Bull. Mater. Sci., Vol. 25, No. 2, April 2002, pp. 127–132. © Indian Academy of Sciences.

Evaluation of thermal shock resistance of cordierite honeycombs

RATHINDRA NATH DAS*, C D MADHUSOODANA, P K PANDA[†] and KIYOSHI OKADA[‡]

Ceramic Technological Institute, Bharat Heavy Electricals Ltd., Bangalore 560 012, India
[†]Materials Science Division, National Aerospace Laboratories, Bangalore 560 017, India
[‡]Department of Metallurgy and Ceramic Sciences, Tokyo Institute of Technology, Tokyo 152-8552, Japan

MS received 24 December 2001; revised 22 February 2002

Abstract. A comparative study on thermal shock resistance (TSR) of extruded cordierite honeycombs is presented. TSR is an important property that predicts the life of these products in thermal environments used for automobile pollution control as catalytic converter or as diesel particulate filter. TSR was experimentally studied by quenching (descending test) the heated samples to water or by heating (ascending test) with an oxy-hydrogen flame along with crack detection by acoustic emission (AE) method. TSR was also calculated by using coefficient of thermal expansion (CTE), modulus of elasticity (MOE) and modulus of rupture (MOR) of the honeycomb samples. Cordierite honeycombs of 200 and 400 cpsi were used for the above study. It was observed that the trends of TSR were same for both the experimental methods as well as by calculation. The ascending test method showed lower TSR values compared to water quench method due to early detection of cracks by AE. Finite element method (FEM) was also used to evaluate the thermal stress distribution in solid cordierite using thermal shock test data. It was observed that the maximum thermal stress calculated by FEM was lower than the strength of the material; therefore, the solid cordierite did not fail during such tests.

Keywords. Cordierite; honeycomb; thermal shock resistance; acoustic emission; FEM.

1. Introduction

The exhaust gases emitted by internal combustion engines utilizing hydrocarbon fuels constitute poisonous gases e.g. CO, NO_X and hydrocarbons responsible for severe pollution of the atmosphere. These exhaust gases are conveniently purified to relatively non-toxic products like CO_2, N_2 and H_2O by employing a catalytic converter consisting of a honeycomb monolith substrate (cordierite) coated with catalyst (Pt/Pd/Rh) and placed in the path of exhaust of an automobile engine. Due to exothermic nature of the conversion reaction and repeated starting and stopping of the engine, the monolith substrate experiences thermal shock. Therefore, the substrate material should have high resistance to thermal shock, a property generally inversely proportional to the coefficient of thermal expansion. Cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5MgO$), known for its low coefficient of thermal expansion, is obviously the right candidate for the above applications (Evans *et al* 1980; Ikawa *et al* 1986). In addition, cordierite honeycombs prepared by extrusion of clay based materials exhibit further low expansion due to preferred orientation of cordierite induced during extrusion (Lachman *et al* 1981). The honeycomb structure with number of parallel channels bounded by thin ceramic walls increases the surface area with less resistance to the exhaust gas flow. Therefore, the advantages of cordierite honeycombs are (i) high surface area per unit volume for higher conversion efficiency, (ii) low thermal expansion for high thermal shock resistance and (iii) light weight, controlled porosity and pore size distribution for optimum wash coat and catalyst loading (Lachman 1986).

Enhanced performance of the cordierite honeycomb is being demanded from newer applications like diesel particulate filter (DPF). In DPF, much higher thermal shock resistance and durability is needed in order to regenerate the ceramic monolith large number of times during its life (Kitagawa *et al* 1990; Lucchini and Maschio 1995). Similarly for developing cold-start emission control systems like in close-coupled and externally heated catalytic converters in modern cars, where the monolith is mounted much nearer to the engine, improved TSR is an essential requirement (Yamamoto *et al* 1990).

