

Combining photo-chemistry, designer molecules, lasers, and digital imaging creates a powerful technique for the quantitative mapping of the velocity field in moving fluids.

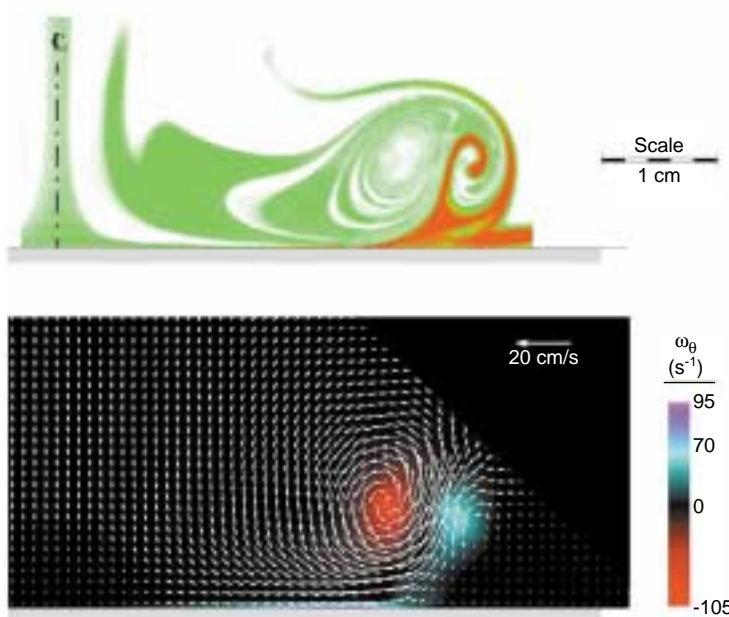
Molecular tagging velocimetry maps fluid flows

Manoochehr M. Koochesfahani and Daniel G. Nocera

The behavior of fluid flows influences, and often determines, the performance of a large variety of devices and processes. Some of the more familiar examples include fuel/air mixing in internal combustion (IC) engines and combustors, aerodynamics of flow around objects moving in air and water, liquid and gas delivery systems, and manufacturing processes such as solidification. There is also renewed interest in fluid flows in biological systems, and in microflow geometries in connection with the design of chemical and biological detection and processing systems.

To understand, and control, the behavior of fluid flows one often needs flow velocity information at many points simultaneously—a quantitative whole-field map of the velocity field. This task is now commonly performed with a technique called particle image velocimetry (PIV). The fluid medium is seeded with many tiny particles that are then illuminated by a sheet of laser light. As the particles move through the laser light they scatter it and their positions are recorded on photographic film or charge-coupled-device (CCD) detectors. The positions are recorded at two different times, separated by a prescribed time interval, and the displacement of individual particles, or more often groups of particles, are determined by well-developed computer-intensive procedures. These displacements over the known time interval provide the estimate of the particle velocity, which gives an accurate estimate of the fluid velocity under certain conditions that are well-understood and often taken into account by the practitioners of PIV.

MANOOCHHR M. KOOCHESFAHANI is a professor in the Department of Mechanical Engineering at Michigan State University in East Lansing, MI. DANIEL G. NOCERA is a professor in the Department of Chemistry at the Massachusetts Institute of Technology in Cambridge, MA. The authors are researchers in the Center for the Sensor Materials (CSM) at Michigan State University, East Lansing, MI; e-mail koochesf@egr.msu.edu and nocera@mit.edu.



Mapping of flow field during vortex-wall interaction shows a vortex ring approaching a wall that causes boundary layer separation and the formation of a secondary counter-rotating vortex ring. Only the right half of the flow is shown just after the formation of the secondary vortex. Broken line indicates the axis of symmetry. Laser induced fluorescence visualization shows the vortex ring and wall-layer fluids marked by green- and red-emitting laser dyes (top). Velocity vectors and vorticity color contour map measured by MTV (bottom).

There are flows, however, in which the presence of seed particles is undesirable—they may interfere with the operation of a device or the flow process itself. There are also situations in which the inertia and buoyancy of the particles can compromise their ability to track the fluid motion. In these cases molecular tagging approaches can offer significant advantages.

