to agglomerate and collect on flow channel walls. This was observed by Miller (7,8). It may perhaps be mitigated with encapsulation.

3 OBSERVATIONS OF MICROPARTICLES

Described here is a very simple scoping experiment designed to elicit the signal levels and other features of phosphor particle aerodynamic measurement. The experiment was conducted in a laboratory hood. It was noticed that simply by removing the lid to a phosphor bottle and placing it in the hood, that the air flow would entrain phosphor particles in an upward draft. Figure 1 shows the arrangement. A Laser Science nitrogen laser emitting about 300 microjoules provided the excitation.

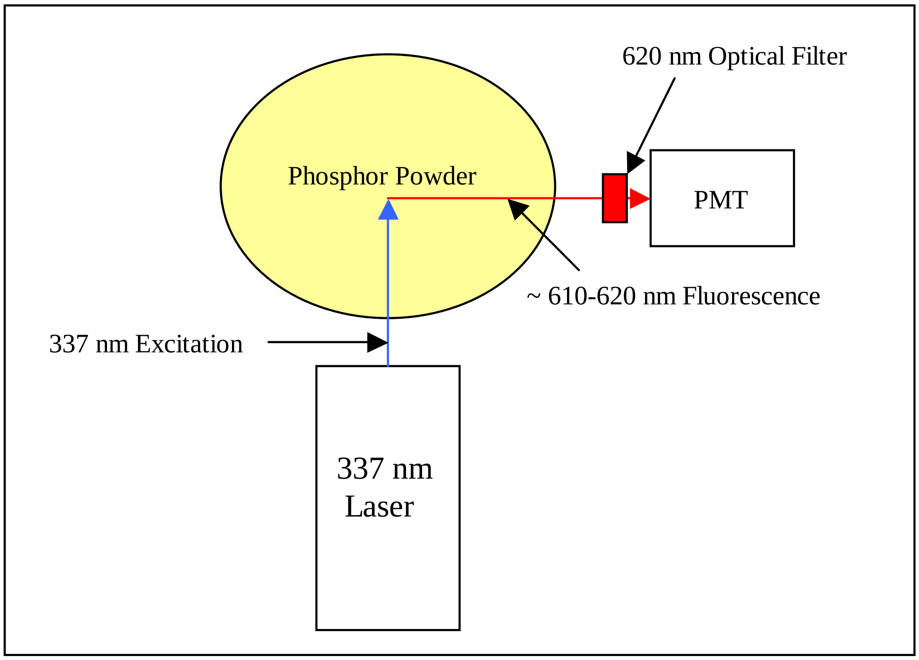

Figure 1. Arrangement for phosphor powder flow measurement.

Figure 2 is a photo using a consumer digital camera of phosphor powder (La_2O_2S:Eu) above the bottle. Simply picking up the bottle and setting it down, then unscrewing the lid, is sufficient agitation to produce the effect. The larger particles that are captured in the photograph do not appear to be entrained in the flow. However, smaller particles are visible to a human observer (in the dark and with laser excitation) and are moving vertically toward the flue of the hood. The distance from the photomultiplier tube (PMT) detector and the center of the powder plume was about 15 cm. The volume element illuminated by the laser and viewed by the detector was located about 7 cm above the bottle opening. Figure 3 is a photograph of fluorescence produced by laser excited particles. The laser path is indicated. The focus is not optimum and the size of the red luminescence images makes the particles appear larger than they really are. A large amplitude signal, typically on the order of a volt, is readily produced from this highly temperature sensitive material.

An additional effort was aimed at quantifying signal levels from a single phosphor particle. Figure 4 shows two particles as sample 1 and 2 respectively. In all likelihood, these entities are probably agglomerations comprised of smaller particles. The sample objects are depicted in relation to the millimeter markings of a plastic ruler underneath for size estimation. The insets show single particle

Figure 2. Agitated phosphor container shows powder in air.