Thermal shock resistance (TSR) of ceramics depend on its material properties such as σ, the modulus of rupture (MOR), E, the modulus of elasticity (MOE), α, the coefficient of thermal expansion (CTE) and ν, the Poisson's ratio and generally represented as maximum temperature gradient that the material can resist

$$\mathrm{TSR} = \Delta T = \frac{\sigma(1-\nu)}{E\cdot\alpha}. \quad (1)$$

*Author for correspondence

Evaluation of thermal shock of honeycomb structure is difficult due to the complexity of shape, anisotropy in properties and difficulty in simulating field conditions. The thermal durability of honeycombs is primarily decided by the thermal stresses during use, which depend on CTE, MOE, MOR, cell density, wall thickness and temperature distribution (Gulati 1985). TSR of honeycombs was also calculated in terms of maximum radial temperature gradient by using the empirical formula incorporating hot zone area factor (Gulati 1988):

$$\Delta T = \frac{\sigma(1-\nu)}{E \cdot \alpha \cdot A_f} \cdot L/t, \quad (2)$$

where, A_f is the ratio of central hot zone to total cross sectional area, L the length of the honeycomb cell and T the thickness of the honeycomb cell.

Conventional water quench method was also used to study TSR of honeycombs. But, this test method does not represent the actual thermal environments that the honeycombs experience during their actual use. Moreover, it is difficult to detect accurately the onset of cracks during the test due to complexity of the honeycomb structure. A solution to the above problem is to test the honeycombs in heating mode and to use AE technique to detect the onset of cracks. This technique measures the more accurate temperature at which the crack initiates. AE was widely used for crack detection of ceramic materials during thermal shock tests. Konsztowicz (1993) used to detect the damage of refractory materials during thermal shock test from the analysis of amplitude and duration of the acoustic signals. Mignard *et al* (1996) used AE to monitor the *in situ* crack growth during thermal shock test of refractory material. Evans and Linzer (1973) and Evans *et al* (1974) used AE for thermal shock study of porcelain and alumina and identified the emissions due to micro and macro cracks. Szmeja and Wala (1994) studied the process of cracking during thermal shock of magnesia and magnesia–chrome refractories by AE technique. They found that each grade of refractory material exhibits a characteristic AE pattern when subjected to thermal shock test and the pattern is generally different during heating than cooling.

In the present study, TSR was measured by flame heating–cum–AE technique, by water quench method and the results were compared with the calculated values. FEM was also used to find out thermal stress developed during testing solid cordierite and the values were correlated with the results.

2. Experimental

2.1 *Fabrication of cordierite honeycombs*

Extrusion paste for forming a single-phase cordierite was prepared by mixing talc (Rajasthan, India), kaolin clay (Kundra, India) and alumina (Indalco, Belgaum, India) to get composition of cordierite ($2MgO \cdot 2Al_2O_3 \cdot 5SiO_2$). The above powders were mixed well with water and plasticizer and were extruded in the form of honeycomb monoliths with cell density of 200 and 400 cpsi (cells per square inch) and wall thickness of 0·22 and 0·17 mm respectively. Extruded bodies were dried at 110°C and sintered at 1390°C for 4 h at the heating rate of 1°C/min. Solid cordierite bodies were also fabricated by following the above process.

2.2 *Characterization of cordierite honeycombs*

Bulk density and apparent porosity of the honeycomb samples were measured by water displacement method. Thermal expansion of extruded honeycombs was measured along the extrusion direction in the temperature range 25–800°C using a push-rod dilatometer. The modulus of rupture (MOR) at room temperature was measured to estimate the flexural strengths of solid and honeycomb samples by a 3-point bend test using an appropriate jig on a Universal testing machine (AG 5000 Shimadzu, Japan). The crush strength in 3 directions were measured using the same equipment. MOE was measured by the resonance technique in an Elastosonic Instrument (NEPL, Bangalore, India).

2.3 *Thermal shock resistance (TSR)*

2.3a *TSR by calculation*: TSR for three different types of honeycombs were calculated by using the measured values of MOR, MOE and CTE in (1). TSR was also calculated using (2) assuming 1/3 of honeycomb area is hot zone ($A_f = 0{\cdot}33$). The Poisson's ratio was taken as 0·1 in all the cases.

