H. Durability of Diesel Particulate Filters (CRADA with Cummins, Inc.)

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Objective

- Identify and implement test techniques to characterize the physical and mechanical properties of ceramic diesel particulate filters (DPFs) and develop analysis tools for predicting their reliability and durability.

Approach
- Identify and implement test techniques to determine the physical and mechanical properties of DPF ceramic substrates.
- Investigate the applicability of probabilistic design tools to DPF ceramic substrates. In particular, the applicability of the Ceramic Analysis and Reliability Evaluation of Structures (CARES) code to predict the reliability of DPF ceramic substrates will be investigated.

Accomplishments

- Developed a procedure to prepare double-torsion test specimens of porous cordierite.
- Determined the fracture toughness and slow-crack-growth behavior of porous cordierite as a function of temperature and environment.
- Carried out a round-robin testing program to determine the precision of 4-point bending test methods to determine the flexural strength of DPF ceramic substrates.

Future Direction

- Implement probabilistic design tools to predict the reliability and durability of DPF ceramic substrates when subjected to arbitrary thermomechanical histories.

Introduction

The pollution emitted by diesel engines contributes greatly to the nation's air quality problems. Even with more stringent environmental regulations set to take effect in 2007, existing trucks and buses will continue to emit nitrogen oxides (NO_x) and particulate matter (PM), both of which contribute to serious public health problems.[1] Fortunately, there are several technologies designed to reduce pollution from existing trucks and buses, such as DPFs. A DPF is a ceramic device that collects PM in the exhaust stream. The high temperature of the exhaust heats the ceramic structure and allows the particles inside to break down (or oxidize) into less harmful components. DPFs reduce emissions of PM, hydrocarbons, and CO by 60 to 90%.

Most DPFs consist of a ceramic honeycomb with hundreds of cell passages partitioned by walls (Figure 1). Each cell passage has a square cell opening at one end and is closed at the other end so that the cell passages are alternately closed at each end. This structure forces the exhaust gases through the porous, thin ceramic honeycomb walls. When the gases carrying the carbon particles flow through the fine pores of the walls, the carbon particles are filtered out. High porosity values, in the range of 60%, heighten filtration efficiency to more than 90% while reducing gas-flow resistance to avoid affecting the engine performance.

The process of diesel PM collection begins as soon as the engine is started and continues while the engine is operating. As the carbon particles are collected on the ceramic walls, the backpressure of the system increases. This problem is alleviated by burning the trapped PM, through a catalytic reaction using exhaust gas heat at 400°C or more, into CO_2 and water vapor that pass through the filter. This process, called regeneration, results in a cleaner filter. The regeneration process is dependent upon exhaust temperature, oxygen, NO_x content, time, and PM levels.

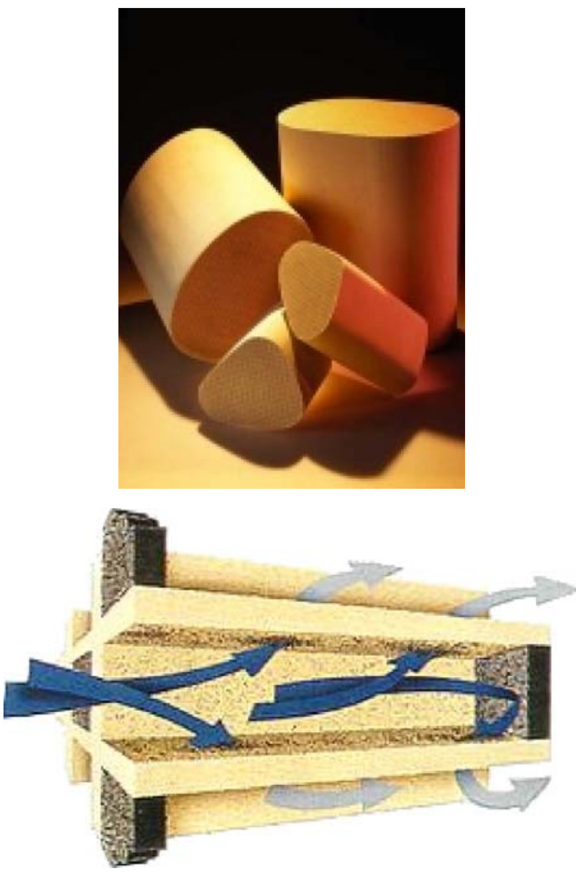


Figure 1. Corning's cordierite-based DPFs.

