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Bonding of partially-stabilized zirconia and nickel with nickel oxide layer

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Bonding of partially-stabilized zirconia (PSZ) and nickel has been investigated. The couples of PSZ and nickel were bonded at 1073 to 1273 K for 300 to 3600 sec under a pressure of 0.75 to 262 MPa in air. PSZ was bonded to nickel with a nickel oxide layer. The nickel oxide layer filled the interstices between PSZ and nickel. The maximum shear strength (about 90 MPa) was obtained in the couple bonded at 1173 K for 900 sec under pressure above 18 MPa. The stresses generated by the differential thermal contraction and the oxide growth stresses are discussed.

1. Introduction

Ceramics have a high heat-resistance, wear resistance, corrosion resistance, insulation, low thermal conductivity, etc. On the other hand, they have weaknesses in thermal shock-resistance, ductility, workability and mass effect [1]. Composite materials made by bonding ceramic and metal are expected to be one of the newly developed functional materials. Since there is a definite difference in macroscopic and microscopic properties between ceramics and metals, the complicated and clever bonding methods of soldering, blazing, organic bonds, inorganic bonds, coating of ceramic, mechanical bonding, direct solid-state bonding, etc, have been reported for bonding technologies [2]. In the case where the bonded materials are used at high temperatures, the direct solid-state bonding method is expected to be best.

Much attention has recently been paid to partially-stabilized zirconia (PSZ) which has superior thermal shock-resistance, high strength and toughness [3]. Nickel is one of the base metals of heat-resistant alloys. The purpose of this paper is to investigate the direct solid-state bonding between PSZ and nickel in the temperature range 1073 to 1273 K in air.

2. Experimental procedures

The chemical composition of PSZ is listed in Table I. The samples of PSZ (3 mm × 4 mm × 5 mm) were prepared by sintering at 1773 K for 7.2×10^3 sec in air. The density was $6.0 \times 10^3\,\mathrm{kg\,m^{-3}}$, and the grain diameter was about 1 μm. The bending strength was 980 MPa. The surfaces of PSZ were ground and polished before observation by scanning electron microscopy (SEM). The SEM photograph of the surface of PSZ is shown in Fig. 1. The nickel ingot was prepared with 99.97% Ni in a high-frequency induction furnace in a vacuum of 1×10^{-2} Pa. The ingot was section-rolled to rod at room temperature. Nickel wire was drawn from the rod to 4 mm diameter at room temperature, and was cut into 6 mm lengths as nickel columns. These columns were annealed at 1473 K for 3.6×10^3 sec in a vacuum of 1×10^{-3} Pa in order to eliminate the effects of working. The cross sections of the nickel columns were ground vertical to the axial direction by 600 emery paper and polished using fine Al_2O_3 powder. Some of them were annealed at 1173 K for 900 sec in air in order to develop the oxide film (NiO) on the polished surface. The nickel columns and PSZ were cleaned by acetone in ultrasonic cleaning equipment.

TABLE I Chemical composition of PSZ (wt %)

ZrO_2	Y_2O_3	Al_2O_3	HfO_2	SiO_2	Na_2O	TiO_2
92.89	4.54	1.24	1.23	0.05	0.03	0.02

The couples which consisted of PSZ and nickel were referred to as couple A, and the couples with PSZ and oxidized nickel were referred to as couple B. Fig. 2 shows a schematic drawing of the bonding apparatus. The couples were introduced to the stainless steel fixtures and pressed under bonding pressures of 0.75 to 262 MPa. They were then annealed in the temperature range 1073 to 1273 K for 3×10^2 to 3.6×10^3 sec in air. The heating rate was about $0.5\,\mathrm{K\,sec^{-1}}$. The bonding time was taken to be the annealing time when the couples were held at the prescribed temperatures. After bonding treatments, the couples were taken out of the apparatus and then were cooled in air. In addition to the bonding treatments, part of couples A bonded at 1173 K for 900 sec under 29 MPa in air were annealed at temperatures between 1073 and 1373 K for 1.8×10^3 sec in air or in evaculated silica capsules. The heating rate was about $0.3\,\mathrm{K\,sec^{-1}}$. The couples were cooled at about $0.5\,\mathrm{K\,sec^{-1}}$ in the temperature range above 1173 K, and then were air-cooled from 1173 K to room temperature.

