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Fabrication and thermal properties of a YSZ–NiCr joint with an interlayer of YSZ–NiCr functionally graded material

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Abstract

Joining yittria stabilized zirconia (YSZ) to Ni–20Cr (NiCr) was fabricated using YSZ–NiCr functionally graded materials (FGM) interlayer by hot pressing process. The thermal properties of this YSZ–FGM–NiCr joint were studied by thermal cycling testing and shear testing. Microscopic observations indicated that the graded distribution of the composition and microstructure in the joint eliminate the macroscopic ceramic/metal interface. The joint shows good thermal stability and good oxidation resistance up to 1000 °C. The thermal fatigue cracks initiate and grow in the way of aggregation of microvoids and their connection in the YSZ-rich NiCr–75 vol.%YSZ interlayer during the thermal cycling to 1000 °C. However, these discontinuous cracks in interlayer release the thermal stress and avoid spallation in the joint interface of direct ceramic/metal bonding. No spallation was found in the joint with three interlayers of 2.0 mm theoretical thick each after 30 thermal cycles. The shear strengths were found to be 207.0 and 75.0 MPa in NiCr–50 vol.%YSZ and NiCr–75 vol.%YSZ interlayers respectively for this joint after 30 thermal cycles.

Keywords: FGM; Joining; NiCr; Thermal properties; YSZ; ZrO_2; Functionally graded materials

1. Introduction

Zirconia ceramics have high fracture toughness and strength, high resistance to wear and corrosion, and high oxide conductivity. They are important in engineering application as both structure and electrical materials. Many applications, such as the gas turbines, engines, solid oxide fuel cell and other industrial applications, require joining of the ceramic to metals or alloys for multiple function and reduced cost.[1–3] The ceramic side offers heat resistance, corrosion resistance or wear resistance, and the metal side provides mechanical strength and thermal conductivity.

Various techniques can be proposed to bond zirconia ceramic to metallic materials. Most of these joints were fabricated by using an inserted metallic interlayer, such as Zr-based alloys,[4] Ag–Cu–Ti alloys,[5] Cu–Ga–Ti and Cu–Sn–Ti alloys,[6] Ti foil[7] and diffusion bonding.[8,9] Due to the difference in the coefficient of thermal expansion (CTE) between ceramic and metals, thermal stress, which would cause crack formation and spallation in the joint, was induce upon cooling from joining temperature or during service of the ceramic components.[10] To overcome this problem caused by the residual stress, multiplayer or functionally graded materials (FGM) have been used for joining ceramic to ceramic or ceramic to metals.[11,12] The continuous changes of constitution between these two sides of FGM can eliminated the macroscopic interface and relax the thermal stress induced by the temperature difference in use. A crack-free Si_3N_4–Al_2O_3 joint showed a dramatic decrease in radial, axial and hoop stress by using a sialon polytypoids as a functionally graded materials interlayers.[13,14] The joint of Al_2O_3 to Ni with Al_2O_3–40 vol.%Ni composite interlayer was produced by hot pressing.[15] Many functionally graded materials have been fabricated and studied for various applications, including metallic–metallic, e.g. W–Mo[16] and W–Cu[17] FGM, ceramic–ceramic, e.g. Si_3N_4–Al_2O_3[13] and Si_3N_4–BN[18] FGM, and ceramic–metal, e.g. SiC–Al 2124,[19] 3Y-TZP-SUS304[20] and ZrO_2–NiAl[21] FGM et al. Joining dense YSZ ceramic to dense NiCr alloy has not been reported. In this study, we fabricated the YSZ–NiCr

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joint using YSZ–NiCr FGM interlayers by hot pressing, and investigated its microstructure and thermal fatigue behaviors.

2. Experimental

A dense fully stabilized zirconia (YSZ) (8 mol.% yittria, 5.9 gcm^{-3} in density) and dense Ni–20 wt.%Cr alloy used to be joined in this study were commercial available, provided from Goodfellow Ltd. UK. They are cylindrical rods with diameter of 12.0 mm. The purity is 99.0% for the YSZ rod and 99.9% for NiCr alloy rod. The YSZ powder (99.9% in purity and less than 1 μm in particle size) and NiCr alloy powder (99.9% in purity and 20 μm in average particle size) from Pi Kem Ltd. UK were used for preparing the FGM interlayers in the YSZ–NiCr joint.