The key to the successful application of DPFs is to reliably regenerate the filter (e.g., to burn the PM that the filter continues to trap or collect). Traditionally, combustion of soot is done in an oxygen atmosphere (air). In air, soot will burn at about 500°C. However, this is not a typical operating temperature for diesel engine exhaust. Therefore, to burn soot in air, an active system—i.e., one that increases the temperature of the exhaust using some external heat source—is required. But if an active system is not carefully controlled, or if too much PM collects on the filter walls, the filters can experience an "uncontrolled burn" where the temperature increases to 600°C or more, resulting in damage to the filter element.

The objective of this project is to develop and implement methodologies to predict the reliability of DPFs. A useful conceptual model for this purpose is the reliability bathtub curve, which describes reliability-related phenomena of a component over its life cycle.[2] A schematic of the reliability bathtub curve is depicted in Figure 2. It consists of three stages: the infant mortality phase is characterized by premature failures due to improper manufacturing or assembly, poor workmanship, or defects introduced during processing. The second stage of the curve corresponds to the useful life of the component and

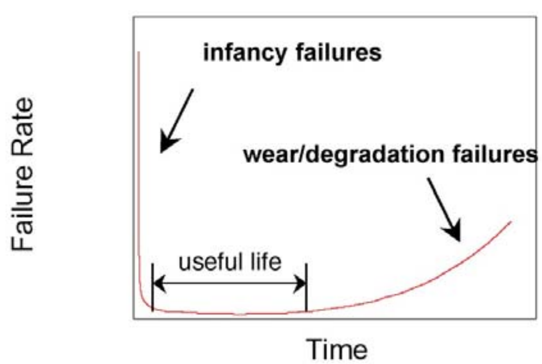


Figure 2. Reliability bathtub curves.

is characterized by a constant failure rate. In this regime, failures are typically associated with random, excessive loads. If sufficiently high safety factors are used during the design process, the magnitude of this failure rate should be negligible. The third stage of the bathtub curve is known as the wear-out phase, where the failure rate increases with time as a result of aging phenomena. Aging phenomena include thermal and mechanical fatigue, corrosion, creep deformation and environmentally assisted crack growth. The time at the onset of wear-out is often regarded as the useful life of the component.

The reliability bathtub curve can be used as a descriptor of how the failure rate of DPFs evolves over time. Infancy failures of DPFs could be related to manufacturing or process defects (large pores, inclusions, cracking) or defects introduced during assembly. Failures of DPFs during their useful life will be dictated by the intersection between the spectrum of thermomechanical loads and the distribution of DPF strengths. Such failures could result from excessive vibration, for example, or unwanted thermal excursions during transients associated with regeneration. Wear-out and degradation failures of DPFs could be associated with the growth of micro-cracks assisted by thermal fatigue, and/or by the chemistry of the environment, and/or by chemical and microstructural changes in the material due to long-term exposure to elevated temperatures in the exhaust environment.

The objective of this project is to develop life prediction methods for DPFs and, by using those methods, to design durable, reliable DPFs.

Approach

Designing DPFs that are durable and reliable poses significant challenges. For example, the porosity of DPFs, which allows them to remove PM from the exhaust gas stream, has a deleterious effect on their mechanical strength. This is important because DPFs will experience demanding thermo-mechanical conditions during service. These include, for example, thermal shock resulting from rapid heating/cooling and stresses that arise from temperature gradients. The approach that will be followed in this project includes identifying and implementing test techniques for the nondestructive evaluation of ceramic substrates to assess their integrity. Their physical and mechanical properties and the mechanisms responsible for their degradation in the various stages of the bathtub curve also will be determined and identified. These properties include thermal expansion, thermal conductivity, heat capacity, density, porosity, elastic properties, strength, fracture toughness, and resistance to crack growth at ambient and elevated temperatures, in air and in relevant environments.

The information generated will be used in turn to implement probabilistic design tools. In particular, the applicability of the CARES code[3] to predict the reliability of DPF ceramic substrates will be investigated. Such probabilistic design methodologies are based on a combination of experimentally determined strength data, stress analyses of the component using a finite-element analysis, and selection of appropriate failure criteria. The durability (service life) of the component can also be predicted using this framework by considering the mechanisms that are responsible for the degradation of material strength, such as slow crack growth or creep. While these methodologies have been successfully applied to design structural components using dense ceramics, their applicability to the analysis and design of porous ceramic components has not been validated.