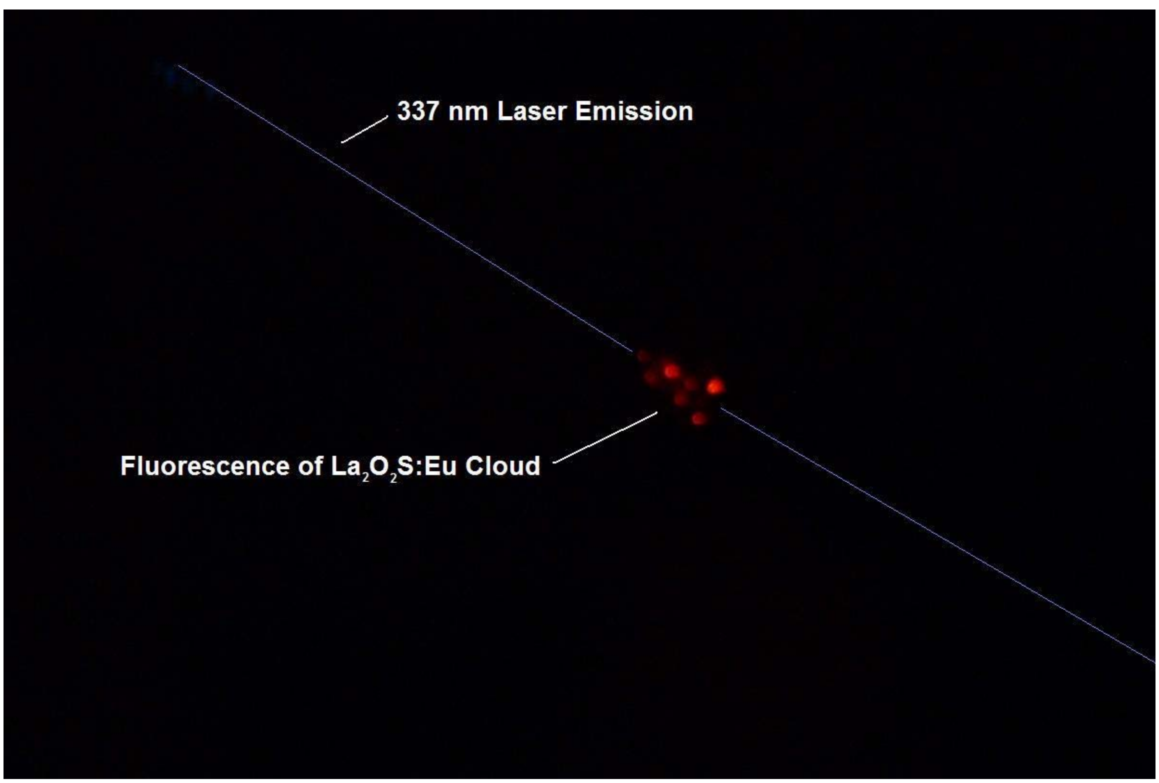

Figure 3. Image of fluorescence.

emission recorded with a digital camera in conjunction with a moderate resolution laboratory microscope. Figure 5 shows the detected signal for both the air flow case above and for the single particles. The collection solid angle was defined by a 1 cm active area of the photomultiplier tube which was located about 3 cm from the particle. The exciting laser was not focused. Signal levels are quite high, in fact the signal for the 500 micron particle appears to be saturating the detector slightly. Clearly use of high quality collection optics and focusing excitation light will lead to the ability to detect ever smaller particles, enabling microscale thermometry and flow measurement. Or, it may indicate lower power light sources such as from light emitting diodes are viable. The measurements described here provide a beginning of a quantitative basis for applying luminescent particles to measuring flow phenomena.