2.3b *TSR by quenching*: TSR was also measured by quenching from different temperatures. One inch cube samples of honeycombs were heated at 1250°C for 2 h and then quenched to water at room temperature. No visible crack was observed. Expecting the geometry effect, bigger samples of 50 mm diameter and 50 mm long cylindrical honeycombs were quenched to water bath at room temperature from a range of high temperatures (1200–1350°C) at every 25°C interval. TSR was measured from the difference of the temperature of water bath and the furnace temperature when the visible cracks appeared.

2.3c *TSR by flame heating–cum–AE technique*: The test equipment consists of a sample holder, a heat generation system, a temperature measurement system and a crack detection system. A close view of the sample with oxy-hydrogen flame heating during the thermal shock test is presented in figure 1. Circular samples (30 mm diameter and 3–5 mm thick) were heated at the centre of the top surface over a localized area of 4–5 mm diameter by an oxy-hydrogen flame torch. The central hot temperature was measured by an IR pyrometer and the cracking of the sample was detected by acoustic emission detection set-up linked to the specimen through a circular Haynes alloy wave-guide. The equipment is useful to carry out thermal shock test of materials up to a maximum temperature of 2000°C in ascending mode. The tests were conducted by placing the sample at the centre of the copper block and heating by oxy-hydrogen flame up to 1400°C that is close to the melting point of cordierite (Panda 1999; Panda *et al* 2002).

2.4 *Thermal stress distribution by FEM*

FEM method was used to model/simulate thermal stress distribution in the cordierite sample due to thermal shock test. Commercially available finite element (FE) software (NISA 7·0 of Engineering Mechanics Research Corporation, Bangalore, India) was used for simulation (NISA II user's manual 1995). The modelling was carried out by creating the geometry of the sample in the form of rectangular finite elements (60 nos, 1 mm × 0·5 mm) corresponding to an axis-symmetric plane of the sample (20 mm × 1·5 mm). The maximum temperature value of 1400°C was applied as thermal boundary to the 2 elements very close to the centre of the top surface to simulate the heating to the maximum temperature by the flame. Based on the temperatures measured earlier at different grid points with the maximum temperature of 1400°C at the centre, the temperatures of the corresponding grid points were provided for calculation. The nodal temperatures at the bottom surface were assumed at the room temperature due to the contact of the sample with water cooled copper block. Temperature and thermal stress distribution pattern during thermal shock test was generated by this method.

3. Results and discussion

The measured properties of honeycombs and solid samples prepared from the same batch are shown in table 1. The variations of mechanical properties are as expected dependent on the design parameters such as pitch, thickness and open frontal area. Bulk densities are in the range of 1·79 to 2·10 g/cc and apparent porosity varies between 25 and 31%. This small variation might result from the variation in extrusion pressure when extruded through

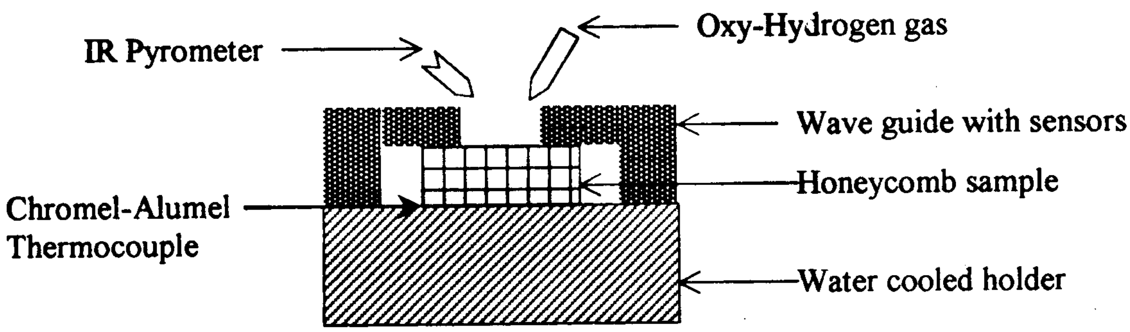

Figure 1. A close view of the sample with oxy-hydrogen flame heating during the thermal shock test.