Molecular tagging velocimetry

This method of velocimetry relies on molecules that can be turned into long-lifetime tracers when excited by photons of an appropriate wavelength. Typically a pulsed laser is used to “tag” the regions of interest, and those regions are interrogated

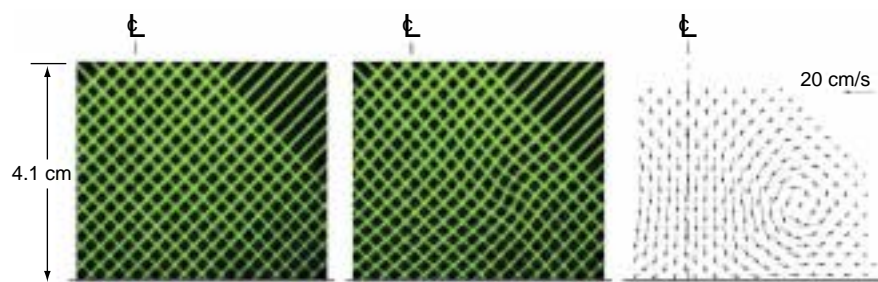


FIGURE 1. Molecular tagging velocimetry image pairs and the resultant velocity vector field show flow from a vortex ring impinging downward on a flat wall at normal incidence. Only the right half is shown and the broken lines indicate the axis of symmetry. Tagged regions imaged 1 μ s after the laser pulse (left). The same grid imaged 8 ms later (center). The derived velocity field using a spatial correlation procedure (right).

at two successive times within the lifetime of the tracer. The measured displacement of the tagged regions provides the estimate of the fluid velocity vector. One might think of the MTV technique as the *molecular* counterpart of PIV. In one implementation of the technique a planar grid of intersecting laser beams, formed from a pulsed excimer laser at 308 nm, is used to turn on the luminescence of water-soluble phosphorescent supramolecules that are premixed in a water flow (see Fig. 1). The flow is that of a vortex ring, generated by discharging a slug of water from a pipe into a fishtank, which impinges vertically downward onto a flat wall.

The tagged molecules form a “glowing net” that convects, bends, and folds as the molecules move with the flow through three-dimensional space. A later image of these tagged regions at a prescribed time delay provides a quantitative map of the fluid velocity field. The conventional imaging shown in Figure 1 provides information on two components of the velocity vector, the projection onto the viewed plane. Stereo imaging with multiple imagers can produce the complete three components of the velocity vector.

From chemistry to fluid flows

A molecular complex is suitable for molecular tagging applications if its lifetime as a tracer is long enough relative to the flow convection time scale to allow sufficient displacement of the tagged regions. While the earliest use of MTV can be traced back at least three decades, this technique has seen significant advances during the past 10 years

as a result of improvements in laser and imaging techniques, data analysis methods, and chemical design and synthesis of novel molecular structures. Liquid-phase flow applications relied, until recently, on photochromic molecules in organic solvents. In a photochromic process absorption by photons, typically UV photons, causes a color change (from clear to dark blue, for example), which can persist for several seconds to minutes. The long-lifetime tracer is the molecule with the new color, and the chemical is reusable since the photochromic process is reversible.

The applications in liquid-phase flows have been expanded considerably by the availability of water-soluble compounds such as caged fluorescent molecules and engineered phosphorescent supramolecules. Among the former, caged fluorescein has been utilized most often. In this compound a chemical group is attached to fluorescein in order to render it nonfluorescent. The caging group is removed upon absorption of UV photons at 350 nm, thereby creating regular fluorescein, which fluoresces with very high quantum efficiency. Here the long-lifetime tracer is the uncaged fluorescein, which persists for a very long time and can be interrogated at the time of interest through its fluorescence. Two sources of photons are therefore needed, one to break the cage and the other to excite fluorescence. In current designs the cage-breaking process is irreversible, so each caged molecule can be used only once.

The work described in Figure 1 takes advantage of phosphorescent supramolecules, a new class of water-soluble

