Available online at www.sciencedirect.com

ScienceDirect

CERAMICS INTERNATIONAL

Ceramics International 36 (2010) 299–306

www.elsevier.com/locate/ceramint

Layered structures in ceramic nozzles for improved erosion wear resistance in industrial coal-water-slurry boilers

Deng Jianxin[a,*], Yun Dongling[a], Zhou Houmin[b], Tan Yuanqiang[b]

[a] *School of Mechanical Engineering, Shandong University, Jinan 250061, Shandong Province, PR China*

[b] *School of Mechanical Engineering, Xiangtan University, Xiangtan 411105, Hunan Province, PR China*

Received 25 March 2009; received in revised form 22 June 2009; accepted 3 August 2009

Available online 22 September 2009

Abstract

The nozzle is the most critical part in the coal-water-slurry (CWS) boilers. Ceramics being highly wear resistant have great potential as CWS nozzle materials. In this paper, Al_2O_3/(W,Ti)C + Al_2O_3/TiC layered ceramics (LN1, LN2, and LN3) with different thickness ratios among constituent layers were developed to be used as nozzles in CWS boilers. CWS burning tests in a boiler with these nozzles were carried out. The erosion wear behavior of the layered nozzles was investigated and compared with an unstressed reference nozzle (N5). Results showed that the layered ceramic nozzles exhibited an apparent increase in erosion wear resistance over the unstressed reference one. The mechanisms responsible were found to be that layered structure in the CWS nozzles can improve the hardness and fracture toughness of the external layer, and reduce the temperature gradients and the thermal stresses at the exit of the nozzle during CWS burning processes. It is suggested that layered structures in ceramic nozzles is an effective way to improve the erosion wear resistance over the stress-free ceramic nozzles in industrial CWS boilers.

Keywords: Nozzles; Ceramic materials; Residual stress; Layered materials; Slurry erosion

1. Introduction

The nozzle is the most critical part in a coal-water-slurry (CWS) burning system. During CWS burning processes, the nozzle is eroded continuously by the abrasive action of CWS, the working environmental temperature of nozzle can reach up to 1000 °C, and there is a very high temperature gradient inside nozzle [1,2]. Therefore, the nozzles in the CWS boiler must have high hardness, and good erosion and thermal shock resistance. In the author's previous studies [1,2], several ceramic composites were produced by hot-pressing for use in CWS nozzles. Detailed observations and analyses of the nozzle wear surface have revealed that the primary wear mechanisms of the CWS ceramic nozzle exhibited thermal shock damage with chipping at the nozzle exit. Greater temperature gradient and higher thermal stress were found to be the main reason that caused the failure of the nozzle exit.

Layered structures constituted by alternate layers with different compositions can be properly designed to induce a surface compressive residual stress [3–6]. The basic idea is to couple material layers with different thermal expansion coefficients (CTE) so that residual stresses arise during cooling from the sintering temperature. Residual stresses arise from a mismatch between the CTE, sintering rates and elastic constants of the constituent phases and neighbouring layers, and the residual stress field depends on the geometry of the layered structure and on the thickness ratio among layers [7–9]. The design of layered ceramics has been proved to be a viable strategy to obtain significant increases of the fracture resistance, wear resistance, and tribological properties [10–12].

The idea of layered structures was first introduced to the design of sand-blasting ceramic nozzles so as to form compressive residual stresses at the nozzle entry (or exit) region in fabrication process, which may partially counteract the tensile stresses resulting from external loadings [13–15]. Results showed that layered structures in ceramic nozzle can induce an excess residual stress in the nozzle during fabrication,

* Corresponding author at: School of Mechanical Engineering, Shandong University, Jinan 250061, Shandong Province, PR China.
Tel.: +86 531 88392047.
E-mail address: jxdeng@sdu.edu.cn (J. Deng).

0272-8842/$36.00
doi:10.1016/j.ceramint.2009.09.003

and this residual stress is compressive whatever the sintering temperature. This kind of compressive residual stress can result in an improved erosion wear resistance of the layered ceramic nozzle compared with the homogeneous stress-free one in sand-blasting processes [13–15].

Al_2O_3/(W,Ti)C and Al_2O_3/TiC ceramics are widely used in industrial applications such as cutting tools and dies [16–18], they both have high hardness and wear resistance. These two materials have different thermal expansion coefficients; and different shrinkage during sintering. The thermal expansion coefficient (CTE) of Al_2O_3/(W,Ti)C is $7.2 \times 10^{-6}\ K^{-1}$, and the CTE of Al_2O_3/TiC is $8.0 \times 10^{-6}\ K^{-1}$ [16–18]. These differences are sufficient to induce residual stresses in the laminated structures, and compressive residual stresses are induced in the layers with lower CTE. As for Al_2O_3/(W,Ti)C + Al_2O_3/TiC layered ceramic material, the CTE of the surface layer (Al_2O_3/(W,Ti)C) is lower than that of the center layer (Al_2O_3/TiC), so compressive residual stresses will be formed in the surface layer of the layered materials during fabrication. In the present study, Al_2O_3/(W,Ti)C + Al_2O_3/TiC layered ceramics with different thickness ratios among constituent layers were produced to be used as the CWS nozzles. The mechanical properties at the surface layers of the layered materials were measured, and the micro-structure was examined. The wear behaviors of the layered ceramic nozzles were investigated and compared to an unstressed reference nozzle. The purpose was to characterize the erosion wear of the layered ceramic nozzle in industrial CWS boilers.