The cylindrical rods of the metal and ceramic were cut into plates with 4.0 mm thick. The flat surface of the plate were polished with the final grit size of 6 μm and ultrasonically cleaned in acetone prior to joining.

The mixtures of metal–ceramic powders with YSZ contents of 25, 50 and 75 vol.% were prepared by ball milling for 12 h using ethanol as a solvent. After being dried and ground, the mixed powders were stacked in the graphite die coated with boron nitride layer by layer between the dense YSZ plate and dense NiCr plate with stepwise compositional distribution, in which composition change from NiCr to YSZ through three interlayers with 25 vol.%YSZ increament. To keep the correct compositional distribution, each layer filled in the die was pre-compacted under a lower pressure before stocking the next layer. The theoretical thickness of 1.0 mm and 2.0 mm of each interlayer calculated from the mixture of fully dense YSZ and NiCr phases was prepared to compare their thermal properties. The joint were performed in vacuum of 5×10^{-5} torr at 1200 °C for 1 h with applied pressure of 10 MPa. The heating and the cooling rate were 10 and 5 °C min^{-1} respectively. The thermal behavior and oxidation resistance were examined by thermal cycling experiment performed in air between room temperature and 1000 °C with an equal heating and cooling rate of 10 °C min^{-1}, kept at 1000 °C for 1 h.

The joints were cross sectioned, polished and observed using optical and scanning electron microscopy (SEM) (Jeol JXA-840) before and after thermal cycling. The phases in the interlayer were identified by XRD analysis (Philip PW 1140/00). The relative densities of each interlayer were obtained by comparing its actual thickness and theoretical one. The mechanical strength was measured by shear testing. The shear strength results are the average of three tests for each condition.

3. Results and discussions

3.1. Joint analysis

A good bond and crack-free YSZ–FGM–NiCr joint was obtained with the FGM interlayers by hot pressing process under this experimental condition. Fig. 1 shows the general view and details of optical microstructures of a cross section for the joint with 1.0 mm theoretical thick of each interlayer. In the microstructures, the YSZ ceramic phase appears dark and NiCr phase appears light. Two ends of the joint are dense YSZ (left in Fig. 1) and dense NiCr alloy (right in Fig. 1). With the variation of composition, the microstructure of interlayer stepwise change from NiCr alloy side (right in Fig. 1) to YSZ side (left in Fig. 1). Metal phase (NiCr alloy) serves as matrix phase and display typical network structure with dispersed YSZ particles in the NiCr-rich region with 25 vol.%YSZ while the YSZ phase becomes matrix phase and the metal phase changes to dispersed one in the YSZ-rich region with 75 vol.%YSZ. The X-ray diffraction patterns from the surface of the cross-section for this joint, shown in Fig. 2, indicated that the phases in the joint were the mixtures of cubic YSZ phase and f.c.c Ni–Cr solution. No new phase was form between YSZ and NiCr alloy during joining process. By comparing the thickness of each interlayer with the theoretical one as we prepared, we obtained that the relative densities for the interlayer with 25, 50 and 75 vol.%YSZ were 99.0, 93.0, and 85.0% respectively, shown in Fig. 3. The relative density of the interlayer is decreases as the YSZ content increases in the interlayer. The fabrication temperature may be still low for sintering YSZ ceramic even under hot pressure.

3.2. Thermal cycling behaviors

To examine the thermal stability, oxidation resistance and thermal fatigue behaviors of YSZ–FGM–NiCr joint, the samples were heated and cooled in air cyclically between room temperature and 1000 °C with heating and cooling rates of 10 °C min^{-1}. Experimental results showed the YSZ–FGM–NiCr joint has good thermal stability and oxidation resistance. For the joint with 1.0 mm theoretical thick of each interlayer, the cracks were found in the YSZ-rich region with 75 vol.%YSZ even after one thermal cycle while there is crack-free in the as-prepared sample. The cracks propagated and grew as thermal cycle increased. Fig. 4 shows the microstructures of the cross-section for this joint after two thermal cycles. From Fig. 4, one can see that a large number of cracks only exist in the NiCr–75 vol.%YSZ interlay instead of others or at interface. These cracks almost parallel to the interface of the interlayer, forming network. The density of cracks in the region close to pure YSZ layer is higher than that in