Table 1. Mechanical and thermal properties of solid and honeycomb cordierites.

		Honeycomb type		
	Units	200 cpsi	400 cpsi	Solid sample
Pitch	mm	1·73	1·28	
Wall thickness	mm	0·22	0·17	
Modulus of elasticity (MOE)	(GPa)	14	16	64
Modulus of rupture (MOR)	kg/cm^2	79	81	511
MOR at 900°C	kg/cm^2	75	86	
Crush strength: *a*-axis	kg/cm^2	378	383	1533
Crush strength : *b*-axis	kg/cm^2	49	63	
Crush strength : *c*-axis	kg/cm^2	4·3	5·9	
Coeff. of thermal expansion (CTE)	$\times 10^{-6}$/°C	1·43	1·35	1·5
Bulk density	g/cc	1·95	1·79	2·10
Apparent porosity	%	26	31	25

different configuration of dies. This range of porosity (25 to 31%) is desirable to get good washcoating for catalyst application. The porosity also reduces the thermal expansion and increases the thermal shock resistance, but decreases the mechanical strength compared to dense cordierite. Therefore, the above porosity range of the sample is optimized for the present study. Little variation in the thermal expansion of samples of the two types (200 and 400 cpsi design) prepared from the same batch of material is attributed to the variation in the degree of preferred orientation of cordierite along the extrusion direction (Madhusoodana *et al* 2001). The orientation of cordierite in honeycomb is imparted during extrusion through honeycomb die that consists of narrow holes and thin slots. Cordierite forming materials contain platy clay and talc which gets aligned due to rheological shear.

The TSR values obtained for different honeycombs using different methods are presented in table 2. Using water quench method, it was observed that TSR was around 1250°C for all types of honeycombs tested.

A typical temperature profile and acoustic emission profile generated during thermal shock test of a honeycomb is presented in figure 2. The experiment was repeated for both types of honeycombs. TSR was calculated from these minimum temperature of crack initiation by deducting cold end temperature which was maintained at 30°C. It was observed that the solid cordierite did not crack even after heating to 1400°C while honeycomb samples were cracked in the temperature range of 1101–1037°C. This shows that the thermal stress generated in the solid sample is lesser than the flexural strength (511 kg/cm^2). Whereas due to the lower flexural strength (79–81 kg/cm^2) of the honeycomb samples, they are able to withstand the thermal stresses only up to 1101–1037°C.

From table 2, it could be noted that the values obtained in AE method are lower than those observed by water quench method. This is because of early detection of cracks in AE method.

By water quenching method, it was also difficult to differentiate the TSR for different types of honeycombs. Whereas, it was possible to differentiate the TSR values by flame heating method due to the immediate detection of the cracks by AE.

Table 2. Thermal shock resistance (TSR) values by different methods (temperature difference = ΔT values are given in °C).

	Honeycomb type (cpsi)	
	200	400
Water quenching	1250	1250
Acoustic emission	1101	1037
Calculated using (2)	1107	1060
Calculated using (1)	355	338
Calculated using (3)	1066	1015

TSR values calculated using (2) are comparable with observed values using AE and the trend is also found to be the same i.e. TSR of 200 cpsi is higher than that of 400 cpsi. The values obtained using (1) are 355 and 338°C, which are much lower than that of water quench (1250°C) and flame heating (1037–1101°C) methods. The lower TSR values by calculation from (1) indicate the need to use some correction factor such as area factor, A_f as suggested in (2). Equation (1), which uses properties of the bulk cordierite material, can be modified by using the thermo-mechanical properties of cordierite honeycomb structures as follows

$$\Delta T_C = \frac{\sigma_h \cdot (1-\nu)}{E_h \cdot \alpha_h \cdot A_f}, \quad (3)$$

where σ_h, E_h and α_h are the MOR, MOE and CTE of the corresponding honeycomb structure, respectively.

