Wiremesh Substrates for Oxidation, TWC and SCR Converters

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Abstract

The primary requirements of exhaust after treatment systems are low back pressure, low system weight, better emission performance and lower cost. Combinations of these properties provide better engine performance and higher system value. Knitted wiremesh substrates with different geometry and channels are used as carriers of diesel oxidation catalyst, three way catalyst and selective catalytic reduction (SCR) NOx catalyst. Wiremesh materials are chosen not only to withstand corrosion and erosion environments but also durable at maximum continuous operating temperature and peak thermal spikes. Wiremesh substrates provide radial and longitudinal flows (depth flow) and turbulent kinetic energy. The back pressure of the substrate is optimized using different material properties and knitting patterns. Substrate geometry is optimized using different brazing materials and heat treatment conditions. The dimensional tolerances and the process capability of the substrate contours are established. The longitudinal and radial depth flows within the body of the substrate is modeled using computational fluid dynamic analysis. The uniformity index, velocity index, back pressure and turbulent kinetic energy are characterized by fluid dynamic modeling. The catalyzability, packagability and physical durability of the substrates are tested using different wire types and wire mesh patterns. Wiremesh substrates with turbulent and torturous flows could be used as a static mixer for the SCR Urea NOx reduction system for increased performance and avoid urea crystallization.

Introduction

Understanding the basic properties of the components of the exhaust system and their relationship to the whole is essential to design the best catalytic converter [1-3]. For the catalytic converter, this includes the substrate, washcoat, catalyst and the mounting system. The substrate inside the catalytic converter acts as a catalyst carrier to provide sufficient active sites for the exhaust gas to be in contact with the catalyst [4,5]. Critical issues when designing a catalytic converter substrate are emission conversion efficiency, long term durability, weight, volume and available space [6-8]. Substrate-related variables that impact one or more of the design criteria are material composition, structure, contour, geometry, length, thermal and acoustic insulation, washcoat formulation, catalyst formulation and container design. Many of the early automotive catalytic converters in the 1970s utilized alumina based pellets and bead-shape supports placed into a steel shell and contained between two screens [9]. To avoid attrition and high back pressure, ceramic monolithic honeycombs using 2 MgO.2 Al_2O_3.5 SiO_2 (cordierite) are used instead of pelleted catalyst [5,6]. Metallic monolithic converters using thin metal foils of ferritic iron-chromium-aluminum alloys and other materials are also used in catalytic converters [10,11]. Different types/shapes of catalyst carriers are also used for improved emission performance of the catalytic converters [12, 13]. Although geometric surface area and open frontal area are two major design criteria of the conventional honeycomb substrate, the light-off efficiency depends more on the mass and heat transfer coefficients. This is true in the case of the non-conventional substrates such as wiremesh since the flow geometry itself is completely different from that of the conventional substrates.

Wiremesh Substrate

A wiremesh substrate forms the core of a catalytic converter to provide support structure and geometric surface area upon which the washcoat and the catalyst are applied. Since the commercial substrate has low surface area, it is necessary to deposit a washcoat with much higher surface area to provide the effective surface area to facilitate the application of precious metal catalysts onto the surface. The pollutant-containing gases enter the substrate and diffuse to and through the washcoat pore structure to the catalytic sites where they are converted catalytically. Wiremesh substrates are made with custom flexible design and fully adaptable to packaging. Wiremesh substrates have extremely high vibration and thermal shock resistances. Wiremesh provides turbulent mixing and radial heat transfer. Figure 1 shows the wiremesh substrates with different dimensions.

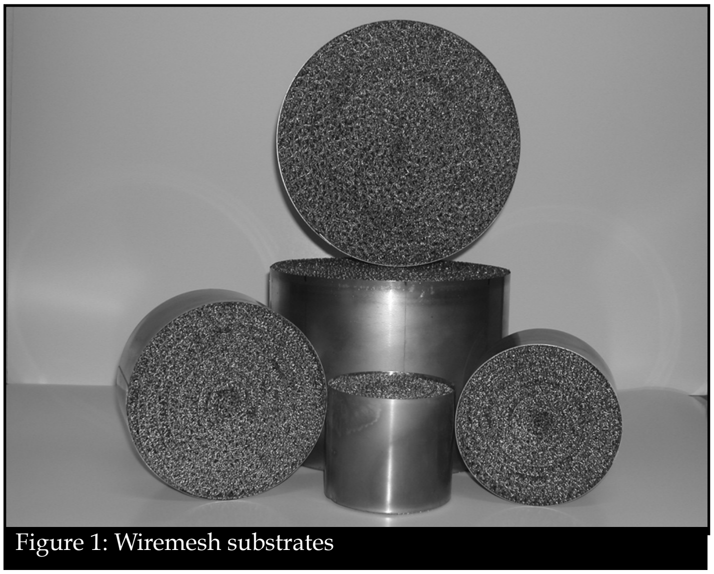

Figure 1: Wiremesh substrates

Substrate Physical Attributes

Several features of the automotive catalyst support contribute to the performance of a catalytic converter system. Certainly high surface area, straight and uniform channels provide active catalytic surface while still showing a comparatively low back pressure. Other properties of the substrate such as mass and specific heat capacity prove detrimental to the rapid attainment of high conversion efficiency. The size and shape of the channel also can have positive and negative effects, depending on the relative values of the factors which contribute to both the back pressure and the heat and mass transfer. In turn, the mass transfer is directly related to the catalyst performance.

Substrate attributes such as material composition, structure, geometry, orientation, wire strength, mesh pitch, depth, crimp angle and the right contour are chosen to get maximum oxidation resistance, high corrosion/erosion resistance, high acoustic advantages, low back pressure and lower weight. The higher surface area to volume ratio and high mass transfer effects provide enhanced emission control efficiency.

Substrate Material

The material used for the wiremesh substrate is application specific. The maximum operating temperature and the environment of the converter mainly determine the material selection. 310S grade stainless steel, offering excellent high temperature properties with good ductility and weldability is designed for continuous operating temperature upto 1150°C. It resists oxidation in continuous service at temperatures up to 1150°C and in intermittent service at temperatures up to 1040°C. Grade 310S is used when the application environment involves moist corroding. The high chromium content, intended to increase high temperature properties give good aqueous corrosion resistance. It has excellent oxidation resistance at normal temperatures and, when in high temperature service, exhibits good resistance to oxidizing and carburizing atmospheres. 310S has good resistance to thermal fatigue and cyclic heating. If the exhaust gas temperature falls below 200°C, ammonium nitrate may be formed which may deposit in the pores of the catalyst in solid or liquid form, leading to its temporary deactivation. 310S resists fuming nitric acid at room temperature and fused nitrates up to 425°C.