Results

During FY 2005, a procedure was developed to prepare double-torsion test specimens to determine the fracture toughness and slow-crack-growth behavior of porous cordierite. The double-torsion test configuration (Figure 3) consists of symmetric four-point loading around a crack or a notch on one end of a rectangular plate. One feature of this loading configuration is that the stress intensity factor is, at a first approximation, independent of crack length for a range of crack lengths. This feature makes double-torsion testing ideally suited for the evaluation of opaque and non-reflective plate-like test specimens where direct crack length measurements could be difficult to make. The double-torsion test method is also attractive for slow-crack-growth studies because of the relative stability of crack extension, in contrast to other testing configurations (e.g., SENB, CT).

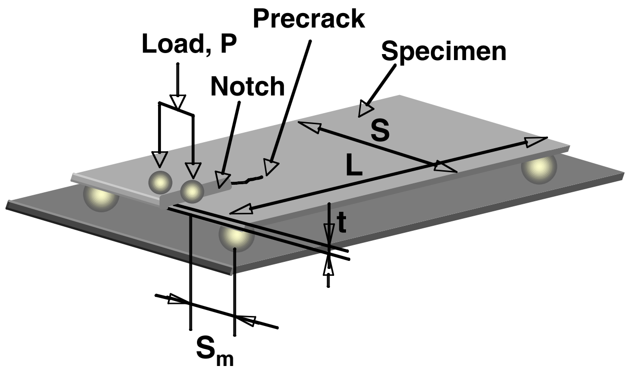


Figure 3. Schematic of the double-torsion test configuration.

The preparation of double-torsion test specimens of porous cordierite, which poses challenges because the thickness of the DPF cell walls is of the order of 300 µm, was accomplished following dry cutting/grinding operations. Sections from a DPF with a cell size of 2.25 × 2.25 mm and approximately 50% porosity were sliced using a diamond cut-off blade. Then subsequent material removal was accomplished using a diamond grinding wheel (150 grit, 4000 rpm, 25 µm down feed). To ensure parallelism and flatness of the surfaces of the test specimen, a special fixture was fabricated using a precision vice and gage blocks. Test specimens were prepared with the notch aligned either parallel or perpendicular to the orientation of the walls, and the final thickness of the test specimens was 280 µm. Figure 4 illustrates the sample preparation process.

The fracture toughness of porous cordierite test specimens was determined by loading pre-cracked test specimens to failure at a constant crosshead displacement rate of 0.01 mm/s. The fracture toughness was calculated from the peak load using Equation 1:

$$K_{IC} = PS_m \left[\frac{3(1+\nu)}{St^4 \xi} \right]^{1/2} , \qquad (1)$$

where P is the peak load, S is the width of the test specimens, S_m is the moment arm, t is the specimen thickness, ν is Poisson's ratio, and ξ is a finite beam thickness correction factor given by

$$\xi = 1 - 1.26 \left(\frac{t}{S} \right) + 2.4 \left(\frac{t}{S} \right) e^{\left(-\frac{\pi S}{2t} \right)} . \qquad (2)$$

Figure 5 shows a cordierite test specimen after a fracture toughness test, illustrating the trajectory of

