Spatially Resolved Capillary Inlet Mass Spectrometer for Improved Emissions Research

ENERGY EFFICIENCY AND RENEWABLE ENERGY

OFFICE OF FREEDOMCAR AND VEHICLE TECHNOLOGIES

Transportation

FOR THE 21ST CENTURY

Background

High-efficiency, advanced diesel engine technology is a leading near-term option for reducing petroleum consumption and oil imports in the United States. However, new regulations by the U.S. Environmental Protection Agency (EPA), to be phased in over the 2007-2009 model years, will require a 90% reduction in emissions of nitrogen oxides (NOx) and particulate matter. Without development of new emission control systems and more advanced fuels, it is unlikely that diesel engines will achieve the new emission standards.

NOx adsorber catalysts provide a promising approach for emissions reduction in the fuel-lean environment of diesel engine exhaust. NOx adsorbers have a finite capacity to store NOx emissions by forming surface nitrogen species at active catalyst sites. These systems are periodically regenerated to restore capacity by injecting pulses of a reductant, which may be the diesel fuel itself. The reductant causes the stored NOx to be desorbed and reduced, ideally to water, carbon dioxide, and nitrogen gas.

Exhaust gas recirculation (EGR), in which exhaust gas is mixed with incoming air before being inducted into the combustion chamber, is another strategy for lowering NOx emissions. The presence of the recirculated exhaust gas in the mixture lowers the peak in-cylinder temperature—the primary factor determining NOx output. Efficient implementation of EGR requires both a well-mixed and uniform port-to-port exhaust gas/air charge.

High-speed instruments capable of temporally resolving the transient emissions associated with these emission reduction processes are required to develop and optimize such emission control strategies. Minimally or non-invasive diagnostics might allow for intra-channel probing of the catalyst chemistry to quantify the axial distribution of participating species, NOx loading, reductant formation, sulfur poisoning, and desulfation. Similarly, measurement strategies for evaluating the exhaust gas/air charge in EGR systems must be minimally invasive, so as not to induce mixing by the measurement procedure itself. Also, measurements must be made quickly if the EGR waves and pulses are to be resolved.

The Technology

Catalysts, such as NOx adsorbers, sulfur traps, and particulate traps, are typically applied to honeycomb type "monoliths" which contain channels only millimeters wide and several inches long. In operation, these systems exhibit temporally varying spatial chemistry distributions along the channel axis. Measurement strategies capable of resolving these intra-channel temporally varying chemistry distributions are required to clarify detailed catalyst parameters and develop catalyst models.

The spatially resolved capillary-inlet mass spectrometer (SpaciMS) has both the temporal response and minimally invasive nature necessary to resolve relevant emission transients accurately, and to analyze intra-catalyst-channel species distributions, and EGR-charge uniformity. The SpaciMS employs a very small glass capillary to extract and transport a sample to the mass spectro-meter where high-speed measurements are made. The capillaries are specified to minimize temporal broadening of transient species during capillary transport. The SpaciMS head uses electron ionization, quadrupole mass filtering and Faraday cup or continuous-dynode electron-multiplier detection. The SpaciMS is routinely applied for measurement of total NOx, O_2, CO_2 and HC fragments indicative of base fuel and reformed olefin and oxygenate products. High-speed single-ion scans are temporally aligned via a trigger (e.g., reductant injection time) to reveal the detailed phase of the various species transients within the NOx-adsorber cycle. The instrument is transportable and has been deployed to several industrial research laboratories for secure evaluation of advanced emission control systems, including NOx adsorbers and EGR.

Commercialization

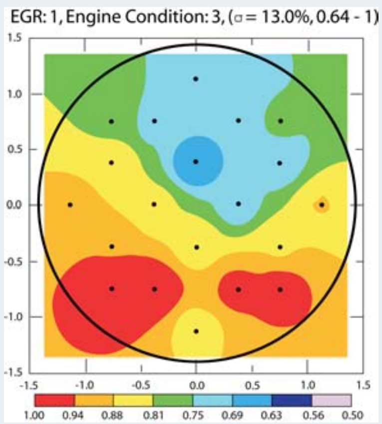
The unique and previously unavailable data provided by the SpaciMS are critical to developing catalyst models; understanding the details of NOx loading, fuel reforming, sulfur poisoning and desulfation; identifying rate-limiting steps; and optimizing catalyst parameters such as reductant quantity required, catalyst aspect ratio and washcoat formulation.

Oak Ridge National Laboratory has decided not to patent this important technology, but instead make it available to all organizations studying exhaust emissions. The SpaciMS measurement strategy is, therefore, being adopted by other government, automotive, industrial, and academic research laboratories.

Benefits

- **High temporal resolution**
- **Minimally invasive**
- **Broad species applicability**
- **Easily transportable**
- **Quantifies intra-catalyst-channel species transients and distributions**

The SpaciMS can rapidly sample and analyze exhaust gases from various locations within the honeycomb of a catalyst-coated monolith.



SpaciMS-measured EGR distribution in a development intake system shows 36% non-uniformity and strong biasing to the bottom of the intake duct. A uniform intake-air/EGR mixture would be indicated by a uniform red field.

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