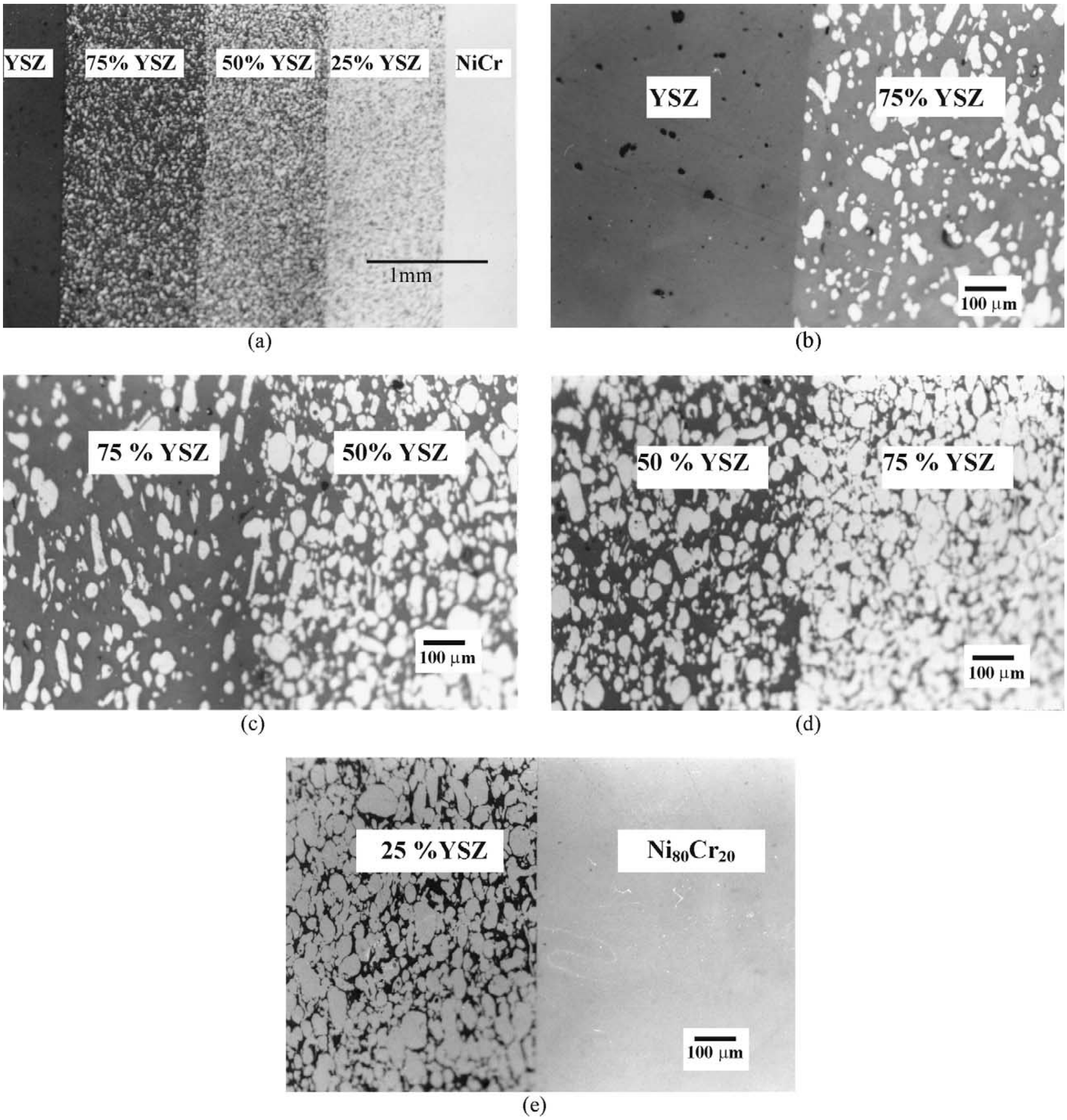

Fig. 1. Optical microstructures of a cross-section for the YSZ–FGM–NiCr joint with 1.0 mm theoretical thickness of each interlayer; general view (a) and the different interfaces (b), (c), (d) and (e).

the region close to the NiCr–50vol.%YSZ interlayer (Fig. 4b and c). The cracks exist in the ceramic region and end by the dispersed metal phase in this composite interlayer. These cracks should be caused by the thermal stress induced by the difference of the coefficients of thermal expansion (ETCs) between two components and two different compositional interlayers, and the low mechanical strength in this interlayer.