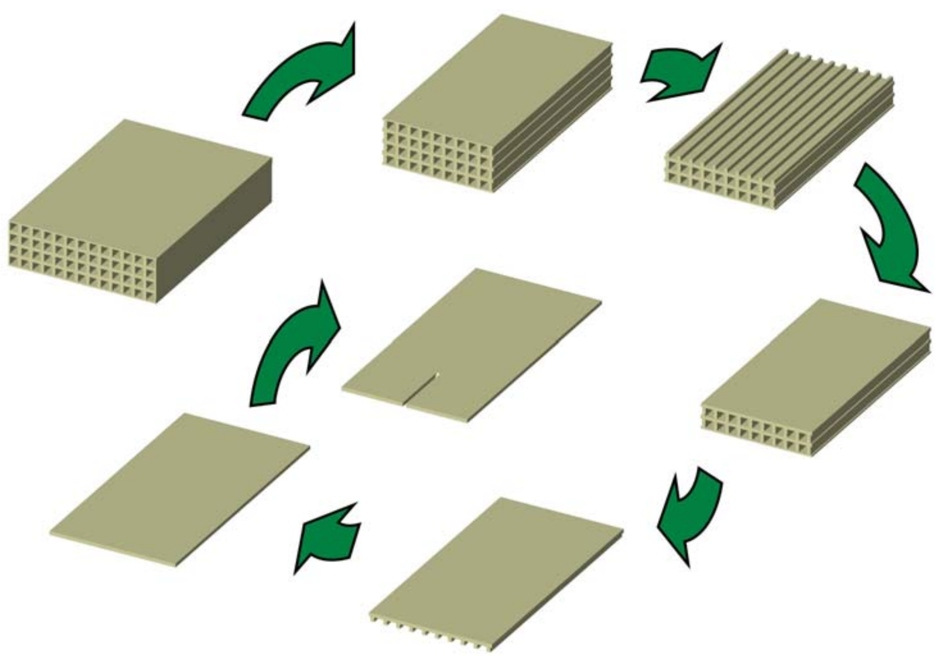
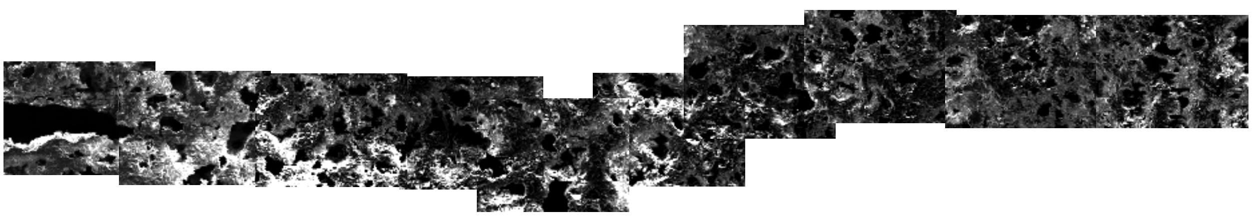


Figure 4. Schematic of procedure followed for the preparation of double-torsion test specimens.

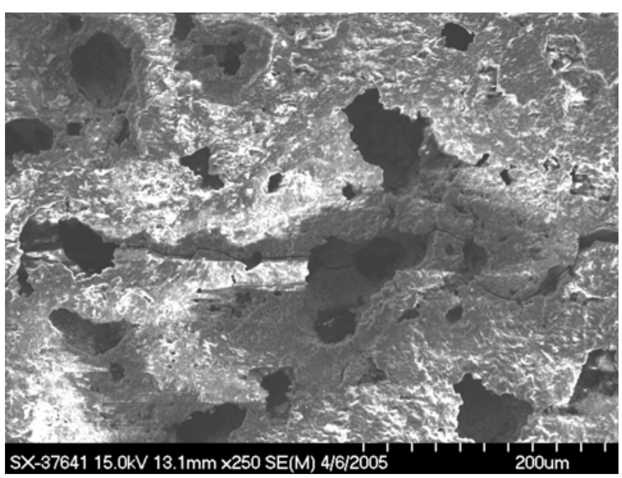
Figure 5. Photograph of double-torsion test specimen after fracture toughness testing.

the crack, which propagated along the major axis of the test specimen. In Figure 5, the notch is located on the right of the test specimen. It was found that the propagation of the crack was significantly influenced by the microstructure of the material, as illustrated by the scanning electron micrograph in Figure 6. To better characterize the interaction between the microstructure and crack propagation, exploratory work was carried out to determine the feasibility of imaging the propagation of the crack through a double-torsion test specimen using X-ray computerized tomography (CT) scans. In addition to providing information on the role of the microstructure in the resistance of the material to crack propagation, this work could lead to the implementation of nondestructive evaluation techniques. Figure 7 shows a reconstructed image of a porous cordierite test specimen. Additional work will be pursued in this area in FY 2006. The fracture toughness of porous cordierite was found to decrease with temperature from a value of 0.45 ± 0.02 MPa$\sqrt{m}$ at 20°C to 0.36 ± 0.07 MPa$\sqrt{m}$ at 800°C. The plot in Figure 8 summarizes those results.