2. Materials and experimental procedures

2.1. Preparation of the Al_2O_3/(W,Ti)C + Al_2O_3/TiC layered ceramic nozzles

The dimension and compositional distribution of the layered ceramic nozzles with different thickness ratios among constituent layers are shown in Fig. 1. These nozzles possess a three-layer symmetrical structure. The composition at the nozzle entry or exit is Al_2O_3/45 vol.%(W,Ti)C, while the composition at the nozzle center area is Al_2O_3/55 vol.%TiC. Three layered nozzles with different thickness ratio p ($p = A_1/A_2$) among constituent layers were produced. The layered nozzles with the thickness ratio of 0.2, 0.5, and 1 are named LN1, LN2, and LN3, respectively (see Fig. 1).

The starting powders used to fabricate the layered nozzles are listed in Table 1 with their physical properties. Composite powders of different mixture ratios were prepared by wet ball milling in alcohol with cemented carbide balls for 80 h. Following drying, the composite powders with different mixture ratios were layered into a graphite mould. The sample was then hot-pressed in flowing nitrogen for 15 min at 1700 °C temperature with 30 MPa pressure. For the purpose of comparison, a homogeneous stress-free ceramic nozzle was also manufactured by hot-pressing. This stress-free nozzle made from Al_2O_3/45 vol.%(W,Ti)C is named N5.

2.2. Hardness and fracture toughness measurements at surface layer of the layered nozzle materials

Fracture toughness measurement was performed using indentation method at the nozzle external layer (entry or exit) in a hardness tester (MH 6) using the formula proposed by Cook and Lawn [19]. Hardness measurements were performed by placing Vickers indentations on external layer of the layered nozzle material. The indentation load was 200 N and a minimum of five indentations were tested. The Vickers hardness (GPa) is given by:

$$Hv = 1.8544 \frac{P}{(2a)^2} \tag{1}$$

where P is the indentation load (N), $2a$ is the catercorner length (μm) due to indentation.

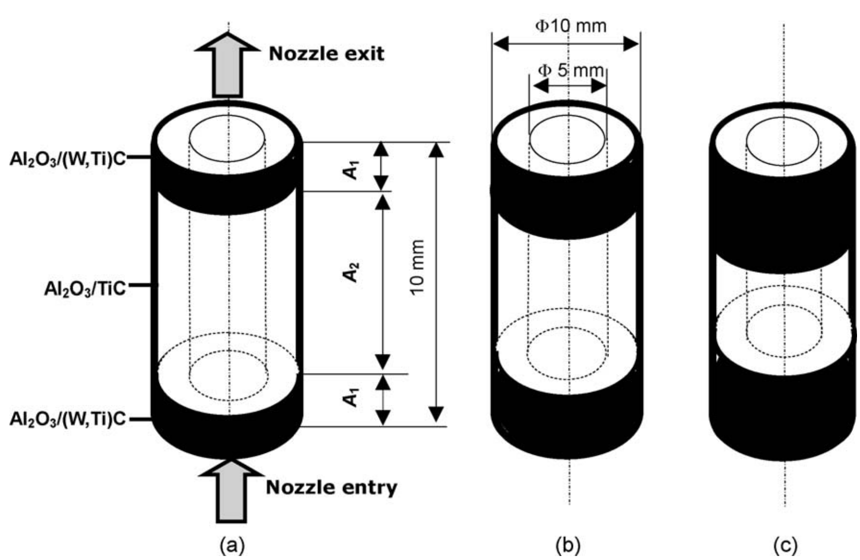

Fig. 1. Compositional distribution of the layered ceramic nozzles with different thickness ratio among constituent layers: (a) N1 nozzle ($p = A_1/A_2 = 0.2$), (b) N2 nozzle ($p = 0.5$), and (c) N3 nozzle ($p = 1$).

Table 1
Physical properties of Al_2O_3, TiC and (W,Ti)C.

Starting powder	Density (g/cm^3)	Young's modulus (GPa)	Thermal expansion coefficient (10^{-6} K^{-1})	Thermal conductivity W/(m K)	Poisson's ratio	Particle size (μm)	Purity (%)	Manufacture
Al_2O_3	3.98	380	8.0	30.2	0.27	1–2	>99	Beijing Antai Advanced Tech. and Mater. Co., Ltd.
TiC	4.93	500	7.4	24.3	0.20	1–2	>99	Zhuzhou cemented carbide works
(W,Ti)C	9.56	480	8.5	21.4	0.25	1–2	>99	Zhuzhou cemented carbide works

Table 2
Properties of the coal-water-slurry.

Consistency (%)	Quantity of heat (MJ/kg)	Ash (%)	Sulphur (%)	Volatility (%)	Adhesiveness (MPa s)	Grit number (μm)
65 ± 2	18.81–20.48	<12	<0.8	>15	1000–2500	40–80

2.3. Coal-water-slurry burning tests

Coal-water-slurry burning tests were conducted with an industrial CWS boiler. The properties of the CWS used in this study are listed in Table 2. The schematic diagram and the photo of the boiler are shown in Fig. 2. The CWS drawn by pump passed through pipeline, accelerated and mixed in the spray-gun (Fig. 3) by gas stream (commonly compressed air). The compressed air pressure was set at 0.4 MPa, and CWS pressure was set at 0.2 MPa. The nozzles have an internal diameter 5.0 mm, external diameter 12.0 mm, and length 10.0 mm. Photo of a layered ceramic nozzle is shown in Fig. 4.