Fig. 3 also gives the ETCs for each interlayer calculated from the mechanical parameters of pure YSZ and pure NiCr using Turner formula:

$$\alpha_0 = \frac{\alpha_m K_m V_m + \alpha_c K_c (1 - V_m)}{K_m V_m + K_c (1 - V_m)}$$

$$K_m = \frac{E_m}{2(1 - \nu_m)}, \qquad K_c = \frac{E_c}{2(1 - \nu_c)}$$

where subscripts m and c indicate metal and ceramic, and E, ν and α are Youngs modulus, Poissons ratio and CTE. The ETC increases linearly from 10 to 14×10^{-6}°C^{-1} as NiCr content increases in the interlayer.

As mentioned in Section 3.1, the relative density of the NiCr–75 vol.%YSZ interlayer was lowest in the

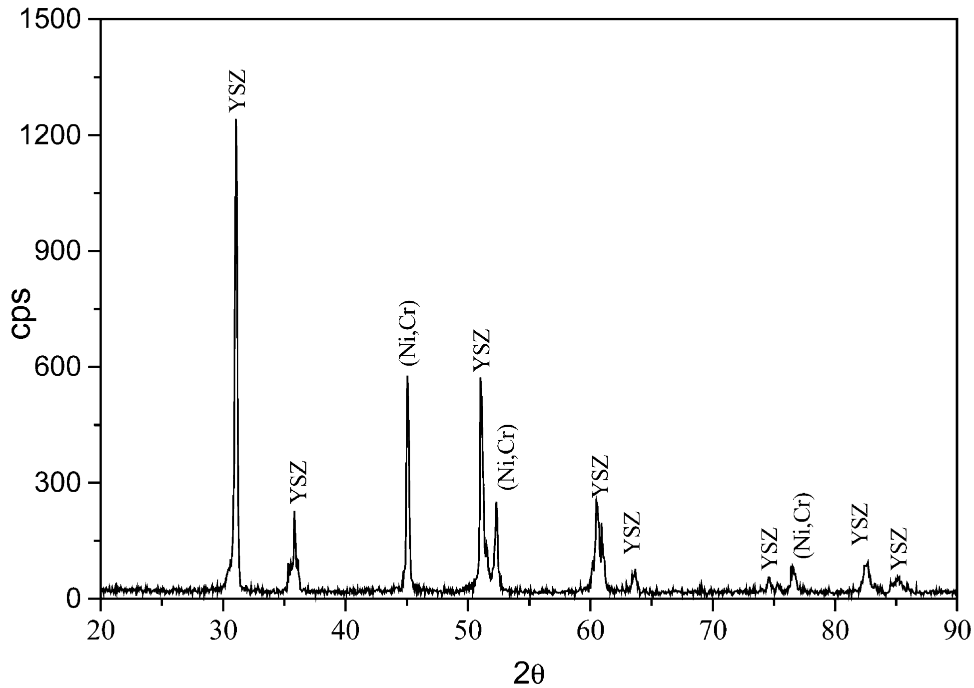

Fig. 2. XRD pattern from the surface of a cross-section for YSZ–FGM–NiCr joint.