When the exhaust gas contains sulfur, as is the case with diesel exhaust, SO_2 can be oxidized to SO_3 with the following formation of sulfuric acid upon reaction with water. 310S has excellent resistance to sulfur dioxide gas. Ferritic iron-chromium-aluminum (FeCrAlloy) and nickel containing alloys are also used for high temperature applications. The chemical and material properties of few commonly used substrate materials are given Table 1.

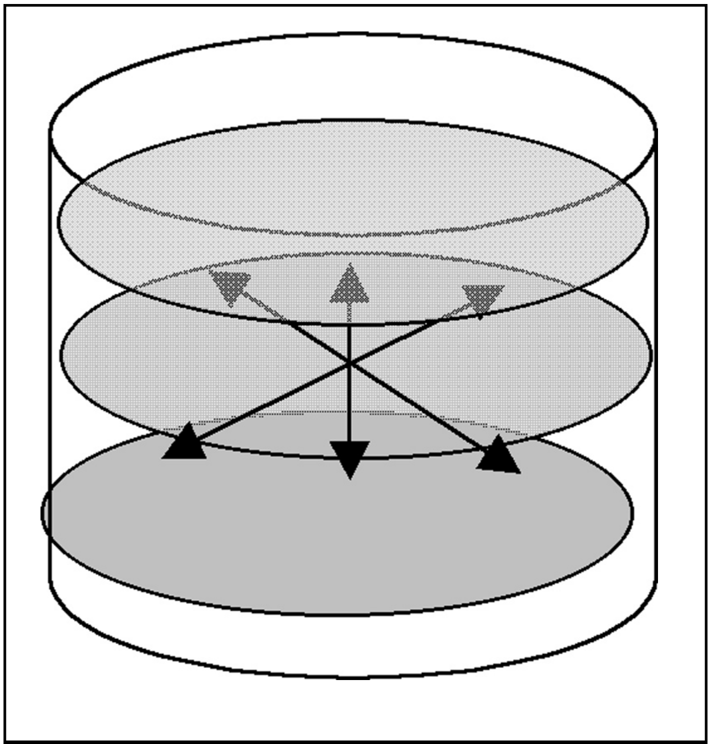

Figure 2: Measurement locations for substrate contour concentricity

Material	304	310S	FeCrAlloy
Chemical Composition			
Chromium %	18 - 20	24 - 26	22
Aluminum %	0	0	4.80
Iron %	68 - 74	48 - 55	73.2
Nickel %	8 - 12	19 -22	0
Carbon %	0.08	0.08	0
Physical Characteristics			
Melting Point (°C)	1400	1400	1500
Maximum operating temperature(°C)	900	1100	1300
Density (g/cm^3)	8.03	8	7.25
Electric resistivity (μohm-m at 20 °C)	0.72	0.72	1.35
Coeff. Of thermal expansion (μm/m K)	17	15.9	11
Thermal Conductivity (W/m K)	16.2	14.2	11
Specific Heat (kJ/kg K)	0.50	0.5	0.46
Mechanical Properties			
Ultimate Tensile Strength (MPa)	621	515	673
Yield Strength (MPa)	290	205	485
Elongation at rupture (%)	55	40	22

Table 1: Wiremesh substrate material properties

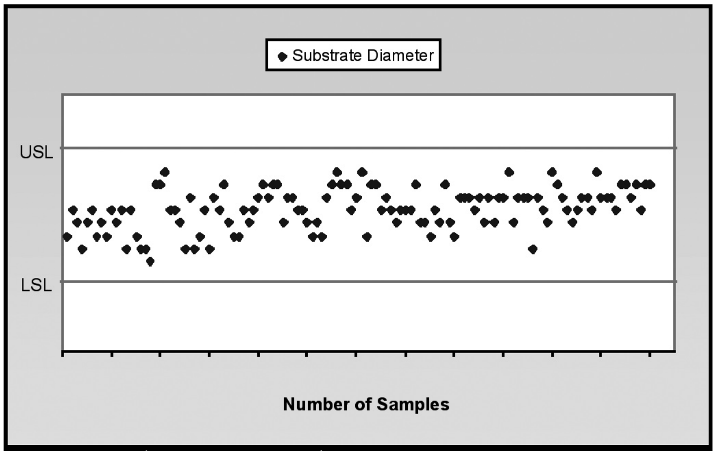

Figure 3: Substrate contour diameter

Substrate Construction

The properties of the substrates are modified by altering wire diameter, number of strands, orientation angle, pitch and depth. Wiremesh with a wire diameter of 0.014” is standard for diesel applications. Current development allows wire diameter of 0.011” also. The thin diameter wires give lower weight and higher surface area and correspondingly faster conversion. The flow distributions within the channels improve intensive exchange between the gas molecules and the surface. The knitted wiremesh is rolled to the required shape applying brazing material. The wiremesh layers are brazed together to avoid fluting and to provide structural integrity and durability. The wiremesh is brazed with brazing alloy using vacuum brazing or controlled reducing atmosphere. Vacuum furnace uses low atmospheric pressures instead of the protective gas atmosphere used in heat treating furnaces. Vacuum brazing is widely used to braze base metals of stainless steel. Vacuum brazing offers the combination of high cleanliness and uniform heating and cooling and rapid cooling. Cold wall furnaces are used in vacuum brazing. Substrates are bright and clean after vacuum brazing because the extremely low oxygen in the vacuum atmosphere prevents oxidation of the parts.