The shear test was used to evaluate the bonding strength. The shear strength of the couples was measured by an Instron-type machine with the implements as shown in Fig. 3. The cross head speed

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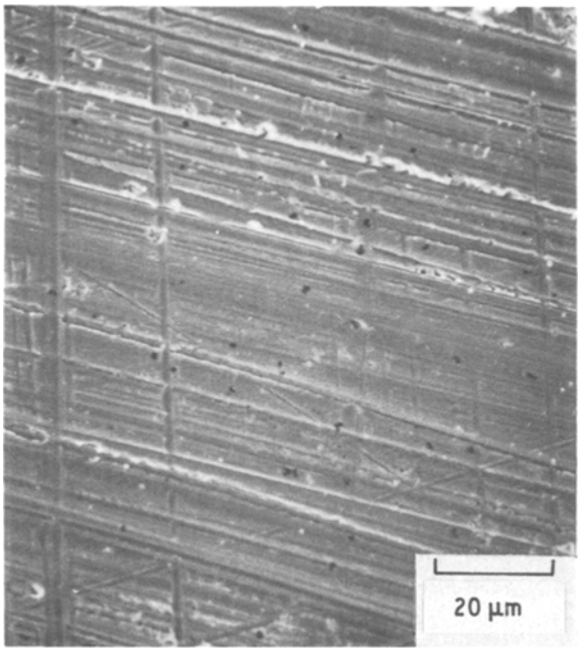

Figure 1 The SEM photograph of surface of PSZ before bonding.

was 0.1 mm $\sec^{-1}$. The fracture surface of the couples and bonding interface were observed by SEM. The concentration distribution of elements in couples was analysed by energy dispersion X-ray microanalysis (EDX), and oxygen was analysed using the wave dispersion X-ray microanalyser (WDX).

3. Results and discussion

Fig. 4 shows examples of the SEM photographs and the profiles of X-ray intensities of zirconium, yttrium, nickel and oxygen in the vicinity of the bonding interface of couples A bonded at 1273 K for 900 sec in air. A layer, about 3 μm thick, forms between PSZ and nickel. Zirconium and yttrium do not penetrate into this layer, and X-ray intensities of the other composite elements (aluminium, silicon, sodium, titanium, hafnium) cannot be detected in this layer. According to the profiles of oxygen and nickel, the layer between PSZ and nickel is the same oxide developed at the surface of nickel columns. This layer is considered to be a nickel oxide layer (NiO). Furthermore, the reaction between PSZ and the nickel oxide layer cannot be observed by SEM and EDX. This is consistent with the report by Econmos and Kingery [4].

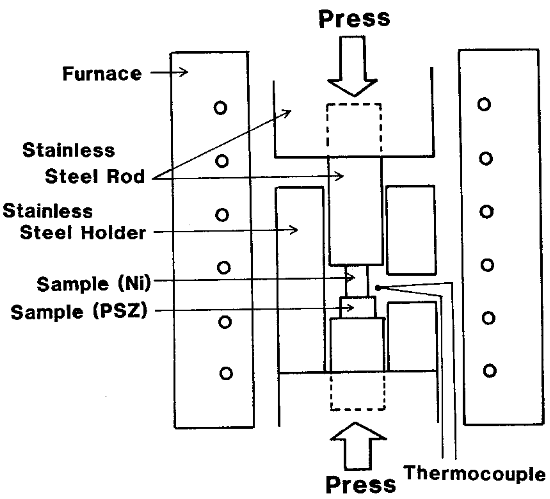

Figure 2 Schematic drawing of bonding apparatus.

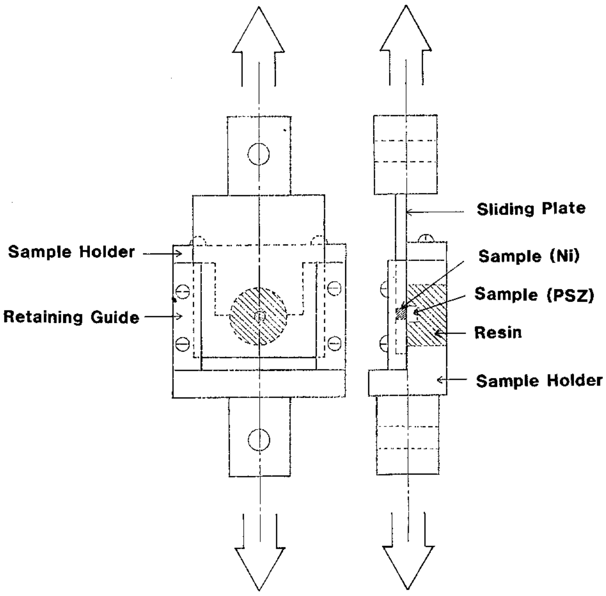

Figure 3 Schematic drawing of stainless steel implements of the shear test.