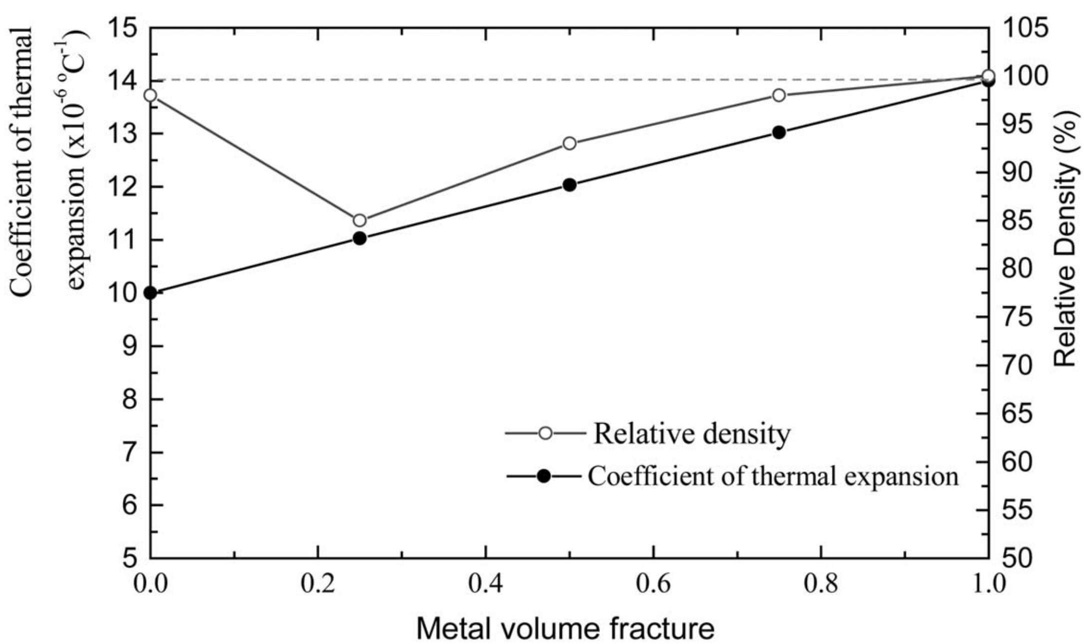

Fig. 3. Coefficients of thermal expansion (CTEs) and relative densities for the layers in YSZ–FGM–NiCr joint.

joint. It leads to large number of microvoids and low mechanical strength in this interlayer. During the thermal cycling, the alternate thermal stress in the interlayer with 75%YSZ induced by the difference of the ETCs between two components and two different compositional interlayers is high enough to cause aggregation of the microvoids and connection, forming cracks and propagating along the stress direction. The cracking in the region close to pure YSZ layer is greater than in the region close to the NiCr–50vol.%YSZ interlayer (Fig. 4b and c) means that the thermal stress should be higher in the first region due to the difference of the ETCs between nearly fully dense YSZ and the low density NiCr–75 vol.%YSZ interlayer. However, the stress energy is released as the formation of and growth of the cracks. The soft NiCr phase in the FGM interlayer acts as dissipating the strain energy, changing the cracking direction, passivating and ending the cracks (Fig. 5).