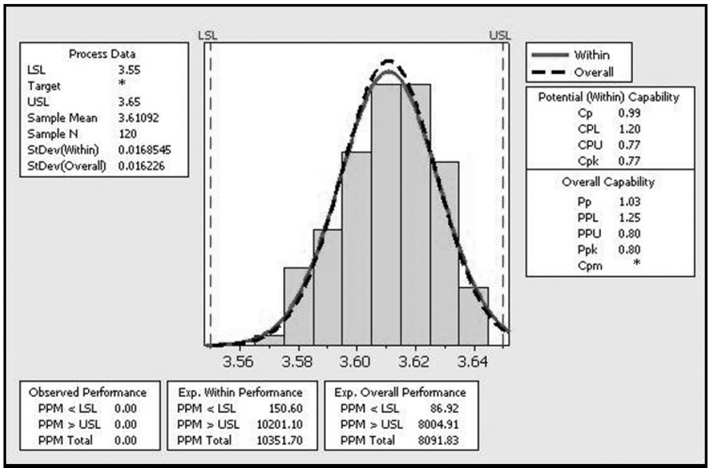

Figure 4: Substrate contour process capability

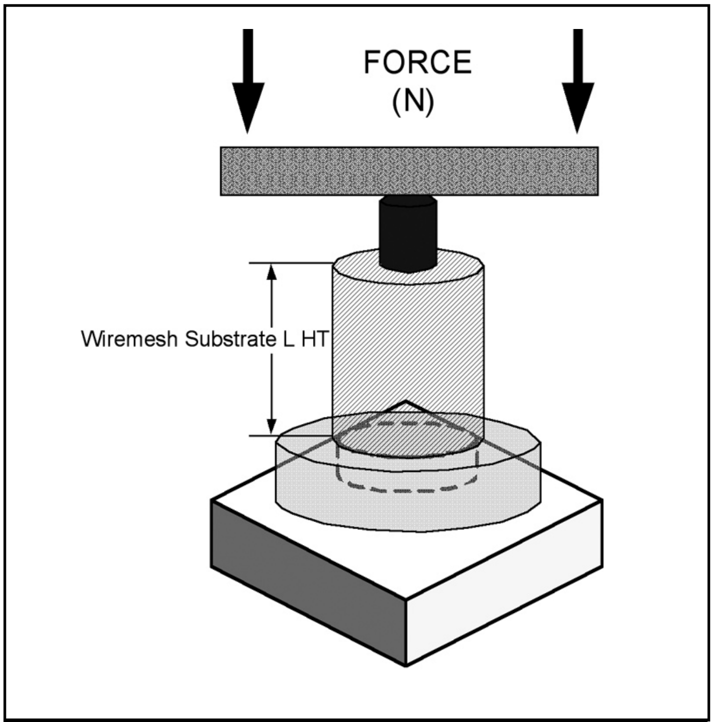

Figure 5: Wireemsh push-out force measurement fixture

The braced rolled mesh is heat treated to form the required stable geometric structure. The dimensional tolerances of the A, B and C axes of the substrate contours are established by controlled rolling, brazing and heat treatment. The concentricity of the substrate geometry is established by measuring the diameter of the substrate at top, middle and bottom locations at every 45 degree angle as shown in Figure 2. The substrate diameter values measured are shown in Figure 3.

The geometric tolerances are optimized and process capability established. A representative process capability of the round substrate is given in Figure 4.

Substrate Braze Strength

The braze strength is optimized by selecting proper filler and binder and is verified by push-out test and hot shake tests. The push out measurement tool is shown in Figure 5. The brazed sample is kept inside the set up as shown and the force necessary to push the core is measured using pre-calibrated instron set up. The measured upshot force as a function of deflection distance is plotted (Figure 6). The plot is different from the typical push out force measurement as this will show a continuous force rather than the conventional push out dislodging curve, as the rolled mesh is a continuous strip.

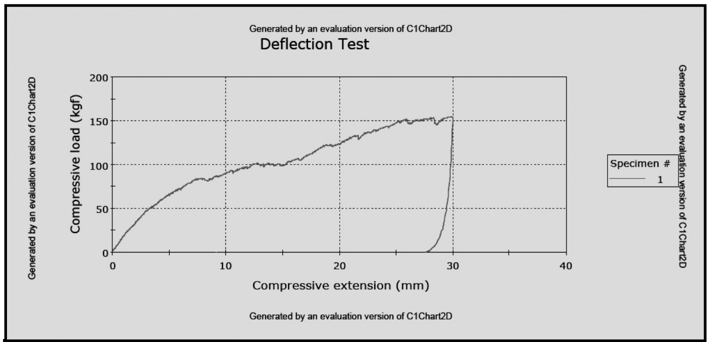

Figure 6: Deflection measurement

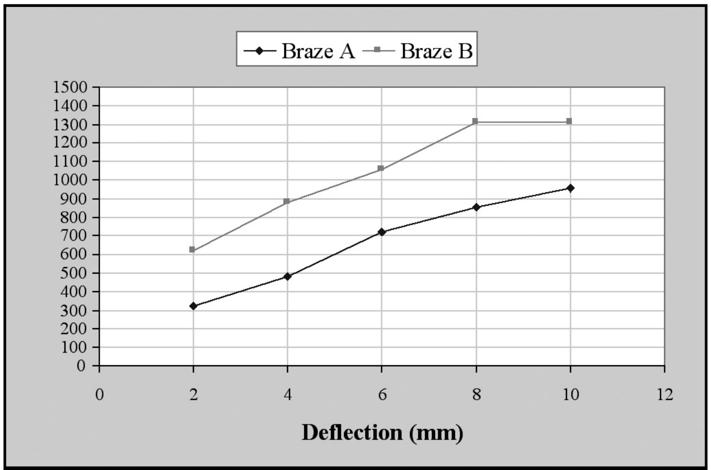

Figure 7: Brazed substrate deflection curve

Figure 7 shows the strengths of the different brazing materials used in the brazing process. Brazing material, brazing environment and brazing conditions are varied to optimize the strength of the brazed substrate. The strength of the substrate is then compared with the minimum back pressure force exerted on the front and back of the substrate. To further confirm the substrate braze strength at elevated temperatures, the substrate mounted with axial support ring is put on the hot shake table and the converter assembly is shaken at a given G load and vibration frequency at elevated temperature. The sample preparation for the hot shake test assures only the failure condition of the brazed material and not the mounting system failure.