The A_f value is dependent on the temperature distribution on the honeycomb face during the test. Considering the central 1/3 of the honeycomb cross-section area is hot zone as assumed in (2), the same A_f value (0·33) is used in (3) for comparing both methods of calculations. TSR values obtained are 1066 and 1015 for 200 and 400 cpsi honeycomb, respectively. These values are comparable to the calculated values using (2) and values obtained from the AE method.

The trend of TSR values observed was the same for all the evaluation methods studied. In all cases 200 cpsi honeycombs had higher TSR values than 400 cpsi honey-

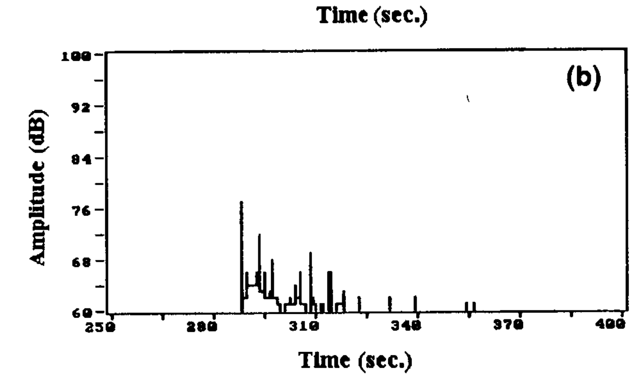

Figure 2. (**a**) Temperature profile of the hot zone and (**b**) acoustic emission signals of the cordierite honeycomb (400 cpsi) recorded during thermal shock test.

Table 3. Thermo-mechanical properties of cordierite used for FEM modelling.

Thermo-mechanical properties	Units	Test sample
Density (ρ)	kg/m^3	2100
Thermal conductivity (K)	W/m °C	2·6
Coefficient of thermal expansion (α)	°C^{-1}	$1{\cdot}5 \times 10^{-6}$
Young's modulus (E)	GPa	64
Heat capacity (C_P)	J/kg C	920
Poisson's ratio (v)	–	0·1
Diameter of the sample	mm	40
Thickness of sample	mm	1·5

combs, even though CTE of 400 cpsi is slightly lower than 200 cpsi. This indicates that the honeycomb design parameters and mechanical properties have more significant influence on TSR.

The FEM modelling generated the temperature distribution as well as thermal stress profiles. The thermo-mechanical properties used for the FEM modelling are presented in table 3. The temperature and thermal stress profile for the solid cordierite samples are shown in figure 3. The maximum thermal stress obtained from analysis is 336 kg/cm^2 which is less than the fracture strength