The mass loss of the worn layered nozzles was measured with an accurate electric balance (minimum 0.01 mg). The erosion rates (W) of the layered nozzles are defined as the nozzle mass loss (m_1) divided by the nozzle density (d) times the burn mass of CWS (m_2).

$$W = \frac{m_1}{d \cdot m_2} \quad (2)$$

where W has the units of volume loss per unit mass of CWS (mm^3/kg).

Finite element method (FEM) was used as a means of numerically evaluating the temperature gradient and thermal stresses inside the CWS nozzle. The eroded wall surfaces of the CWS nozzles were examined by scanning electron microscopy (SEM).

3. Results and discussion

3.1. Surface layer properties and microstructural characterization of the layered nozzle materials

The results of hardness and fracture toughness of the layered nozzles with different thickness ratios are presented in Table 3. It is indicated that the hardness and fracture toughness decrease gradually from LN1 nozzle to N5 nozzle. The external layer of the nozzle with low thickness ratio shows the highest hardness and fracture toughness. By comparison with the stress-free nozzle (N5), the hardness at the external layer of the layered nozzles is much more improved, and rose from 19.9 GPa for N5 stress-free nozzle to 21.6 GPa for LN1 layered nozzle, representing a maximum increase of 1.7 GPa. The fracture toughness rose from 4.9 MPa $m^{1/2}$ for N5 stress-free nozzle to 9.8 MPa $m^{1/2}$ for LN1 layered nozzle, representing a maximum increase of 4.9 MPa $m^{1/2}$. As reported in Ref. [20], layered structures in ceramic material can induce an excess compressive residual stress at the nozzle external layer during fabrication. These compressive residual stresses are beneficial for the increase of hardness and the fracture toughness at the

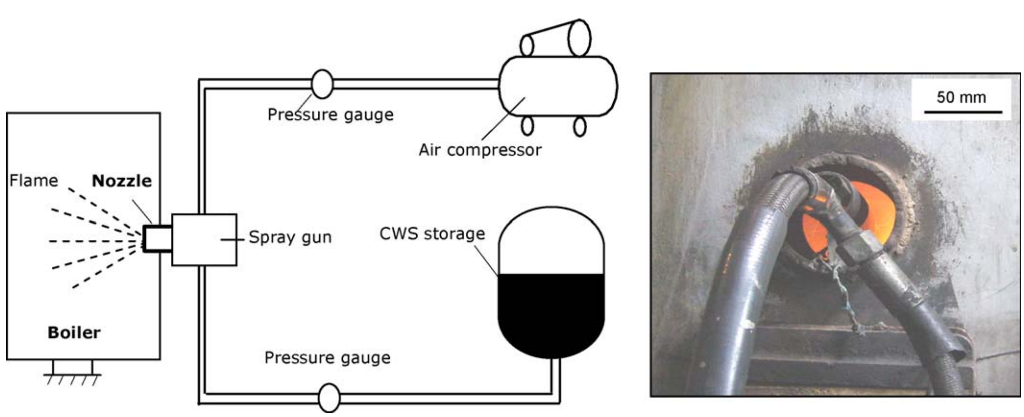

Fig. 2. Schematic diagram and photo of the industry coal-water-slurry boiler.

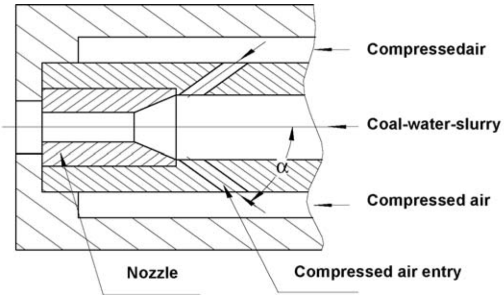

Fig. 3. Structure of the spray-gun.

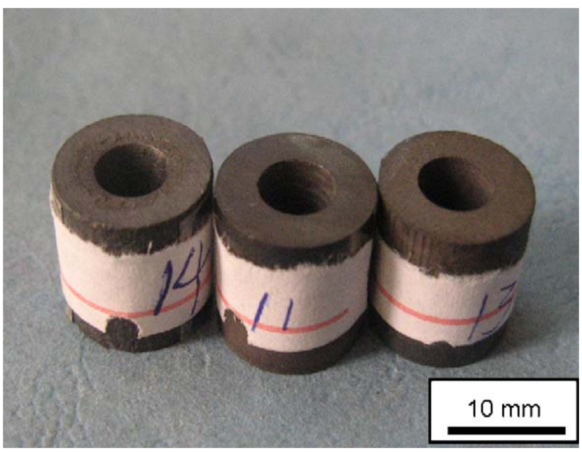

Fig. 4. Photo of the layered ceramic nozzles.

external layer, which is also reported by other researchers [4–6].

The SEM micrographs of the cross-section surface of the LN3 layered nozzle material are shown in Fig. 5. A layered structure can be clearly seen.

3.2. Hole diameter variation and erosion rate of the layered ceramic nozzles

Fig. 6 shows the comparison of hole diameter variation of the CWS nozzles after 120 h operation. It is indicated that the

Table 3
Hardness and fracture toughness at the external layer (entry or exit) of the layered ceramic nozzles with different thickness ratios among constituent layers.