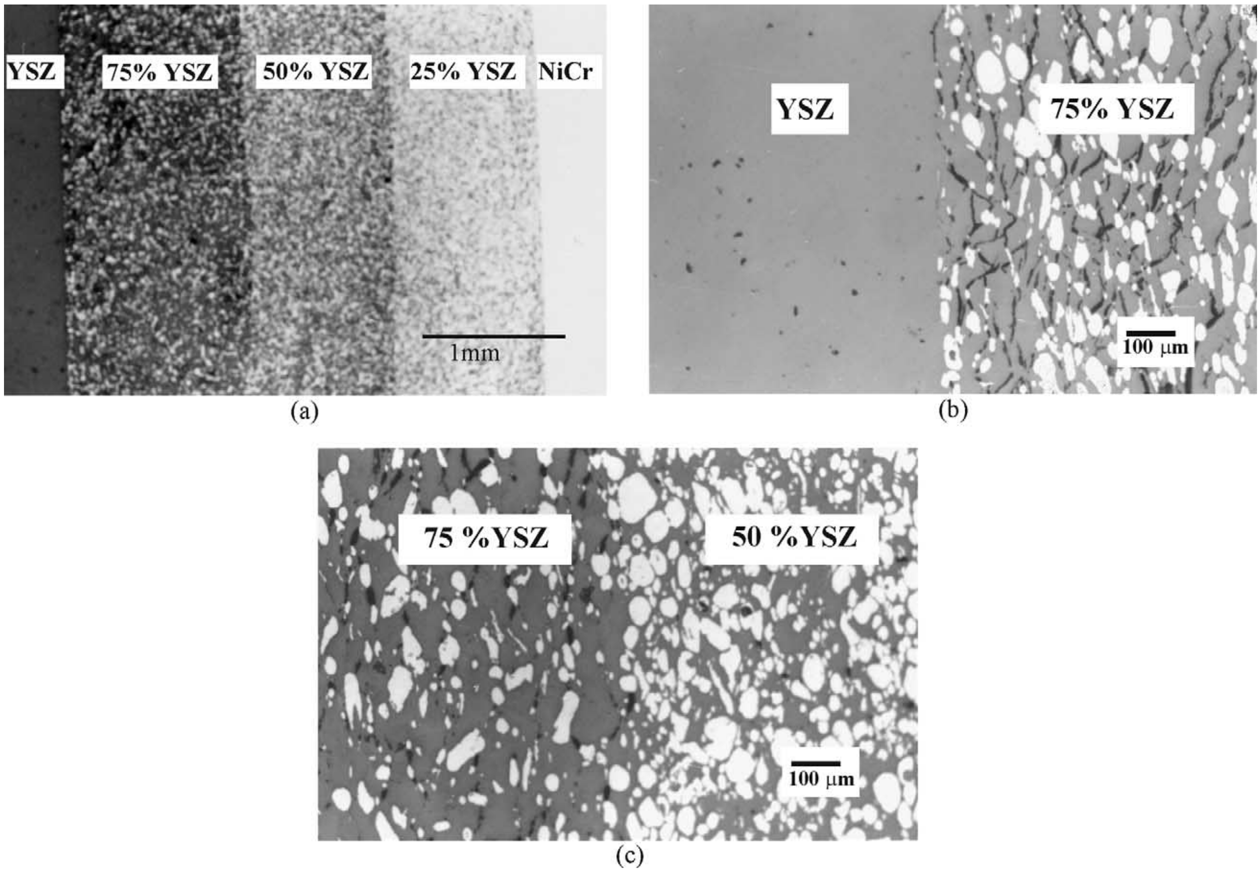

Fig. 4. Optical microstructures of a cross-section for the YSZ–FGM–NiCr joint with 1.0 mm theoretical thickness of each interlayer after two thermal cycles up to 1000 °C; general view (a), the NiCr–75 vol.%YSZ interlayer near by YSZ (b) and the NiCr–75 vol.%YSZ interlayer near by the NiCr–50 vol.%YSZ interlayer (c) showing thermal cracking.

The cracks can be developed and propagated anywhere in this region independently, so they are discontinuous. These discontinuous cracks in interlayer release the thermal stress and avoid spallation at the joint interface of direct ceramic/metal bonding.

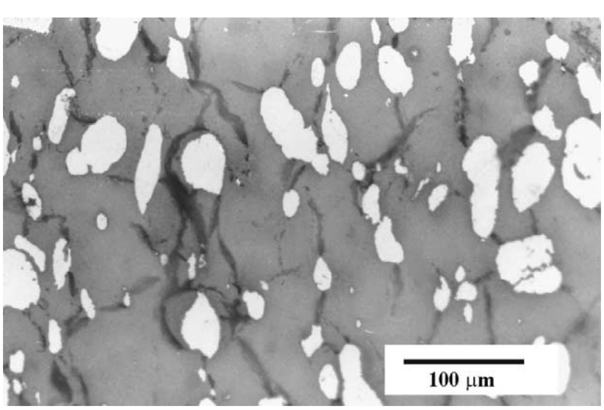

Fig. 5. Optical microstructures showing the cracks ended by soft NiCr phase in the NiCr–75vol.%YSZ interlayer of YSZ–FGM–NiCr joint with 1.0 mm theoretical thickness of each interlayer after two thermal cycles up to 1000 °C.