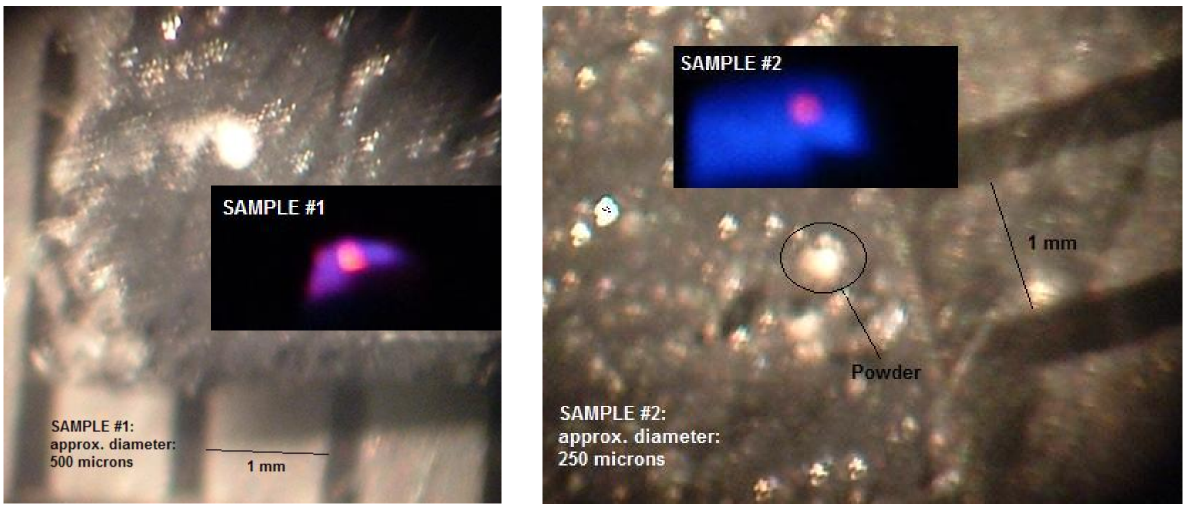

Figure 4. Single particle images.

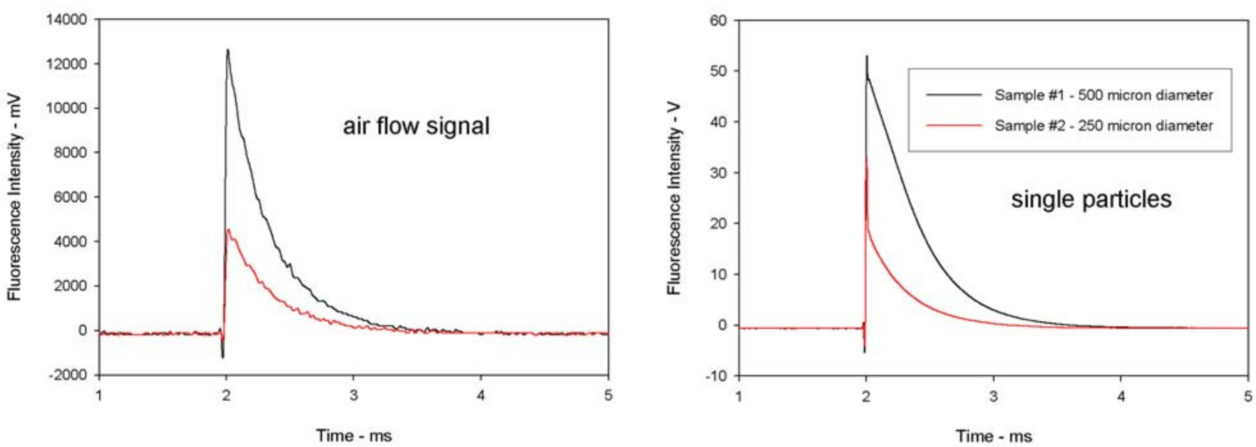

Figure 5. Detected signal from phosphor powder in air flow and isolated single particles.

4 NANOSCALE PARTICLE APPLICATIONS

We have a history of using phosphor particles for a variety of temperature and other sensing applications for customers both private and governmental. Phosphor particle sizes are typically greater than 1 micron in diameter. Recently, some researchers have begun studying common phosphor compounds whose size is on the order of nanometers. They are termed variously as nanophosphors or nanocrystals. As is well known, spectral properties change in several ways, eg. Excitation bands shift and emission distributions change as particles become smaller (10).

Whereas the impetus for much of this research relates to display and photonic applications, our interests are in these materials for sensing purposes. The size of these materials should make possible new uses.