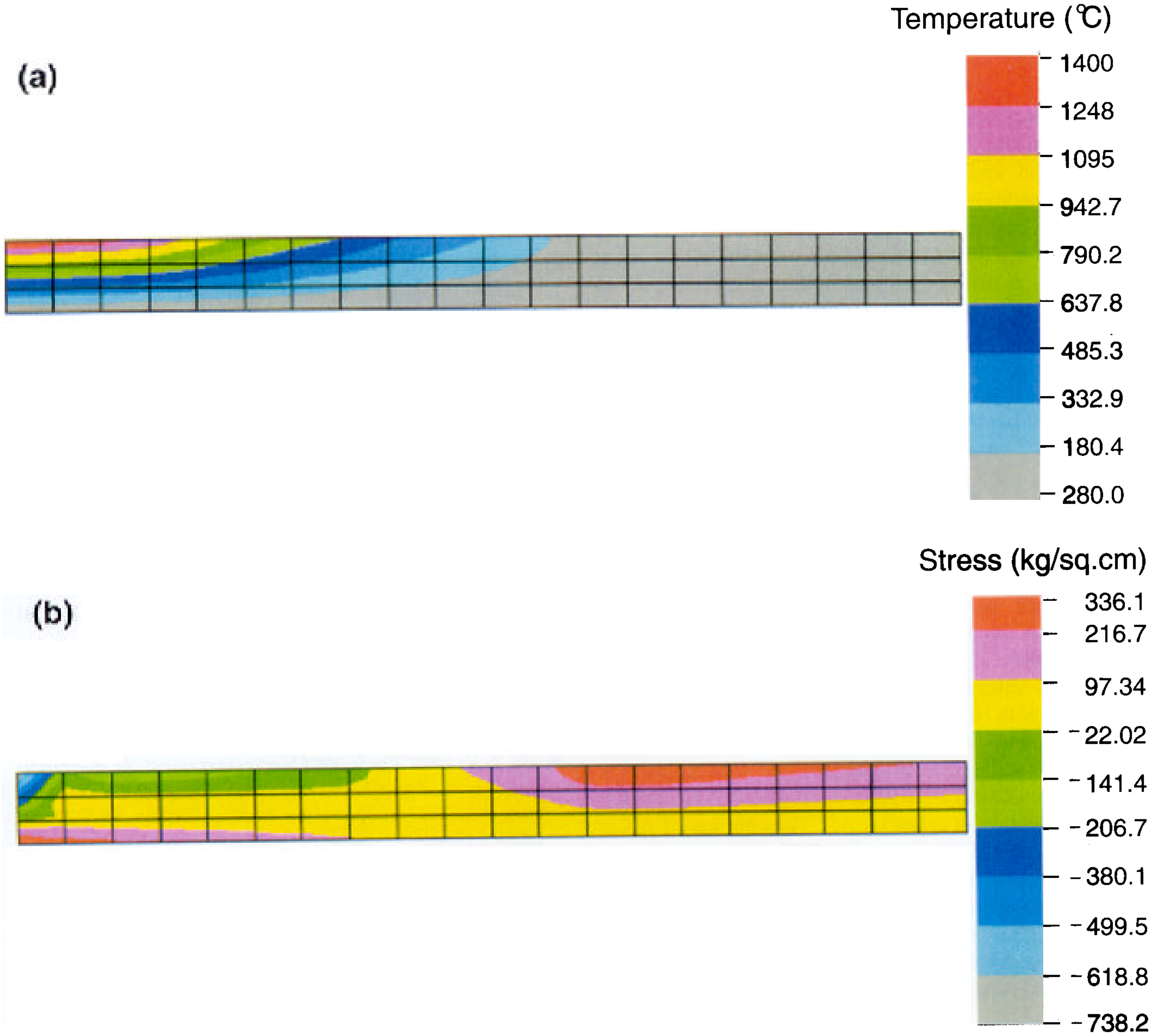

Figure 3. (**a**) Temperature and (**b**) thermal stress distribution in an axis-symmetric plane of solid cordierite body corresponding to a maximum temperature of 1400°C at the centre of top surface as calculated by FEM.

of the material i.e. 511 kg/cm^2. The maximum compressive stress observed is 738 kg/cm^2 which is much lesser than compressive strength of the sample (1533 kg/cm^2). Thus the material can withstand the thermal stresses without failure. This substantiates the observation that during flame heating, the acoustic emission was not detected up to 1400°C, indicating that no crack generated by this thermal shock. As can be observed from the stress plot that maximum tensile stress was concentrated on one edge away from the heating zone and next higher stress concentration on the opposite side of hot zone. Thus, by knowing the cordierite material properties and using FEM analysis, the thermal stresses and material failure temperature can be estimated. Similar analysis can be carried out for cordierite honeycomb structure.