Code name	Thickness ratio $P = A_1/A_2$	Hardness (GPa)	Fracture toughness (MPa $m^{1/2}$)
LN1 layered nozzle	0.2	21.6 ± 1.2	9.8 ± 0.6
LN2 layered nozzle	0.5	21.1 ± 1.1	8.4 ± 0.5
LN3 layered nozzle	1	20.9 ± 1.1	7.3 ± 0.5
N5 stress-free nozzle		19.9 ± 1.0	4.9 ± 0.5

hole diameter variation of N5 stress-free nozzle is much higher than those of the LN1, LN2, and LN3 layered nozzles under the same test conditions, and the hole diameter variation was also influenced by the thickness ratio among constituent layers. The LN1 nozzle with thickness ratio of 0.2 between adjacent layers had the smallest hole diameter variation among the layered nozzles.

The comparison of the erosion rates of the nozzles in CWS burning processes is shown in Fig. 7. It is obvious that the erosion rates of N5 stress-free nozzles are much higher than that of the layered nozzles, and the erosion rates of the nozzles from low to high is LN1, LN2, LN3, and N5 in turn. The LN1 layered nozzle with thickness ratio of 0.2 exhibited the highest erosion wear resistance.

3.3. Worn surface of the layered nozzles

The exit hole profiles of the CWS nozzles after 120 h operation are shown in Fig. 8. It was found that the exit of N5 stress-free nozzles exhibits fracture marks. For determination of erosion mechanisms, the worn CWS nozzles were sectioned axially. Fig. 9 shows the wall surface profiles of the worn CWS nozzles after 120 h operation. It is obvious that the hole diameter of N5 stress-free nozzles enlarges greatly compared with that of the layered nozzles.

Characteristic SEM pictures taken on the nozzle wall surface of the LN1 and N5 nozzles after 120 h operation are showed in Fig. 10. The wall surface of the LN1 layered CWS nozzle is

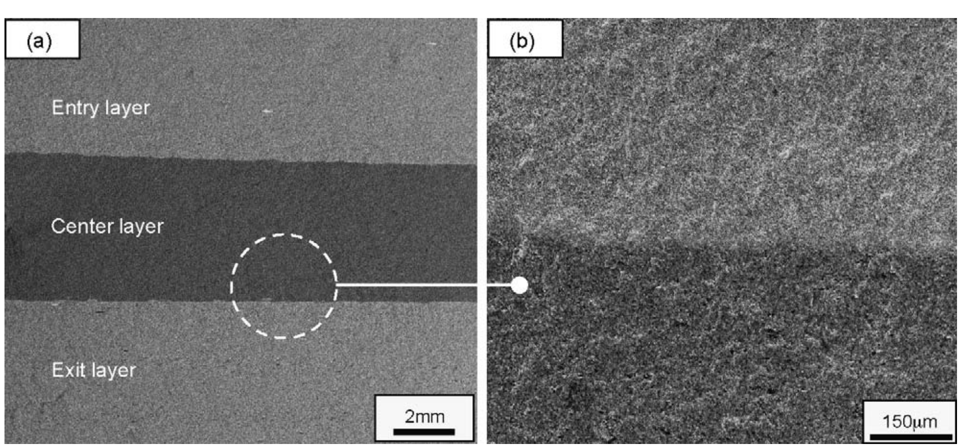

Fig. 5. SEM micrographs of the cross-section surface of the LN3 layered nozzle material.

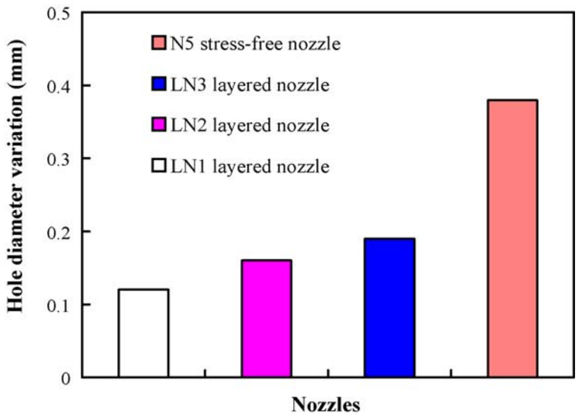

Fig. 6. Comparison of the hole diameter variation of the LN1, LN2, LN3 layered nozzles and N5 stress-free nozzle.

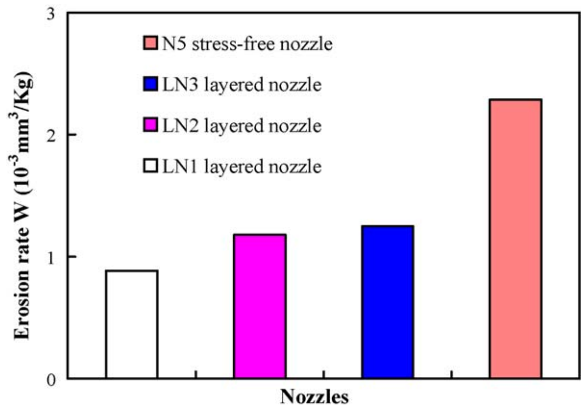

Fig. 7. Comparison of the erosion rates of the LN1, LN2, LN3 layered nozzles and N5 stress-free nozzle.

relatively smooth in contrast to that of the N5 stress-free nozzle. From Fig. 10d, the micro-structure can be seen clearly. In this structure, the ‘‘white’’ phase with clear contrast is (W,Ti)C, and the grey phase is Al_2O_3. No sign of plowing was found both at the wall surface of LN1 nozzle.