Back Pressure

The back pressure of the wiremesh substrate is studied using the flow bench presented in Figure 8. The wiremesh substrate is fixed inside a metallic tube ensuring no leak and the tube is clamped to the flow bench pipe. By adjusting the control valve the flow is varied and for specific flow the corresponding pressure drop is measured. The crimping pattern with controlled crimp angle, pitch, and depth alters the flow characteristics. The back pressure of the substrate is varied by changing the channel angle. The back pressure of the wiremesh substrate as a function of flow is shown in Figure 9.

Figure 8: Substrate assembly for back pressure study

Thermal Expansion and Thermal Shock Resistance

The thermal expansion of substrate material is the most important property which affects the thermal shock resistance of the substrate. The thermal expansion on the A, B and C axes is attributed to its complex structure and the anisotropic of lattice expansion. Due to preferential orientation of the materials in the substrate, anisotropic expansion can occur in a preferred orientation and it can result in low thermal expansion on the C axis. The thermal shock parameter (TSP) is defined as the ratio of mechanical strain tolerance to thermal strain imposed by the radial temperature gradient. TSP is often used as a relative measure of the material ability to withstand steep temperature gradients [6,7]. Due to nonuniform heat flow and heat loss to ambient the center region of substrates experience high temperature than its periphery. This creates temperature gradients and induces tensile stresses causing fracture of the substrate. The higher the TSP, the better the thermal shock resistance (TSR) capability of the material.

$$TSP = (MOR/E) / [\alpha c(tc - 25) - \alpha p(tp - 25)]$$

where MOR is modulus of rupture(Pa), E is elastic modulus(Pa), αc and αp are coefficient of thermal expansion at the center and peripheral region of the substrate(1/C), tc and tp are the temperature at the center and peripheral region of the substrate(1/C). Thermal expansion of material affects the TSR and becomes one of the important design characteristic. The lower the thermal expansion, the higher is the TSR. The mechanical properties which influence the TSR are mainly mechanical strength and elastic modulus. High strength and low elastic modulus is required for getting high TSR. The temperature distribution across the wiremesh substrate is uniform and thus the temperature gradient is low. The wiremesh substrate has high thermal shock resistance.

Wiremesh Flow Modeling

Fluid flow of the converter was investigated using computational fluid dynamics. It is well known that the catalytic converter performance and the pressure drop are substantially affected by fluid distribution of the exhaust gas within the substrate [14]. A porous media model is used to treat the substrate in the flow analysis, which represents a pressure resistance to the fluid. Computational

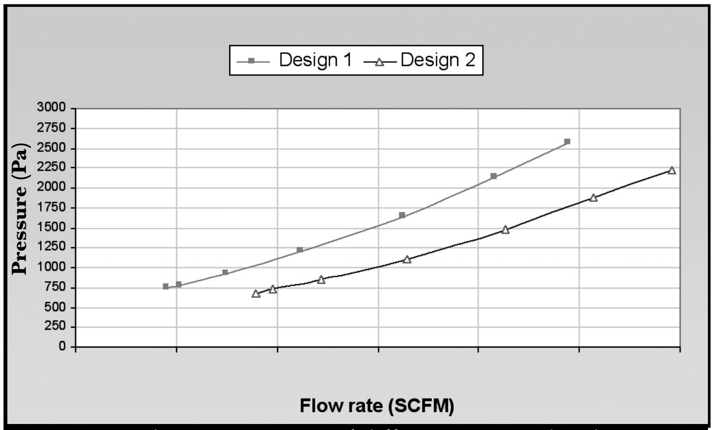

Figure 9: Back pressure curve of different wiremesh substrate

fluid dynamics analysis is done on wiremesh substrate using Fluent 6.2 software. The boundary condition specified is mass flow inlet and constant pressure outlet (mass flow rate 1700 kg/hr @ 575°C).. The geometry of the converter design examined in the analysis is shown in Figure 10.

The fluid flow solver used in this analysis is a three-dimensional incompressible turbulent flow. The governing equations are Navier-Stockes equations, which can be found in most common fluid dynamics books [14,15]. The κ-ε turbulence model is used.

This porous media model simulates a pressure resistance to the fluid. Within the substrate, the flow is assumed to be a mix of turbulent and laminar flows. The momentum equation takes the form:

$$\frac{\partial p}{\partial z} = -\alpha V^2 - \beta V$$

$$\alpha = \frac{Lc_1 \rho R_e{}^n}{2\theta^2 A}, \quad \beta = \frac{Lc_1 c_2 \mu R_e{}^n}{2\theta^2 A D_h}$$

$$R_e = \frac{\rho V' D_h}{\mu}, \quad V' = V/\theta$$

Where z is the flow direction; V the velocity in the substrate channel; c_1, c_2 and n are constants pertaining to the type of flow; θ is the porosity of the substrate. The effects of the substrate properties (material, cell density, wall thickness and coating) on flow are included in the α, β parameters.

Flow uniformity can be defined in several ways [16-18]. By defining a flow uniformity index C_d, the effect of the geometry of the exhaust manifold and converter on flow mal-distribution at the inlet cross-section of the monolith could be better understood. The average velocity of the inlet space is calculated as follows.

$$V = \Sigma_i V_i / n$$

Where V_i is the velocity component in the stream V is the direction for a single cell and n is the total number of cells at the selected plane. A local uniformity index for a single cell is then defined as;

$$C_i = (V_i - V)\ 2 / V_i$$

The uniformity index C_d is then calculated using

$$C_d = 1 - \Sigma_i C_i / 2n$$

Hence, C_d has values from zero (worst case) to one (uniform flow). A more uniform flow increases the useful life of the catalyst, while reducing the risk of "in-use" emissions and/or thermal, mechanical, and chemical durability failures.

Characteristics for efficient flow defines high velocity flow area where the minimum flow velocity is 65% of the maximum flow velocity and the low velocity area as the area where the minimum flow velocity is 35% of the maximum flow velocity (17). By rule of thumb, the high velocity area must utilize 40% of the catalyst frontal area and the low velocity flow must utilize about 90% of the catalyst front face of the substrate.