4. Conclusions

Thermal shock resistance (TSR) of extruded cordierite honeycombs was studied using a novel acoustic emission (AE) method and compared with calculated and conventional methods. Cordierite honeycombs of 200 and 400 cpsi were fabricated by extrusion and were characterized for CTE, MOE and MOR. The values were used for calculation of TSR using two equations. In the developed flame heating–cum–acoustic emission set up, the cracks were induced during ascending thermal shock by oxy-hydrogen flame heating. These cracks were detected by AE. Cracks were detected during heating, at 1101°C and 1037°C for 200 and 400 cpsi honeycombs, respectively. Thus 200 cpsi honeycomb showed higher TSR values than 400 cpsi honeycomb, even though no significant variation was found in CTE between these two honeycombs. The variation of TSR is mainly attributed to the variation in MOE and MOR. These values depend on design parameters such as cell density, wall thickness and open frontal area. The influence of honeycomb design parameters is thus found to be significant for TSR values.

The TSR values obtained from AE measurement was compared with that of calculated and water quench methods. Trend observed was the same for all the types of evaluations. In all tested samples, AE method showed lower TSR values compared to water quench method due to early detection of crack by AE. Thus the use of AE is suggested as more appropriate for TSR evaluation.

Finite element method (FEM) was used to evaluate the thermal stress distribution of solid cordierite during thermal shock test. The maximum stress observed by FEM was lower than flexural strength of the material. This supports experimental observation of the sample, which did not show failure during flame heating up to melting point. Thus combination of acoustic emission and FEM approach is a good tool for predicting and more accurately evaluating thermal shock resistance of honeycomb structures.

Acknowledgements

(RND) and (CDM) thank the Management of CBU, BHEL, for their encouragement during this research. Thanks are also due to Prof. A M Umarji, MRC, IISc, Bangalore, for his help in thermal expansion studies. The Japan Society for the Promotion of Science, is also gratefully acknowledged for the RONPAKU fellowship for (RND). We also thank Dr T S Kannan for the encouragement and Mr A Cheluvraju for the assistance during the thermal shock test.

References

Evans A G and Linzer M 1973 *J. Am. Ceram. Soc.* **56** 575

Evans A G, Linzer M and Russel L R 1974 *Mater. Sci. Engg.* **15** 253

Evans D L, Fisscher G R, Geiger G R and Martin F W 1980 *J. Am. Ceram. Soc.* **63** 629

Gulati S T 1985 SAE Technical Paper **850130**

Gulati S T 1988 SAE Technical Paper **881685**

Ikawa H, Otagiri T, Imai O, Suzuki M, Urabe K and Udagawa S 1986 *J. Am. Ceram. Soc.* **69** 6

Kitagawa J, Toshihiko H and Mikio M 1990 *SAE technical paper series* (Warrendale, PA, USA: SAE) **900113** p. 33

Konstowicz K J 1993 *Thermal shock and thermal fatigue behaviour of advanced ceramics* (eds) G A Schneider and G Petzow (Netherlands: Kluwer Academic Publishers) pp 429–441

Lachman I M 1986 *Sprechsaal* **119** 1116

Lachman I M, Bagley R D and Lewis R M 1981 *Am. Ceram. Bull.* **60** 202

Lucchini E and Maschio S 1995 *Mater. Sci. & Technol.* **11** 605

Madhusoodana C D, Das R N, Umarji A M and Okada K 2001 *Ceramic Trans. Am. Ceram. Soc.* **112** 539

Mignard F, Olagnan C, Saadaoui M and Fantozzi G 1996 *J. Mater. Sci.* **31** 2437

NISA II user's manual 1995 Version 6·0, Engineering Mechanics Research Corporation, Michigan 48099, USA

Panda P K 1999 *Etude de L'endommagement de Materiaux Ceramiques Par Choc Thermiques Ascendant: Comportement a la Fatigue Thermique Et Modelisation*, Ph D thesis, INSA de Lyon

Panda P K, Kannan T S, Dubois J, Olagnon C and Fontozzi G 2002 *Sci. & Tech. Adv. Mater.* (in press)

Szmeja B J and Wala T B 1994 *Ceram. Int.* **20** 367

Yamamoto H, Hideaki H, Kitagawa J and Machida M 1990 *SAE technical paper series* (Warrendale, PA, USA: SAE) **900614**