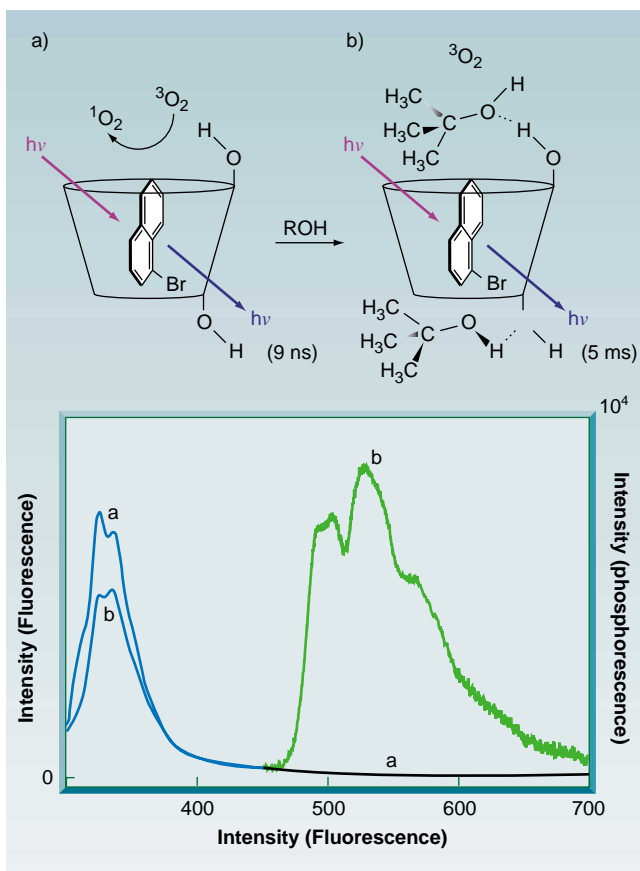


FIGURE 2. Emission from 1-BrNp in Gb-CD. Only blue fluorescence is exhibited in the absence of an appropriate alcohol (ROH) (a). Bright green phosphorescence plus the initial fluorescence is seen upon the addition of ROH, which prevents the quenching of 1-BrNp phosphorescence by oxygen (b).

compounds suitable for molecular tagging diagnostics. When a phosphorescent compound is used for molecular tagging, excitation by photons produces a long-lived excited state that is interrogated through its phosphorescence emission as the molecule radiatively returns to its ground state. The long-lifetime tracer is the excited state molecule itself. Here, only one source of photons is needed, and the excitation/emission process is reversible, which means the chemicals are reusable. The difficulty is that long-lived luminescence suffers from quenching primarily from oxygen, and so suitable molecular complexes have not been available until recently.

A new design prevents the quenching of a lumophore by mixing certain alcohols (indicated collectively by "ROH") with an aqueous solution of a cyclodextrin (CD) that contains the lumophore (see Fig. 2). Cyclodextrin is a molecule

that is constructed from sugars connected in a head-to-tail arrangement forming a cup-shaped cavity, much like a Dixie cup but with the bottom punched out. The outside of the CD cup is totally water soluble, but the inside is hydrophobic. The lumophore is 1-bromonaphthalene (1-BrNp), which absorbs efficiently at 308 nm but its phosphorescence is effectively quenched by oxygen. Because the CD cup inside is hydrophobic, 1-BrNp, which is also hydrophobic, ends up inside any nearby CD cup. The addition of alcohol, typically cyclohexanol, forms a ternary complex, where the alcohol hydro-

gen bonds to the rim of the CD cup and acts as its lid, thereby shielding 1-BrNp from oxygen. The resulting long-lived, green phosphorescence has a typical lifetime of about 5 ms. The design described here acts in a way similar to the design of a glow-in-the-dark frisbee where the glowing phosphorescent material is encased in plastic to protect it from oxygen quenching. In gas-phase applications, the knowledge of photochemistry and advances in lasers have led to several approaches for tagging and interrogation. Oxygen can be tagged by driving it into an excited vibrational state via stimulated Raman scattering, which is then interrogated through its fluorescence. Oxygen can also be tagged by generation of ozone via oxygen photodissociation, followed by interrogation of the tagged regions for ozone dissociation and the subse-

that is constructed from sugars connected in a head-to-tail arrangement forming a cup-shaped cavity, much like a Dixie cup but with the bottom punched out. The outside of the CD cup is totally water soluble, but the inside is hydrophobic. The lumophore is 1-bromonaphthalene (1-BrNp), which absorbs efficiently at 308 nm but its phosphorescence is effectively quenched by oxygen. Because the CD cup inside is hydrophobic, 1-BrNp, which is also hydrophobic, ends up inside any nearby CD cup. The addition of alcohol, typically cyclohexanol, forms a ternary complex, where the alcohol hydro-