The uniformity index was modeled throughout the length of the wiremesh substrate. The uniformity index increased through the body of the depth flow substrate due to radial and longitudinal

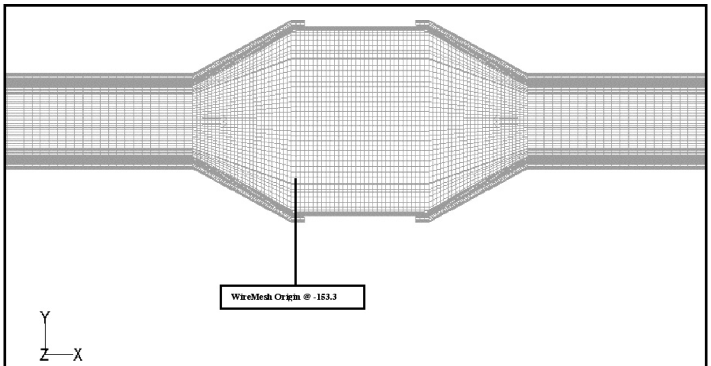

Figure 10: Wiremesh substrates mesh cut through center plane

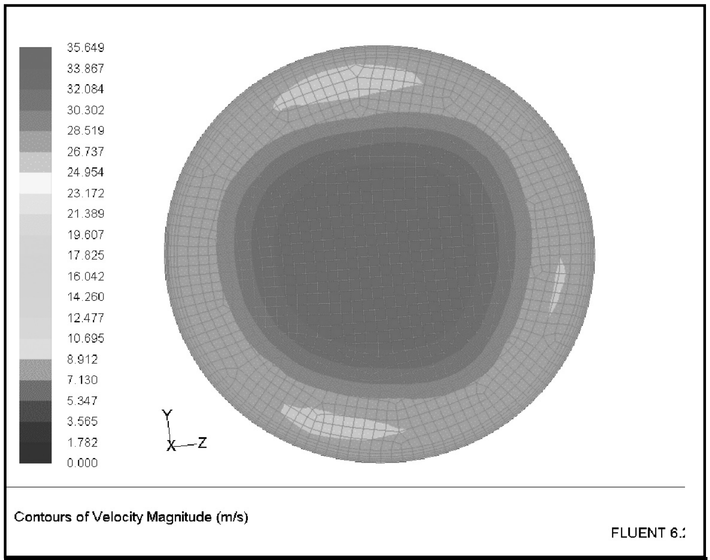

Figure 11: Flow distribution at 15 mm from front face of wiremesh substrate (Gamma = 0.9433)

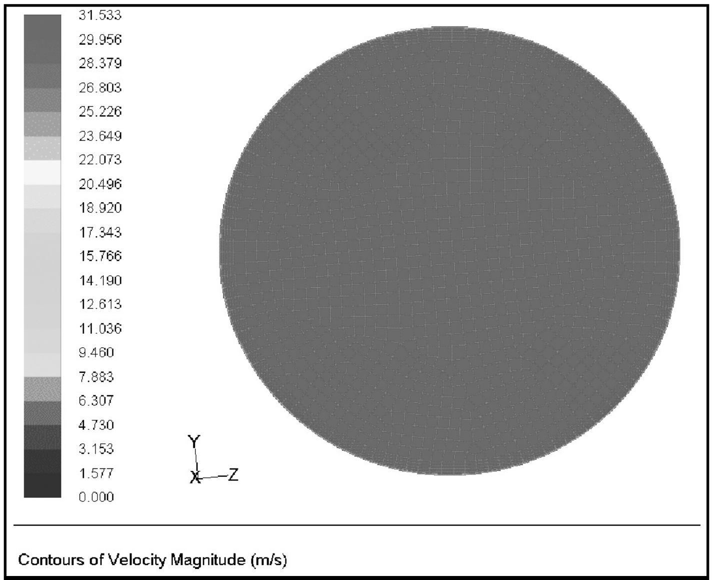

Figure 12: Flow distribution at 35mm from front face of wiremesh substrate (Gamma = 0.9975)

flows occurring through the body of the substrate. Computational fluid dynamics studies show the increase in uniformity index from the front face of the substrate to the body of the substrate. The flow is found to be very well distributed just 35 mm from the front face of the wiremesh substrate. The flow is very stable in wiremesh substrate and attained flow uniformity index (γ) of 0.99 at a length of 65 mm from the front face of substrate, where the uniformity index is about 0.94 at 15 mm from the front face of the substrate (Figures 11, 12). In the wiremesh substrate the uniformity (faster improvement in γ) is easily achieved by depth flow inside the substrate.

Wiremesh substrates provide uniform distribution of the flow vectors (Figure 13-15). As seen in Figure 13 and 14, the velocity profile is found to be fully uniform within 35 mm from the front face of the susbtrate due to longitudinal and radial flow through the wiremesh. The computer simulated static pressure model of the wiremesh substrate is given in Figure 16. The turbulent and torturous flow path provides higher residence time for the exhaust gas to be in contact with the catalyst on the surface of the substrate. The wiremesh substrate enhances the mass transfer between the exhaust gas and the substrate material due to tortuous radial and longitudinal (zigzag) flow. Flow through the wiremesh substrate results in higher turbulent kinetic energy aiding better heat and mass transfer (Figure 17). Wiremesh catalysts are less prone to clogging due to their design. The increased mass transfer co-efficient increases the emission performance. The turbulent and torturous flow path provides higher residence time and contact time. The