Fig. 5 illustrates the effect of the bonding pressure on the shear strength of couples A bonded at 1173 K for 900 sec. The open circles indicate the fracture in the nickel oxide layer between PSZ and nickel, and the solid circles indicate the fracture in PSZ. Couples having a shear strength lower than 36 MPa fracture in the nickel oxide layer between PSZ and nickel. The fracture in PSZ appears in some of the couples having a shear strength higher than 36 MPa. The shear strength increases linearly to about 90 MPa with bonding pressures up to about 18 MPa. Further increases in bonding pressure above 18 MPa does not increase the bond strength. However, a large plastic deformation of nickel is observed in the couples bonded above bonding pressures of about 66 MPa. A similar tendency is observed in the bonding between niobium and Al_2O_3 [5]. Bonding pressures above 18 MPa are necessary for bonding treatments at 1173 K for 900 sec in order to obtain a strong bonding between PSZ and nickel. Fig. 6 shows the SEM photographs of the sheared bond faces of couples A bonded under bonding pressures of 1.8 and 20 MPa. The adherent area between PSZ and nickel in couple A bonded under a low bonding pressure is smaller than that under a high bonding pressure. The high bonding pressure gives very intimate contact between PSZ and nickel, although the optimum bonding pressure depends on the degree of flatness of nickel, bonding temperature and bonding time.

Fig. 7 illustrates the effect of bonding temperature on the shear strength of couples A bonded for 900 sec under 29 MPa. The shear strength increases steeply with temperature up to 1173 K, and then decreases gradually. Fig. 8 shows SEM photographs of the bonding interfaces of couples A bonded at various temperatures for 900 sec under 29 MPa. In the case of couple A bonded at 1073 K, the interstices remain in the interface between PSZ and nickel. The nickel oxide

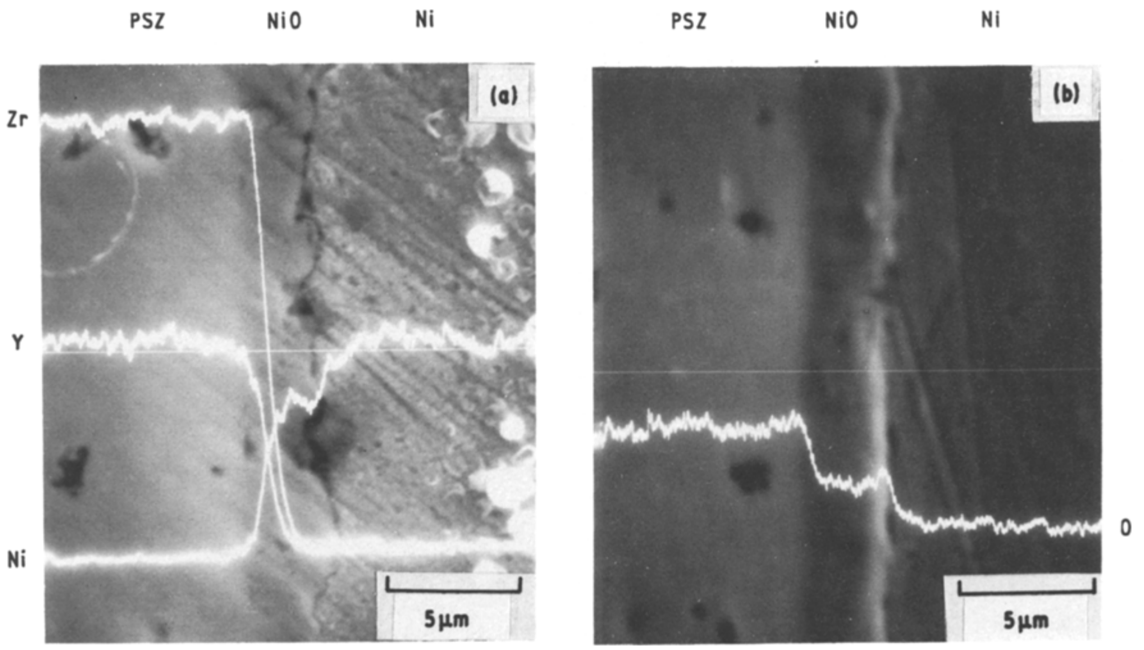

Figure 4 The profiles of X-ray intensities of the elements of (a) zirconium, nickel, yttrium and (b) oxygen in couples A bonded at 1273 K for 900 sec under 15.6 MPa in air.