4.1 Biological infusion as nanoscale tracers.

Positive pressure infusion of therapeutic agents directly into the bulk brain tissues holds promise of providing a new therapeutic modality for addressing this disease. The infusion process circumvents the screening effects of the "blood-brain barrier" and provides a pathway for delivery of therapeutic agents through the interstitial space. It can thus be used to target and reach the infiltrating tumor cells (1). But there is a need to be able to model and study the infusion process for various agents. Phosphor particles less than about 20 nm are sufficiently small to be transported through the blood brain barrier and may function as flow tracers. Steps have already been taken toward this end by Gillies et al. (11), who infused a slurry of Y_2O_3:Eu into a 0.6% agarose gel. This material is used routinely as a brain phantom material in tests of protocols for positive pressure infusion therapy within the brain. In such a therapy, the neurosurgeon seeks to deliver a chemotherapeutic agent through the interstitial space in the brain by pumping it from a catheter at flow rates of approximately 1 to 5 microliters per minute. The agent is thus driven through the porous structure of the extracellular matrix, the mean pore size of which is on the order of 20 nm in healthy tissue, but which distends considerably in the presence of pathologies and/or edema. In attempts to optimize protocols for infusion therapies, an inexpensive yet robust medium was sought to serve as a surrogate for *in vivo* brain tissues, and after extensive studies, agarose gel at a concentration of 0.6% was found to work. To investigate the nature of the correspondence between the sub-microscale properties of the gel and the brain which allow the gel serve this function, particles of the Y_2O_3:Eu roughly 8 to 12 nm in diameter were infused into a 0.6% gel and tracked via their fluorescence, to confirm that they could traverse the interstitial space within the gel matrix and thus establish a lower limit on pore size in the matrix. Figure 6 shows the results of several infusions of the nanoparticles (which were in suspension in deionized water), and reveals that they were indeed able to flow through the gel.

4.2 Micro and nanoscale thermometry.

The recent explosion of interest in nanotechnology brings with it the need for measurement tools suited to that scale. In a separate series of experiments (12) related to the above infusion investigation, the fluorescence-based temperature dependence of YAG:Ce nanoparticles was measured within the physiological range. Part of the motivation was to eventually infuse such particles as well, in further studies of convection-enhanced delivery of therapeutic agents within the gel surrogate. For instance, measurement of thermal conductivity and transport across nanotubes and other structures could be addressed with nanophosphors.

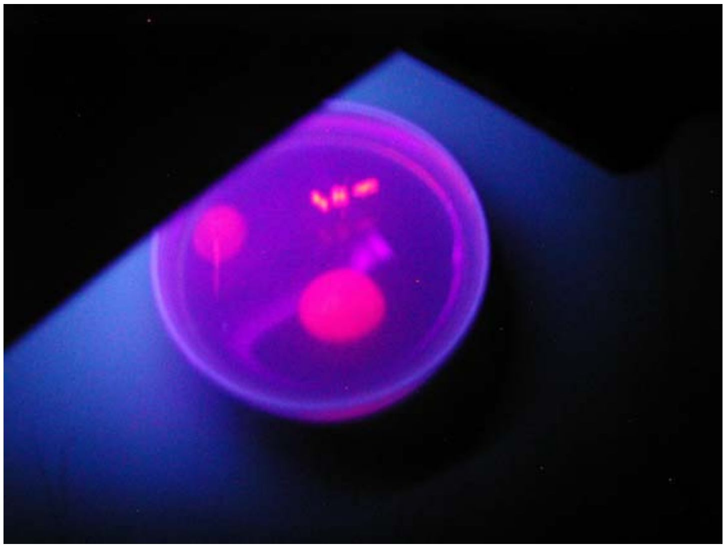

Figure 6. Luminescence of nanoparticles injected into brain phantom gel.

4.3 Physical parameter sensing.

Because there is a larger surface-to-volume ratio as particle-size decreases, phenomena which affect the spectral response of the surface molecules will be detectable. For instance, it is known that when placed in suspension, the decay time of particles will change as a function of pH (13). There is no discernible change for larger sized particles (>8 nm). This suggests that there may be a response to such parameters as oxygen concentration. If true, this would make possible pressure measurements. In recent work Vetrone gives spectral characteristics for a number of rare-earth doped luminescent materials and notes that for nanoparticles in a liquid, the emission characteristics change with the liquid's index of refraction.(14).

5 TRIBOLUMINESCENCE

The above results and observations point the way to several approaches utilizing particulates in a liquid or gaseous stream as temperature and flow path indicators. In addition, it may be noted in closing that another characteristic that may be exploited is triboluminescence. Certain phosphor materials, most notably ZnS:Mn, show a pronounced luminescence when struck or stressed (15). With as little as 0.1 J energy striking a coating of this material, triboluminescence may be observed. It would seem that particulates in a high speed flow which strike such a coating should also triboluminesce. Several approaches to coating health monitoring and surface impact degradation are therefore possible.

6 ACKNOWLEDGEMENT

The work described here was sponsored by the U. S. Department of Energy under contract DE-AC05-00OR22725 with Oak Ridge National Laboratory, managed and operated by UT-Battelle, LLC.

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