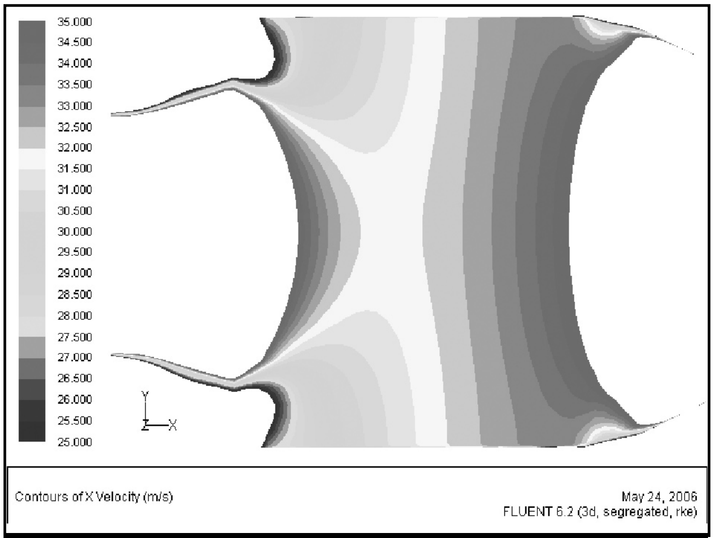

Figure 15: Velocity contour of wiremesh substrate on mid-plane

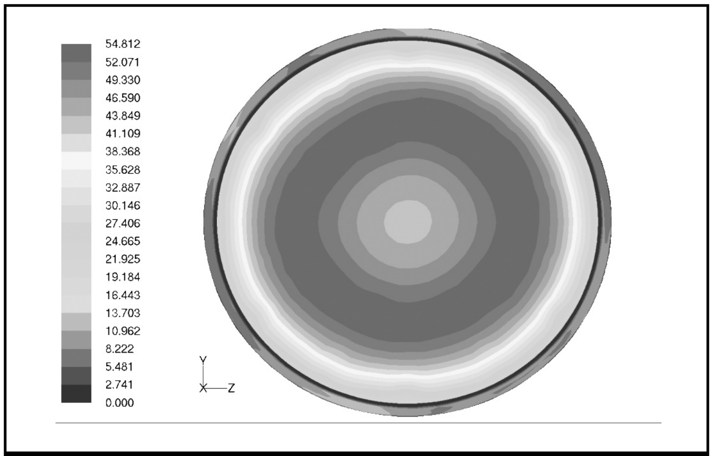

Figure 13: Velocity contour at front face of wiremesh substrate

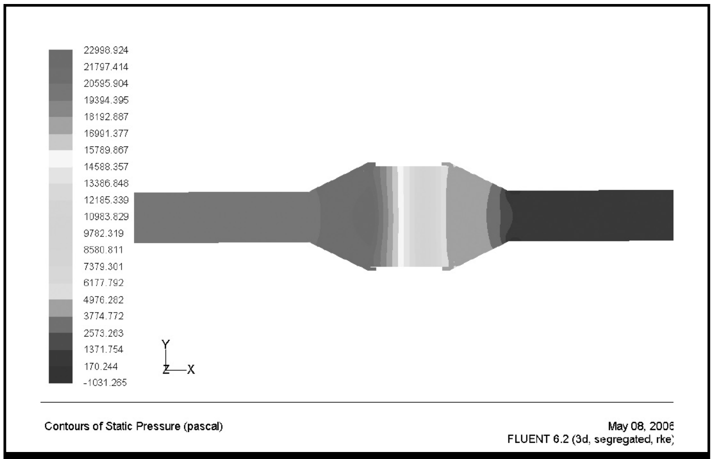

Figure 16: Wiremesh substrate static pressure model

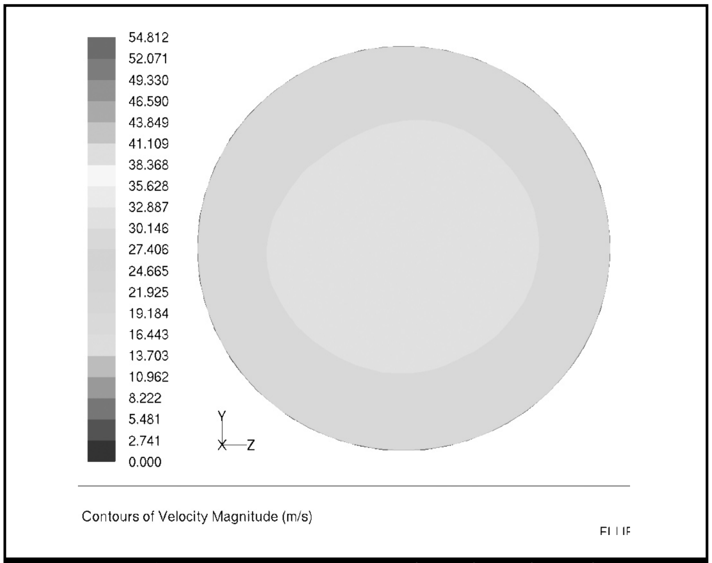

Figure 14: Velocity contour at 35 mm from front face of wiremesh substrate

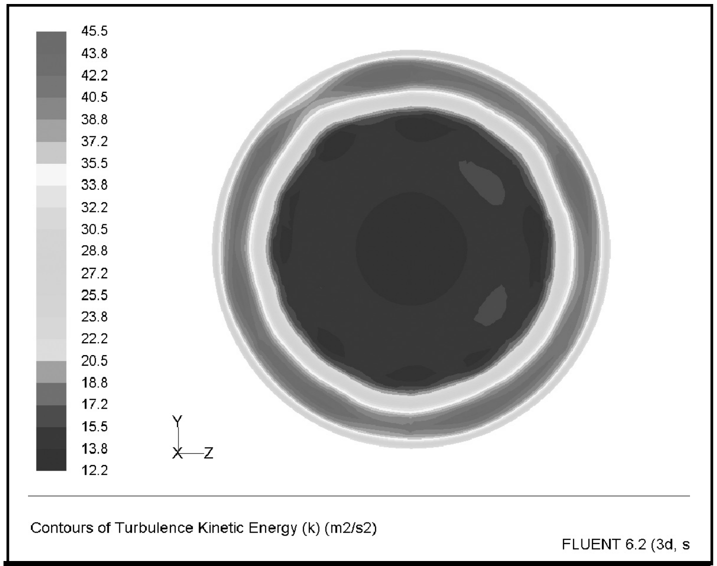

Figure 17: Turbulent kinetic energy of wiremesh substrate at 35 mm from front face

mass transfer co-efficient is increased and the performance enhanced. The wiremesh substrate enhances the mass transfer between the exhaust gas and the substrate material due to radial flow. Wiremesh substrate acts as a static mixer for the SCR catalyst application. Flow through the wiremesh substrate results in higher turbulent kinetic energy aiding better heat and mass transfer.

Wiremesh Catalyzability

The wiremesh substrate is washcoated and catalyzed with formulations for diesel oxidation catalyst (DOC), urea selective catalytic reduction (SCR) and three way catalyst (TWC). Figure 18 shows a typical catalyzed wiremesh substrate. The washcoat adhesion, washcoat pick up and catalyst performance durability can be optimized for specific applications.