layer is too thin to fill them because nickel oxide grows slowly at low temperature. In couple A bonded at 1173 K, the interstices between PSZ and nickel are compactly filled with the nickel oxide layer. Since the nickel oxide layer grows by a thermal activated process, the nickel oxide layer grows more thickly and fills the interstices more compactly with increasing temperature. Therefore, the shear strength increases steeply with temperature up to 1173 K. In the case of couples A bonded at 1273 K, voids are observed in the nickel side of the nickel oxide layer. This is consistent with reports [6–8] that the oxide become more porous at high temperatures and that voids develop in NiO via cracks parallel with and close to the metal–oxide interface. These voids develop with temperature and reduce the shear strength.

Fig. 9 illustrates the bonding time dependence of the shear strength of couples A bonded at 1173 K under 29 MPa. The shear strength has a maximum at about 900 sec. This tendency is similar to that of temperature dependence. The bonding time is closely related to the bonding temperature. At short bonding time, the interstices remain at the interface between PFZ and the nickel oxide layer because of a thin layer of nickel oxide. At longer bonding times, voids were observed in the nickel oxide layer. The average value of the shear strength of couple B bonded at 1173 K for 900 sec under 29 MPa is 70 MPa, and is lower than 90 MPa of couple A bonded under the same conditions. This lower bond strength is caused by the interstices which are observed at the interface between the PSZ and the nickel oxide layer. The nickel oxide layer was reported to grow with time according to the parabolic law [9]. Therefore, since the nickel oxide layer developed during the initial annealing (900 sec) before bonding, grows more slowly during the second annealing (bonding treatments), the interstices remain at the interface between the PSZ and the nickel oxide layer.

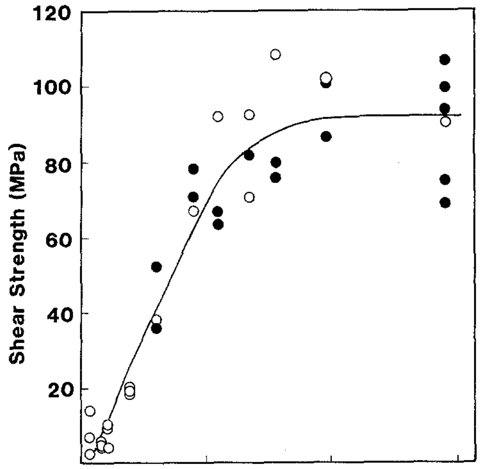

Figure 5 The pressure dependence of the shear strength of couples A bonded at 1173 K for 900 sec in air. (○) Fracture in the layer of NiO between PSZ and Ni; (●) fracture in PSZ.

When the couples cool from the bonding temperatures to room temperature, thermal contracting occurs. Stress will be generated in the vicinity of the bonding interface if the thermal expansion coefficients are dissimilar for the ceramic and metal. The thermal expansion coefficients and elastic modulus are listed in Table II. The thermal expansion coefficients of nickel and NiO are nearly equal. Since the stress at the

TABLE II The values of thermal expansion coefficients and elastic modulus used in calculation

	Ni	NiO	PSZ
Thermal expansion coefficients (10^{-6} K^{-1})	17.6 [10]	17.1 [10]	11.0
Elastic modulus (GPa)	205 [13]	277 [12]	208