During FY 2005, work was initiated to investigate the slow-crack-growth behavior of porous cordierite using the load relaxation version of the double-torsion test specimen. According to this test procedure, a pre-cracked test specimen is loaded at a constant crosshead displacement rate that is lower than the load associated with fracture toughness. Then the crosshead is arrested and the load is monitored as a function of time. From the rate of load relaxation, which results from the increase in



(a)



(b)

Figure 6. Scanning electron micrograph illustrating the influence of microstructure on crack propagation: (a) low magnification and (b) high magnification.

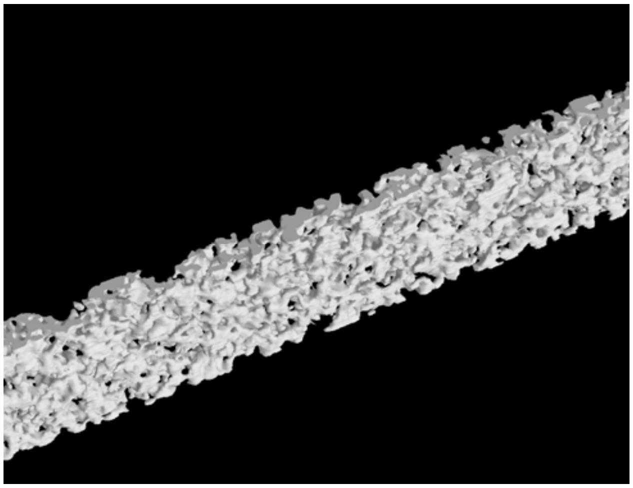


Figure 7. Reconstructed image of porous cordierite test specimen from X-ray CT scan.

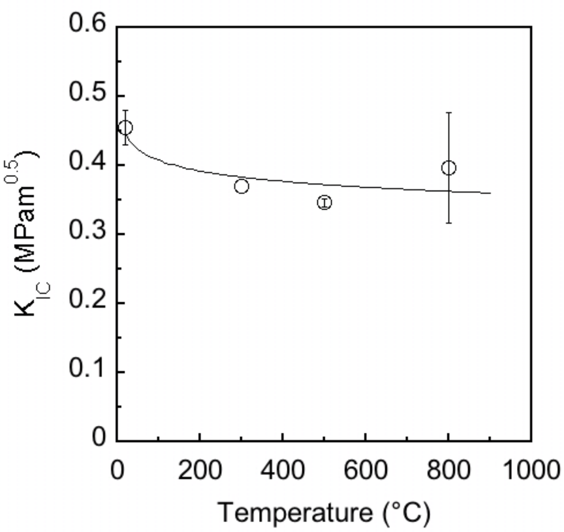


Figure 8. Fracture toughness of porous cordierite as a function of temperature.

compliance of the test specimen, it is possible to determine the rate of crack growth using Equation 3:

$$\frac{da}{dt} = -\frac{P_i}{P^2}\left[a_i + \frac{D}{B}\right]\left(\frac{dP}{dt}\right) , \qquad (3)$$

where P_i is the initial load, a_i is the initial crack length, and D and B are experimental constants associated with the load train compliance.

In parallel with these efforts, a series of dynamic fatigue tests were also initiated in FY 2005 to determine the slow-crack-growth behavior of porous cordierite according to ASTM C1465-00, "Standard Test Method for Determination of Slow Crack Growth Parameters of Advanced Ceramics by Constant Stress-Rate Flexural Testing at Elevated Temperatures." According to this test method, the flexural strength of honeycomb test specimens is determined as a function of applied stress rate. The strength degradation exhibited with decreasing applied stress rate is the basis of this test method, which enables the evaluation of slow-crack-growth parameters of a material.

This dual approach to the characterization of the slow-crack-growth behavior of porous cordierite is necessary to distinguish between material behavior, which will be obtained through the double torsion testing of test specimens obtained from the walls of cellular structures, and structural behavior, which will be obtained through dynamic fatigue flexural testing of beam test specimens with cellular structure. The results from these two sets of tests will provide the kinetics for crack growth to enable the prediction of the service life of porous cordierite DPFs using CARES. The slow-crack-growth behavior of porous cordierite and porous cordierite DPFs with cellular structure will be determined in air, water-vapor, a reducing environment, and simulated diesel exhaust environments.