Wiremesh Substrate Packaging

Canning process has a substantial impact on the quality and performance of the converter. The four basic canning methods used in converters are summarized in Table 2 and Figure 19 [19].

Closing the can using a fixed gap has the advantage of offering a fixed final dimension of the converter, which simplifies the design and welding of the cones and shells. Closing the can using a fixed force has the advantage of offering a more accurate GBD control since the dimensional tolerance influence of the shell, mat and substrate is eliminated.

The single seam converter design is usually preferred for round, trapezoidal or oval converters with low aspect ratios because it offers uniform GBD distribution. Moreover, using a single rolled and cut shell provides greater manufacturing flexibility, allowing fast design changes without the need of expensive stamping tool modifications.

Control Parameters	Single Seam	Split Shell
Fixed Gap	Stuffing	Clamshell
Fixed Force	Tourniquet	Shoebox

Table 2: Converter mounting methods

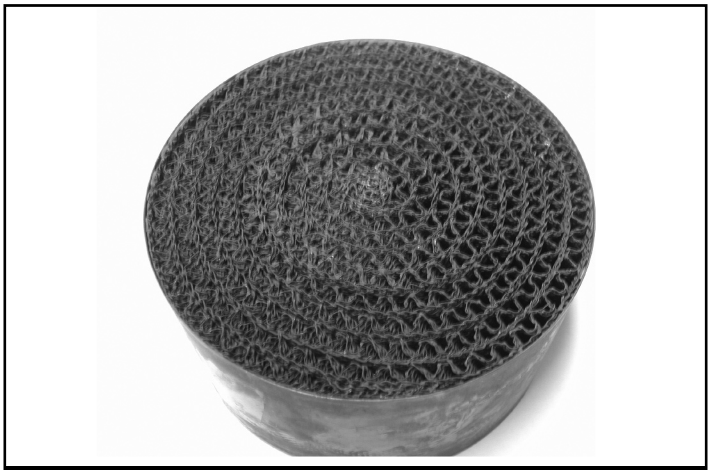

Figure 18: Catalyzed wiremesh substrate

The wiremesh substrates can be mounted in different ways depending on the application and the location. Wiremesh substrates are made with a wrapping of metallic shim/mantle of 0.5 to 1.5 mm thick. The wiremesh substrate with extended mantle (5 mm) is intended to weld the converter on the exhaust system with air gap insulation. Wiremesh substrates with flushed shim/mantle can be mounted with axially and/or radially like conventional monoliths (Figure 20). The wiremesh seal provides axial and/or radial support mounting required. The radially mounted wiremsh substrate in a split shell and the single seam shell are shown in Figures 21 and 22. The metallic shim/mantle prevents the deterioration of the mat/insulation blanket due to the exhaust gas radial flow.

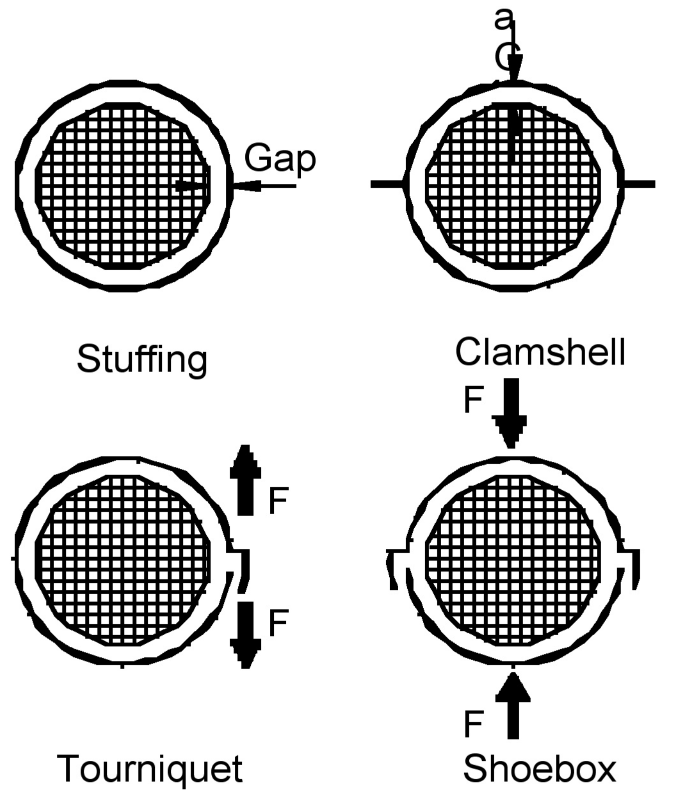

Figure 19: Monolithic substrate canning methods

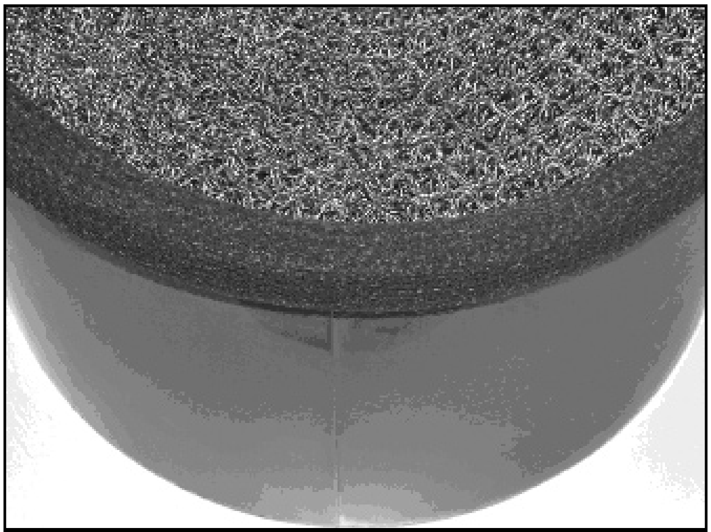

Figure 20: Wiremesh substrate with shim/mantle and L-seal.