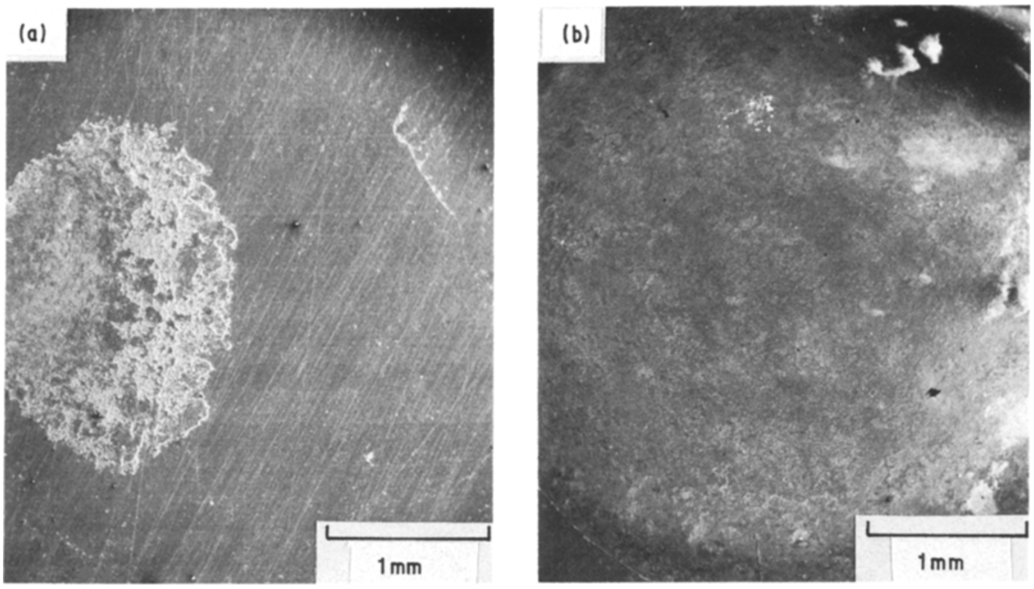

Figure 6 The SEM photographs of the sheared bonding faces on PSZ side of couples A bonded at 1173 K for 900 sec under pressures of (a) 1.8 MPa and (b) 20 MPa in air.

interface between nickel and NiO is expected to be small enough to ignore, the residual stress at the PSZ/NiO interface is discussed here. The magnitude of the stress generated by differential thermal contraction has been given [10],

$$\sigma_1 = \frac{E_1 \Delta T(\alpha_1 - \alpha_2)}{1 + 2E_1 t_1/(E_2 t_2)} \qquad (1)$$

where σ_i is the stress, α_i the thermal expansion coefficient, E_i the elastic modulus (listed in Table II), t_i the thickness, and ΔT the temperature change. Subscripts ($i = 1, 2$) indicate the layers. The stress caused by the thermal contraction is calculated to be the tensile stress of about 560 ~ 770 MPa by using Equation 1 with the values in Table II. On the other hand, it was reported that the oxide growth stresses were developed during the oxidation of metals [11]. Ueno [12] pointed out that very high compressive stresses were caused in the layer of NiO by the oxidation of nickel and evaluated them to be more than 940 MPa. Since this compressive stress caused by isothermal oxidation can be added to the stress generated by differential thermal contraction, the tensile stress is eliminated by the high compressive stress and the compressive stress eventually exists in the NiO layer.

Fig. 10 illustrates the effect of the additional annealing in vacuum or air on the bonding strength of couples A which were bonded at 1173 K for 900 sec under 29 MPa. The shear strength of the couples annealed in air decreases slightly with annealing temperature. This decrease is mainly caused by the development of voids as mentioned above. The shear strength of the couples annealed in vacuum decreases linearly to a large

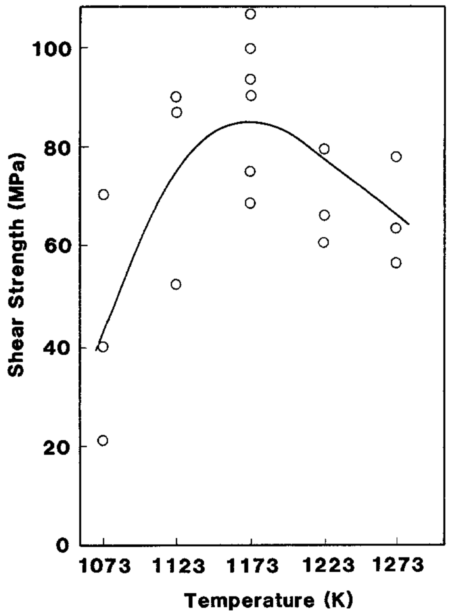

Figure 7 The temperature dependence of the shear strength of couples A bonded for 900 sec under 29 MPa in air.