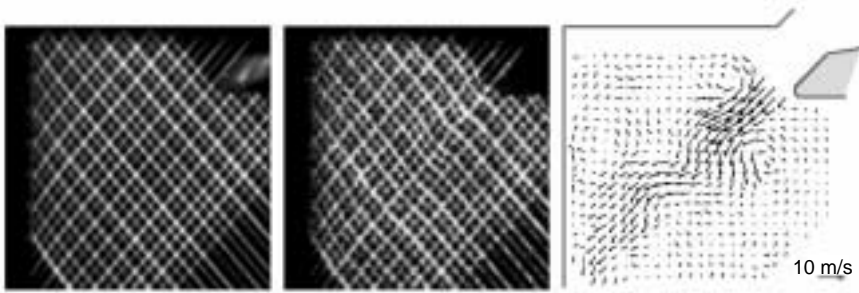


FIGURE 3. Molecular tagging velocimetry image pair using nitrogen seeded with biacetyl and the resultant velocity vector field in a steady flow rig model of IC engine. A 3×3 -cm field of view of the initial tagging pattern right after laser firing. Part of the valve body and the left wall of the cylinder are visible in the picture (left). Tagged regions imaged $50 \mu\text{s}$ later (center). The velocity field derived from images at left and center, showing the details of the intake flow (right).

quent oxygen fluorescence. In yet another method suitable for flows containing water vapor, photon excitation can generate OH by photodissociation of water. The long-lifetime tracer is OH, which is then interrogated through its fluorescence using a second photon source. Use of nitrous oxide tagging, generated by pho-

todissociation, has also been recently reported. Some of the popular fluorescent tracers such as biacetyl and acetone, which have often been used for flow visualization and laser induced fluorescence measurements, also exhibit phosphorescence, which can be used for molecular tagging velocimetry. In this case only one

photon source is needed. However, because phosphorescence of these molecules is effectively quenched by oxygen, their use is limited to oxygen-free environments (typically nitrogen is used).

Applications

Molecular tagging velocimetry is being used in several laboratories around the world to investigate fundamental flow phenomena and applied fluid engineering problems. Detailed measurements are possible with this technique (see figure, p. 103). The method has been used in flows over a wide range of speeds, from liquid-phase flows with speeds in the tens of microns per second to gas-phase flows at supersonic speeds. The scope of the studies covers a range including pulsatile flow in tubes, internal circulation in droplets, unsteady boundary layer separation from aerodynamic sur-

faces, convective flow in directional solidification, flows during intake and compression inside motored IC engines, free and wall-bounded turbulent flows, and highly three-dimensional vortex flows.

One engineering application that is benefiting from these developments is the mapping of flows in automotive IC engines under motored conditions. These studies are being carried out by Harold Schock's group at Michigan State University's Automotive Research Experiment Station (ARES). A steady flow rig configuration is commonly used in the IC engine research community to study the fundamentals of the intake flow (see Fig. 3). In a study sponsored by the US Department of Energy, this technique is now being used to map the high level of cycle-to-cycle variability during the late compression phase of IC engines and determine strategies for its control.

Molecular tagging velocimetry has continuously evolved and is now being utilized as a tool. As new molecular tracers become available, and lasers and imaging techniques advance, its utility is expected to grow. □

FURTHER READING

It is impossible within the scope of this article to cite all the pioneering work that has been responsible for the current state of this measurement technique. For those interested in further information, the following references may be of interest.

1. C. P. Gendrich, M. M. Koochesfahani, and D. G. Nocera, *Exp. Fluids*, 23, 361 (1997).
2. M. M. Koochesfahani, R. K. Cohn, C.P. Gendrich, and D. G. Nocera, in *Developments in Laser Techniques and Fluid Mechanics*, Chapter 2, section 1, Eds. Adrian, Durao, Durst, Maeda, and Whitelaw; Berlin: Springer-Verlag (1997).
3. M. M. Koochesfahani, American Inst. of Aeronautics and Astronautics and Astronautics Fluid Conference, paper AIAA-99-3786 (July 1999).
4. W. R. Lempert in *Flow Visualization: Techniques and Examples*, Eds. A. J. Smits and T. T. Lim, Imperial College Press, London (2000).
5. *Measurement Sci. and Tech.*, "Special feature: Molecular tagging velocimetry," 11(9) (2000).

Bulk reprints of all Laser Focus World articles can be ordered from Cathleen Johnson, reprints manager, at (727) 843-0628 or FAX (727) 845-7598.