Figure 21: Wiremesh substrate in single seam converter assembly

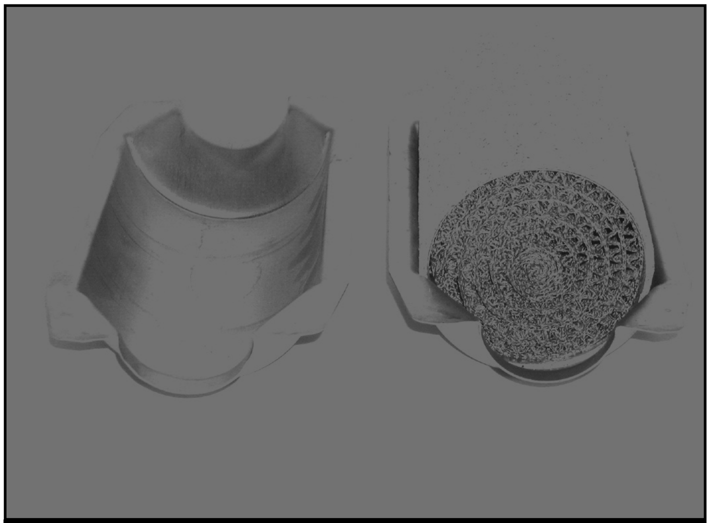

Figure 22: Wiremesh substrate in split shell assembly

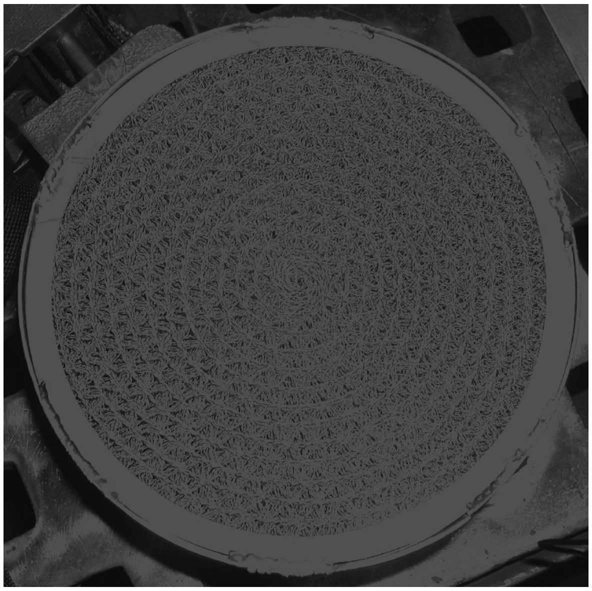

Figure 23: Wiremesh substrate with retainer ring

Figure 24: Hot vibration test set up

Substrate Durability

A hot vibration test is performed to investigate the mechanical stability of the brazed substrate in an environment of elevated temperature. Since this is a test for the brazing strength of the substrate and not for mounting, the converter is canned with two retainer rings welded on the front and back of the substrate to make the mounting robust (Figure 23). The test piece is installed on a vibration table. Figure 24 gives the picture of the hot shake test bench. Before testing begins, the initial position of the substrate was noted. The temperature through out the converter was monitored with thermocouples which include converter skin, inlet, outlet and substrate skin thermocouples. The location of the converter skin thermocouples were as follows; T/Cs are located on opposite sides of the converter shell approximately 3/8" from the leading edge of the substrate inlet face, other T/Cs are located on opposite sides of converter shell approximately 3/8" from the leading edge of the substrate outlet face, and also on opposite sides of the converter shell at the middle of the substrate. The tests were run at 1023 K and 28 G and at a vibration frequency of 185 Hz. Upon completion of the test sequence, the substrate position was recorded and a visual inspection was also performed.

Conclusion

Knitted wiremesh substrates are used in the diesel oxidation catalyst, three-way catalyst and urea selected catalytic reduction (SCR) NOx reduction catalyst.

Wiremesh materials are chosen to withstand the corrosion/erosion environments and durable at maximum continuous operating temperature and peak thermal spikes.

The wiremesh substrate is brazed using different brazing materials. The brazing process is optimized using different binders, heat treatment conditions and environments. Substrate contour geometry is optimized using different brazing materials and heat treatment conditions.

Wiremesh substrates provide radial and longitudinal flows (depth flow) and turbulent kinetic energy. The back pressure of the substrate is optimized using different material properties and knitting patterns.

The dimensional tolerances and process capability of the substrate contours are established.

The longitudinal and radial depth flows within the body of the substrate is modeled using computational fluid dynamic analysis.

The uniformity index, velocity index, back pressure and turbulent kinetic energy are characterized by fluid dynamic modeling.

The catalyzability, packagability and the physical durability of the substrates are tested using different wire types and wire mesh patterns.

Wiremsh substrates with turbulent and torturous flows could be used as a static mixer for the SCR Urea NOx reduction system to enhance performance and avoid urea crystallization.

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BIOGRAPHY

Dr. Sivanandi Rajadurai is the Vice President of ACS Industries Inc. In his role, he directs exhaust product development efforts. Dr. Sivanandi Rajadurai has been involved in Catalyst and Exhaust Products Development for the last 30 years. Dr. Sivanandi Rajadurai received his Ph.D. (1979) in Physical Chemistry (Heterogeneous Catalysis) from Indian Institute of Technology, Chennai. Rajadurai has a mix of academic and industrial experience. Dr. Rajadurai worked as Assistant Professor of Chemistry at Loyola College, Chennai from 1080-85 and Research Associate Professor at University of Notre Dame from 1985-90. Rajadurai worked as a Research Leader and Director of Research at Cummins Engine Company and Molecular Technology Corporation (1990-96), Director of Advanced Development at Tenneco Automotive (1996-02) and Director of Emissions Systems at ArvinMeritor Inc.(2002-2004) and now as Vice President of ACS Industries.

Dr.Rajadurai is a Fellow of the Society of Automotive Engineers. He is a life member of North American Catalysis Society, North American Photo Chemical Society, Instrumental Society of India, Bangladesh Chemical Society and Indian Chemical Society. He was the UNESCO representative of India on low-cost analytical studies (1983-85). He was awarded the Tenneco Innovation Award in 1998, 1999 for developing computer-aided tools for converter design and for validating low noble metal catalytic converter. He received the General Manager's Leadership Award (1998) and also the 2000 Vision Award for developing strategies for cleaner, quieter, and safer transportation. Dr.Rajadurai is a panelist of the Automotive R&D Scientists and Technologists of Indian Origin, New Delhi 2004. and Member of the Core-Group Automotive Research (CAR), India.