3.4. Discussion

When the erosive particles hit the target at low angles such as the nozzle wall surface in CWS burning processes, most of the CWS particles traveled almost parallel to the nozzle wall, and

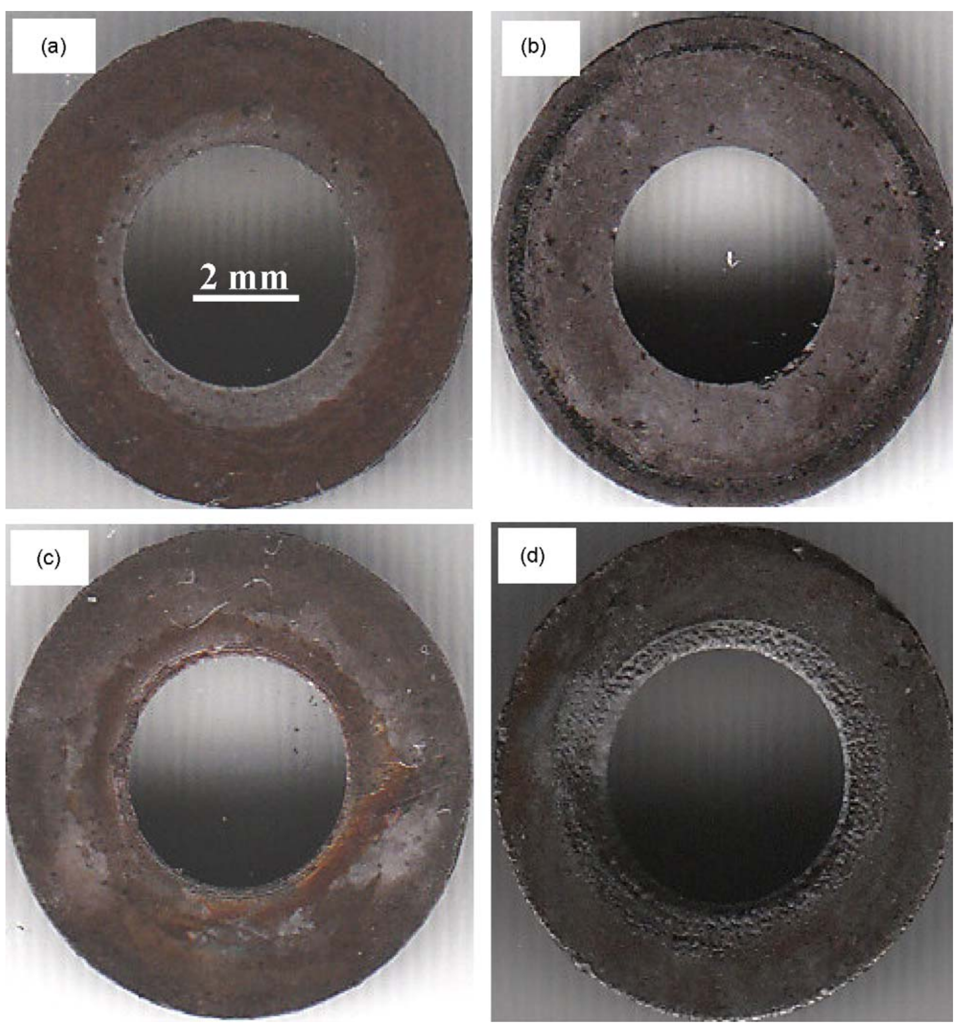

Fig. 8. Exit hole profiles of the worn CWS nozzles after 120 h operation: (a) LN1 layered nozzle, (b) LN2 layered nozzle, (c) LN3 layered nozzle, and (d) N5 stress-free nozzle.

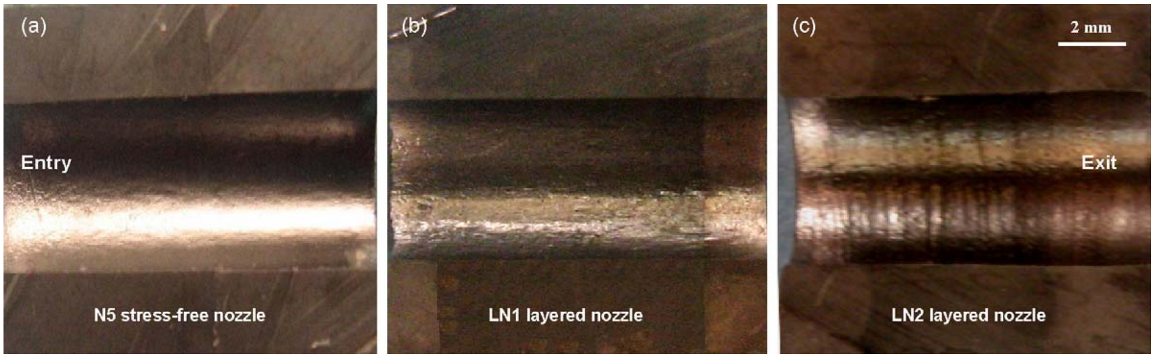

Fig. 9. Wall surface profiles of the worn CWS nozzles after 120 h operation: (a) N5 stress-free nozzle, (b) LN1 layered nozzle, and (c) LN3 layered nozzle.