In FY 2005, a round-robin testing program was organized between Oak Ridge National Laboratory (ORNL) and Cummins to determine the precision in the determination of the 4-point flexural strength of cordierite DPFs. Each laboratory was responsible for testing 40 test specimens with nominal dimensions of 100 × 26 × 13.5 mm. Tests were carried out at ambient conditions under a constant crosshead displacement of 0.35 mm/min, using inner and outer spans of 19 and 89 mm. The test results, which were analyzed using Weibull statistics, are presented in Figure 9. It was found that the average and standard deviations of all tests were 6.73 and 0.83 MPa, respectively, and that the repeatability and reproducibility standard deviations were 0.35 and 0.9 MPa, respectively. The Weibull modulus, which was determined according to the maximum likelihood method, was found to be 24.1. The discrepancies in the characteristic strength values were associated with procedures used to determine the dimensions of the test specimens.

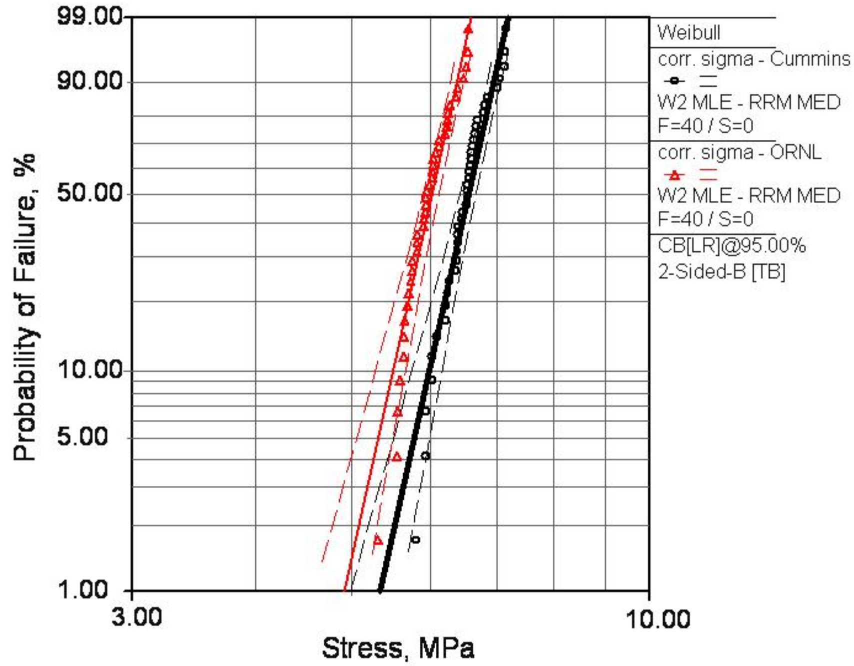


Figure 9. Weibull plot summarizing flexural strength results from round-robin testing program. (△: ORNL data; ○: Cummins data).

Summary

A procedure was developed to prepare test specimens from cellular DPFs to determine the fracture toughness of porous cordierite by the double-torsion test method. Using this test method, the fracture toughness of porous cordierite was found to decrease with increasing temperature from 0.45 ± 0.02 MPa$\sqrt{m}$ at 20°C to 0.36 ± 0.07 MPa$\sqrt{m}$ at 800°C. Work was initiated to determine the slow-crack-growth behavior of porous cordierite and cellular structures of porous cordierite, using the load relaxation variation of the double-torsion test method and the constant stress-rate flexural test method according to ASTM C1465, respectively. The results from these tests will enable prediction of the service life of porous cordierite DPFs. A round-robin testing program was completed to assess precision in the determination of the 4-point flexural strength of cordierite DPFs. It was found that the average and standard deviations of all tests were 6.73 and 0.83 MPa, respectively and that the repeatability and reproducibility standard deviations were 0.35 and 0.9 MPa, respectively. The Weibull modulus was found to be 24.1

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Collaborators

Amit Shyam, H-T. Lin, Randy Parten, and Claire Luttrell.

Presentations and Publications

A. Shyam and E. Lara-Curzio, “The Double-torsion Testing Technique for the Determination of Fracture Toughness and Slow-Crack Growth Behavior of Materials: A Review,” accepted for

publication in the *Journal of Materials Science* (2005).

T. Yonushonis and E. Lara-Curzio, "Durability of Particulate Filters—PM 10461," presented at the FY 2005 Heavy Vehicle Materials Program Review, Oak Ridge National Laboratory, Oak Ridge, Tennessee, September 14, 2005.