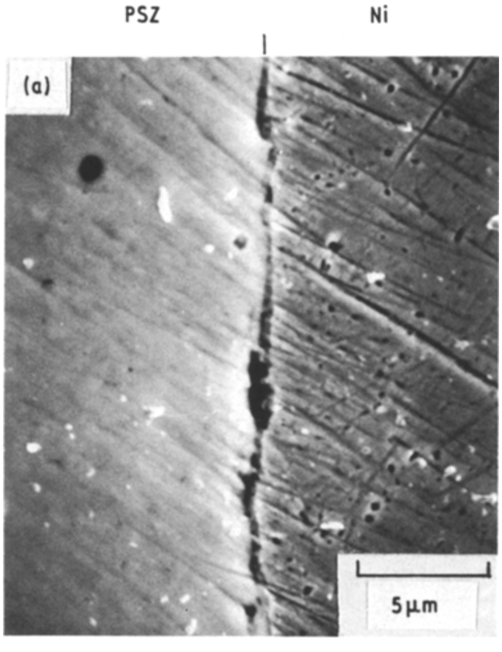

Figure 8 The SEM photographs of the bonding interfaces of couples A at (a) 1073 K, (b) 1173 K and (c) 1273 K for 900 sec under 29 MPa in air.

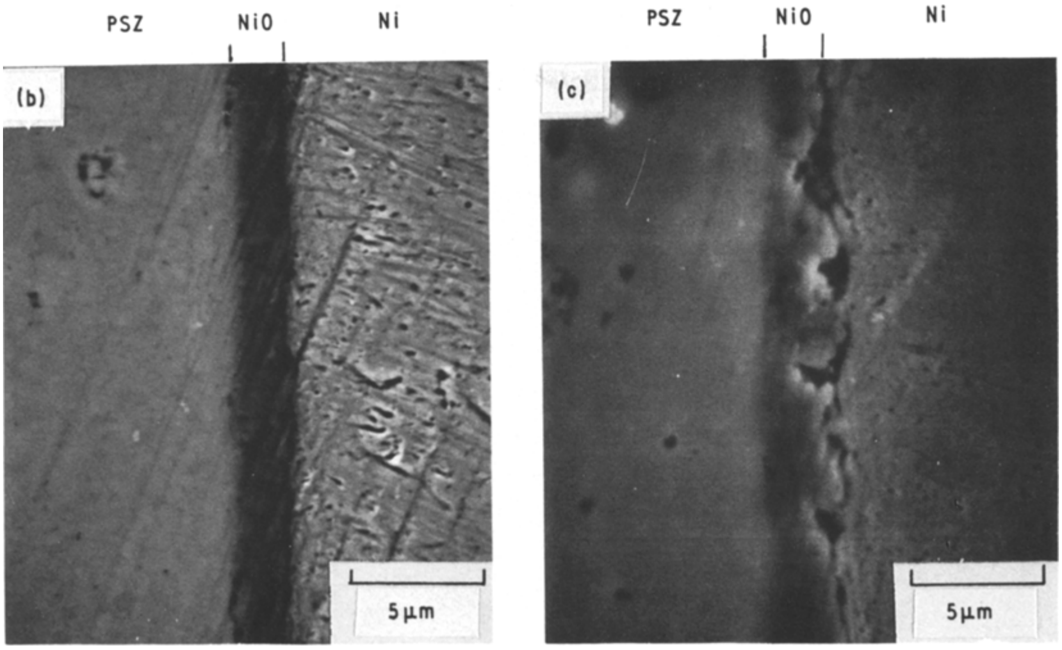

Figure 8 Continued.

degree with the temperatures of additional annealing. During annealing in vacuum, the NiO does not grow and the compressive stress caused by isothermal oxidation cannot be generated in the oxide layer. Furthermore, the compressive stress caused by oxidation during the bonding treatments is eliminated by additional annealing in vacuum. The tensile stress produced during cooling only acts on the NiO layer and lessens the bonding stress of the couples annealed in vacuum. Fig. 11 shows the interfaces of bonded couples A after annealing at 1273 K for 1.8×10^3 sec in air and vacuum. The couple annealed in air has a sound layer of NiO, although some voids appear in the nickel oxide layer. On the other hand, the nickel oxide layer in couples annealed in vacuum is smashed to pieces. This indicates that the compressive stress caused by isothermal oxidation protects the nickel oxide layer from damage by the tensile stress generated during cooling and maintains the high bonding strength.

4. Conclusions

1. PSZ was bonded to metal nickel with a layer of NiO.
2. The bonding strength depends on bonding pressures, times and temperatures. A maximum shear strength of 90 MPa is gained in the PSZ and nickel couples bonded at 1173 K for 900 sec under 29 MPa in air, where the interstices at the bonding interface are compactly filled with a layer of NiO.
3. The compressive stress caused by isothermal oxidation protects the layer of NiO from damage during cooling and maintains the high bonding strength.