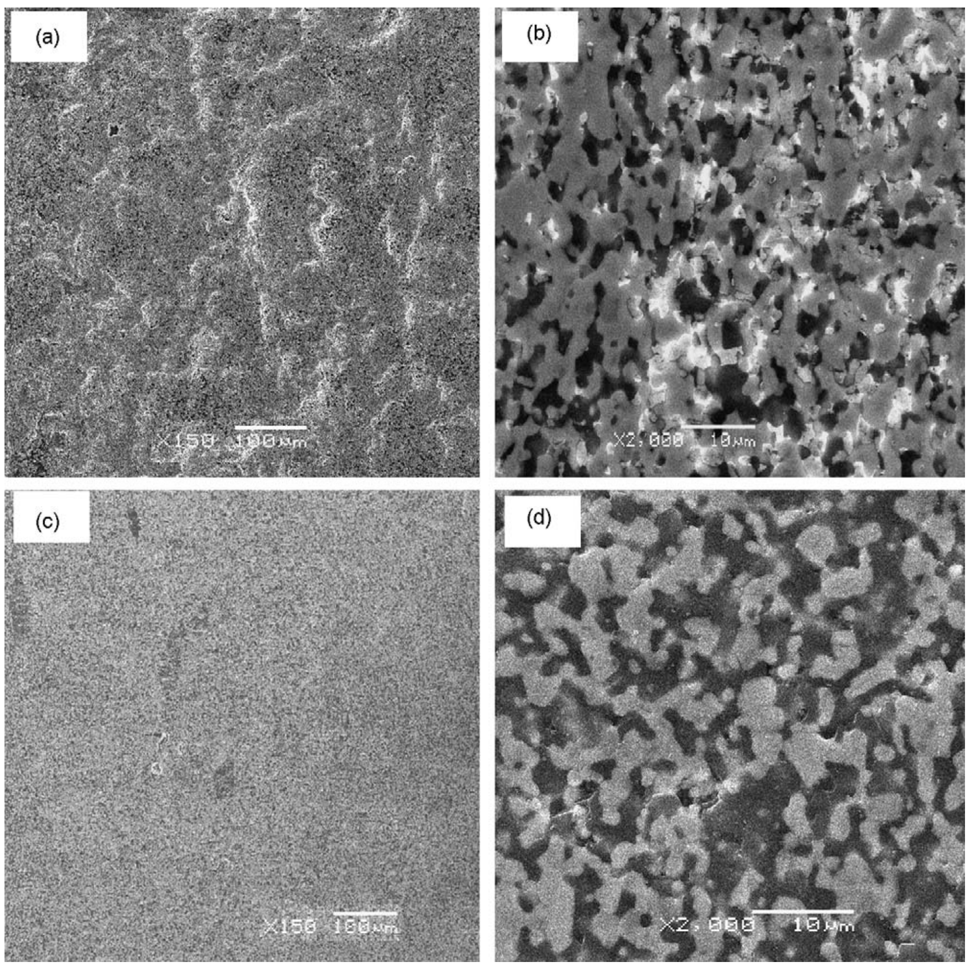

Fig. 10. SEM micrographs of the wall surface of the CWS nozzles after 120 h operation: (a) wall surface of N5 stress-free nozzle, (b) enlarged SEM micrograph corresponding to (a), (c) wall surface of LN1 layered nozzle, and (d) enlarged SEM micrograph corresponding to (c).

impact the wall surface of the nozzle at low angles. As the erodent particles in CWS were much softer than the ceramic nozzles, sliding abrasive particles in the CWS can act as a polishing medium on the nozzle wall surface. Therefore, the eroded wall surface of the nozzle is very smooth (see Fig. 10), and the hardness of the CWS nozzles plays an important role with respect to their erosive wear. The ceramic nozzles with high hardness had smaller erosion rates. The high erosion resistance of the layered nozzles may be explained by their high surface hardness (see Table 3).

The damage of ceramics subjected to temperature gradient environments is another major limiting factor in relation to service requirements and lifetime performance. Repetitive thermal shock also results in thermal fatigue, which has a significant effect on the life of the CWS nozzles. For determination of the thermal stresses inside the nozzle when

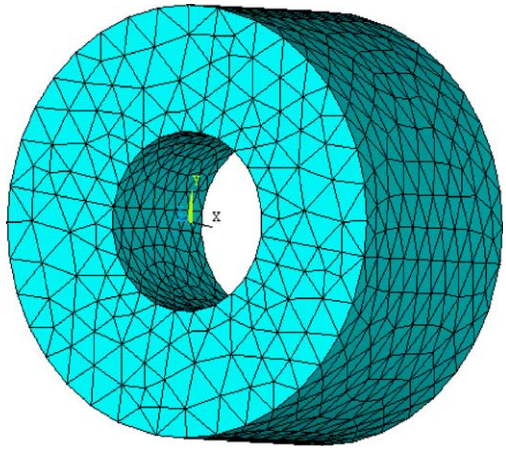

Fig. 11. Finite element method gridding model of the nozzle.

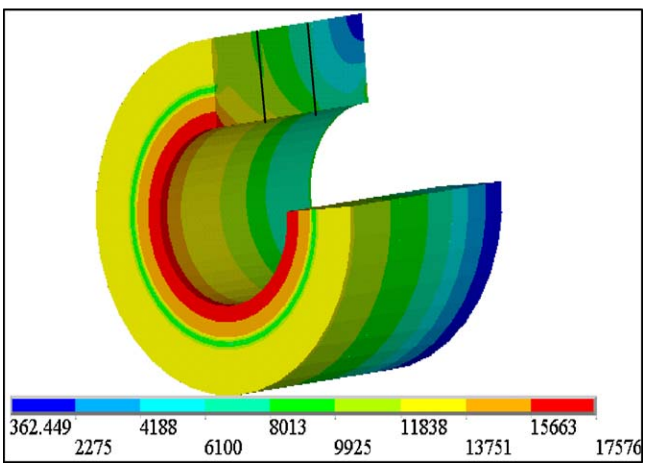

Fig. 12. Temperature gradients in the LN3 layered nozzle (°C/m).

used in a CWS boiler, three-dimensional finite element method (FEM) was used. Owing to the symmetry, an axisymmetric calculation was preferred and steady state boundary conditions were invoked. Details on the FEM model approach and the boundary conditions employed are described in Refs. [1,21]. The materials constants are listed in Table 1. Fig. 11 shows the FEM gridding model of the nozzle.