Increasing the mechanical strength by increasing its relative density or reducing the thermal stress by increasing the thickness of the interlayer can improve the mechanical and thermal properties of the YSZ–FGM–NiCr joint. A crack-free YSZ–FGM–NiCr joint with 2.0 mm theoretical thick of each interlayer was prepared under the same fabrication conditions. Thermal cycling test for this joint indicated that much improvement of the thermal properties could be obtained by increasing the thickness of the interlayer. The cracks were also developed first in the NiCr–75 vol.%YSZ interlayer, a few was found in the NiCr–50 vol.%YSZ interlayer in this joint after five thermal cycles, but the cracks is much smaller in dimension and much lower in density as compared to that in the joint with the 1.0 mm theoretical thick of each interlayer. The general view and details of optical microstructure of the cross-section showing the cracks developed in the NiCr–75 vol.%YSZ interlayer for this joint with 2.0 mm theoretical thick of each interlayer after 10 thermal cycles are shown in Fig. 6a and b. From the figure, one can see the cracks formed in this joint after 10 thermal cycles are shorter in dimension and lower in density than those formed in the joint with 1.0 mm theoretical thick of

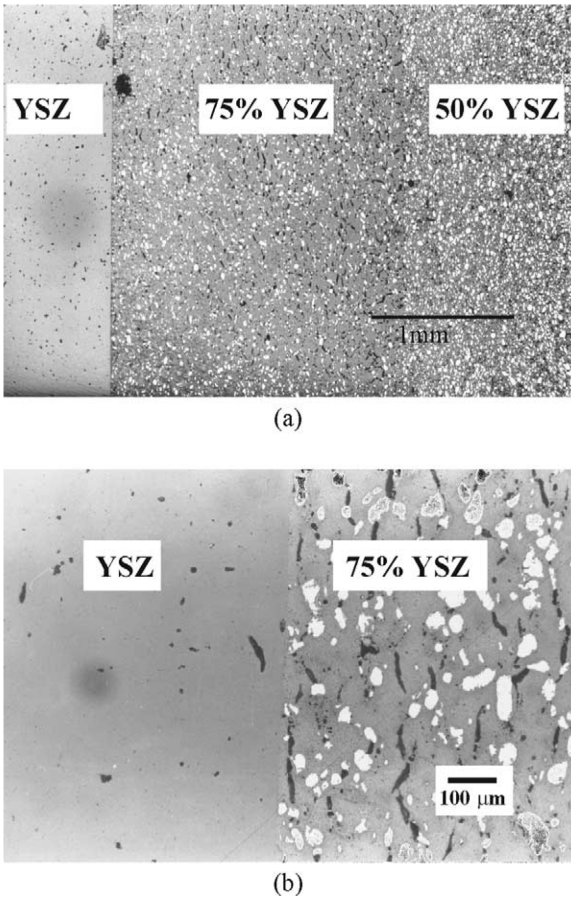

Fig. 6. Optical microstructures of a cross-section for YSZ–FGM–NiCr joint with 2.0 mm theoretical thickness of each interlayer after 10 thermal cycles up to 1000 °C; general view (a) and detail of the NiCr–75 vol.%YSZ interlayer showing thermal cracking.

each interlayer even after two thermal cycles. Ref. 22 gave the relationship between the plastic deformation of FGM and the thickness of the interlayer. The thicker interlayer has lower thermal stress. The cracks generated in this joint after five thermal cycles and grew very slowly for further thermal cycles. No spallation was found in this joint after 30 thermal cycles. The shear test in this joint showed that the shear strength is still as high as 207.0 MPa for NiCr–50 vol.%YSZ interlayer and 75.0 MPa for NiCr–75 vol.%YSZ interlayer after 30 thermal cycles.

4. Conclusion

We fabricated YSZ–FGM–NiCr joints by hot pressing process and studied their thermal properties by thermal cycling test and shear testing. The joint shows good thermal stability and good oxidation resistance up to 1000 °C. The cracks were initial and grew in the ceramic-rich NiCr–75 vol.%YSZ interlayer . The stepwise compositional change of interlayer turns the metal/ceramic interface of traditional metal/ceramic bonding into a large number of metal/ceramic interfaces, thus it reduce the thermal stress during fabrication and thermal cycling. The soft NiCr phase in the FGM interlayer acts as dissipating the strain energy, passivating and ending the cracks. The continuous cracks in interlayers release the thermal stress and avoid spallation at the joint interface of direct ceramic/metal bonding. No spallation was found in the joint with each interlayer thickness of 2.0 mm after 30 thermal cycles. The joint showed shear strength of 75.0 MPa in NiCr–75 vol.%YSZ interlayer after 30 thermal cycles.

Acknowledgements

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