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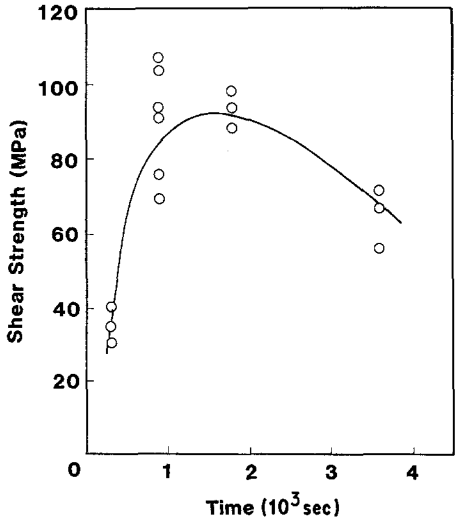

Figure 9 The time dependence of the shear strength of couples A bonded at 1173 K under 29 MPa in air.

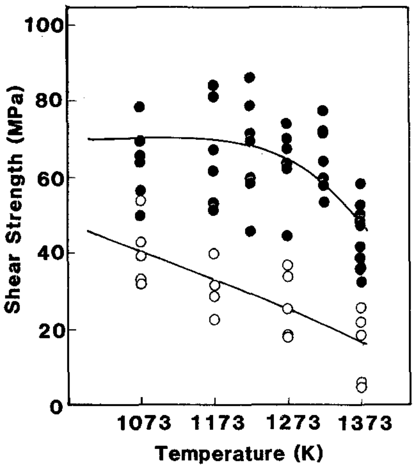

Figure 10 The effect of the additional annealing (○) in vacuum and (●) in air on the bonding strength of couples A bonded at 1173 K for 900 sec under 29 MPa in air.

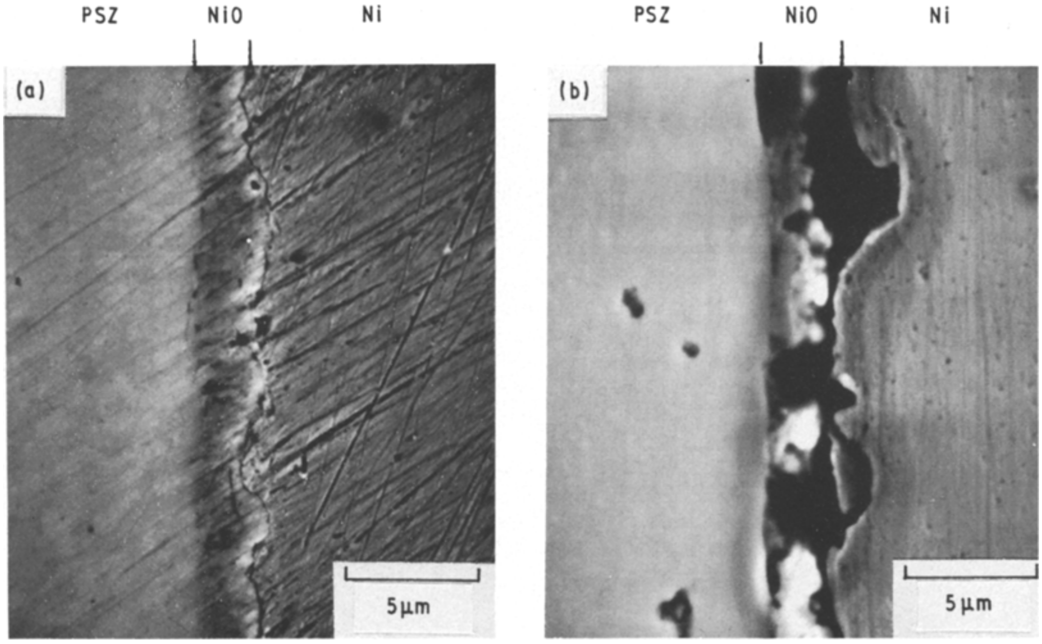

Figure 11 The SEM photographs of the bonding interface of couple A bonded at 1173 K for 900 sec under 29 MPa in air, and then annealed at 1273 K for 1.8 × 10^3 sec (a) in air, (b) in vacuum.

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