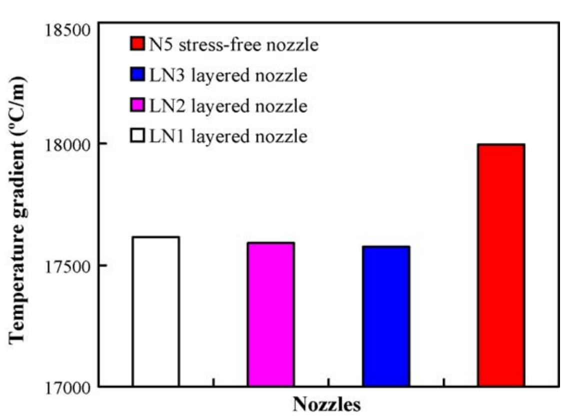

Fig. 13. Comparison of maximum temperature gradients at the exit in different nozzles (°C/m).

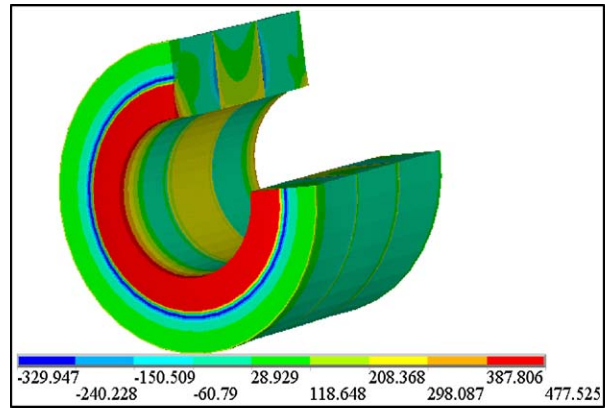

Fig. 14. Thermal stresses in the LN3 layered nozzle (MPa).

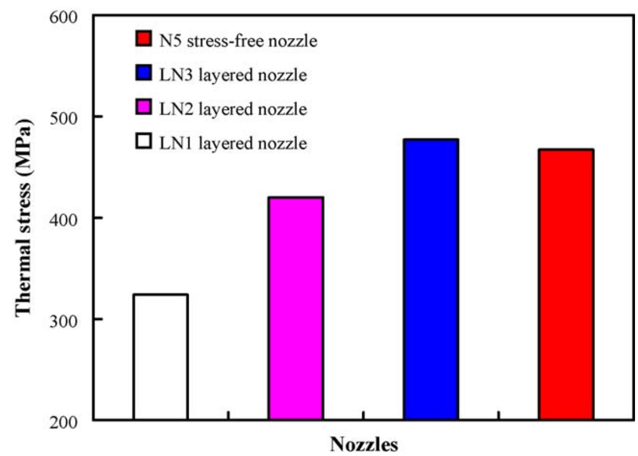

Fig. 15. Comparison of maximum thermal stress at the exit in different nozzles (MPa).

The temperature gradient in LN3 layered nozzle is shown in Fig. 12. It was found that there is greater temperature gradient inside the CWS nozzles, and the highest temperature gradient is 17576 °C/m, and is located at the nozzle exit. Fig. 13 shows the comparison of maximum temperature gradient of LN1, LN2, LN3, layered nozzles and N5 stress-free nozzle at the exit. It is indicated that the maximum temperature gradient of N5 stress-free nozzle is much higher than that of the LN1, LN2, and LN3 layered nozzles.

Fig. 14 shows the thermal stress distribution in LN3 layered nozzle. As can be seen, there are higher thermal stresses inside the CWS nozzles, and the highest thermal stress is located on the exit of the nozzle, and the maximum value is 477.525 MPa. Fig. 15 shows the comparison of thermal stress of LN1, LN2, and LN3 layered nozzles and N5 stress-free nozzle at the exit. It is indicated that the layered nozzles possess lower maximum thermal stresses.

As calculated above, there are higher temperature gradient (Figs. 12 and 13) at the exit of the nozzle, which lead to large thermal stresses (Figs. 14 and 15) during the CWS burning processes. These thermal stresses can lower the fracture strength of the ceramic material, which, in some cases, may be sufficient to cause considerable damage or even catastrophic failure of CWS ceramic nozzles.

There may be several factors that affect the erosive wear resistance of the CWS nozzles. One is the hardness of the CWS nozzles, and the other is the thermal stress inside the CWS nozzles. Once the CWS ceramic nozzle possesses a layered structure, the hardness and fracture toughness at the nozzle external layer are improved (see Table 3), and the temperature gradients and the thermal stresses at the nozzle exit can be greatly reduced due to the different thermal expansion coefficients of the individual layers during CWS burning processes. Therefore, the layered ceramic nozzles exhibited an apparent increase in erosion wear resistance over the unstressed reference one, the mechanisms responsible were found to be that layered structure in the CWS nozzles can improve the hardness and fracture toughness of the external layer, and reduce the temperature gradients and the thermal stresses at the exit of the nozzle.

4. Conclusions

Al_2O_3/(W,Ti)C + Al_2O_3/TiC layered ceramics with different thickness ratios among constituent layers were developed to be used as nozzles in CWS boilers. The performance and wear characteristics of these nozzles were investigated. The following conclusions were obtained:

The layered ceramic nozzles (LN1, LN2, and LN3) exhibited an apparent increase in erosion wear resistance over the unstressed reference nozzle (N5) in industry CWS boilers. The mechanisms responsible were found to be that layered structure in the CWS nozzles can improve the hardness and fracture toughness of the external layer, and can reduce the temperature gradients and the thermal stresses at the exit of the nozzle during CWS burning processes.

It is suggested that layered structures in ceramic nozzles is an effective way to improve the erosion wear resistance over the stress-free ceramic nozzles in industry CWS boilers.

Acknowledgements

This work was supported by the "Taishan Scholar Program Foundation of Shandong", the "Outstanding Young Scholar Science Foundation of Shandong", and the "Key Science and Technology Project of Shandong Province (2006GG2204017)".

References

[1] J. Deng, Z. Ding, D. Yun, Erosion wear mechanisms of coal-water-slurry (CWS) ceramic nozzles in industry boilers, Materials Science Engineering A 417 (2006) 1–7.

[2] Z. Ding, J. Deng, J. Li, Wear behavior of ceramic nozzles in coal water slurry burning, Ceramics International 34 (2004) 591–596.

[3] R. Lakshminarayanan, D.K. Shetty, R.A. Cutler, Toughening of layered ceramic composites with residual surface compression, Journal of American Ceramic Society 79 (1996) 79–87.

[4] P.Z. Cai, D.J. Green, G.L. Messing, Mechanical characterization of Al_2O_3/ZrO_2 hybrid laminates, Journal of the European Ceramic Society 5 (1998) 2025–2034.

[5] G. de Portu, L. Micele, G. Pezzotti, Laminated ceramic structures from oxide systems, Composites: Part B 37 (2006) 556–567.

[6] V.M. Sglavo, M. Bertoldi, Design and production of ceramic laminates with high mechanical resistance and reliability, Acta Materialia 54 (2006) 4929–4937.

[7] S. Guicciardi, M. Nagliati, C. Melandri, G. Pezzotti, D. Sciti, Effects of residual stresses on the fracture properties of non-oxide laminated composites, Journal of the European Ceramic Society 27 (2007) 351–356.

[8] H. Tomaszewski, H. Weglarz, A. Wajler, M. Boniecki, D. Kalinski, Multilayer ceramic composites with high failure resistance, Journal of the European Ceramic Society 27 (2007) 1373–1377.

[9] V. Sergo, D.M. Lipkin, G. De Portu, D.R. Clarke, Edge stresses in alumina/zirconia laminates, Journal of American Ceramic Society 80 (1997) 1633–1638.

[10] G. De Portu, L. Micele, D. Prandstraller, G. Palombarini, G. Pezzotti, Abrasive wear in ceramic laminated composites, Wear 260 (2006) 1104–1111.

[11] A. Tarlazzi, E. Roncari, P. Pinasco, S. Guicciardi, C. Melandri, G. de Portu, Tribological behaviour of Al_2O_3/ZrO_2–ZrO_2 laminated composites, Wear 244 (2000) 29–40.

[12] M. Hadad, G. Blugan, J. Ǩubler, E. Rosset, L. Rohr, J. Michler, Tribological behaviour of Si_3N_4 and Si_3N_4 – % TiN based composites and multi-layer laminates, Wear 260 (2006) 634–641.

[13] J. Deng, Wear behaviors of ceramic nozzles with laminated structure at their entry, Wear 266 (2009) 30–36.

[14] J. Deng, L. Liu, M. Ding, Effect of residual stresses on the erosion wear of laminated ceramic nozzles, Materials Characterization 59 (2008) 1–8.

[15] J. Deng, L. Liu, M. Ding, Erosion wear behaviours of SiC/(W,Ti)C laminated ceramic nozzles in dry sand blasting processes, Materials Science Engineering A 444 (2007) 120–129.

[16] X. Ai, Z. Li, J. Deng, Development and perspective of advanced ceramic cutting tool materials, Key Engineering Materials 108 (1995) 53–66.

[17] J. Barry, G. Byrne, Cutting tool wear in the machining of hardened steels. Part I. Alumina/TiC cutting tool wear, Wear 247 (2001) 139–151.

[18] J. Deng, X. Yang, Wear mechanisms of Al_2O_3/TiC/Mo/Ni ceramic wire-drawing dies, Materials Science Engineering A 424 (2006) 347–354.

[19] R.F. Cook, B.R. Lawn, A modified indentation toughness technique, Journal of American Ceramic Society 66 (1983) 200–201.

[20] J. Deng, D. Yuan, Y. Tan, Design, fabrication and performance of Al_2O_3/($W_{0.7}Ti_{0.3}$)C + Al_2O_3/TiC multilayered ceramic nozzles, International Journal of Refractory Metals & Hard Materials 27 (2009) 734–739.

[21] Z. Ding, Development of assembled coal-water-slurry ceramic nozzles and study on their failure mechanisms, Ph.D. Dissertation, Shandong University, 2004.