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ScienceDirect
Ceramics International 36 (2010) 741–747
CERAMICS INTERNATIONAL
www.elsevier.com/locate/ceramint

Alumina–copper joining by the sintered metal powder process

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Received 2 February 2009; received in revised form 20 March 2009; accepted 4 October 2009
Available online 3 November 2009

Abstract

Sintered metal powder process (SMPP) is one of the high technology methods in ceramic–metal joining domain. The present study examines the effect of temperature and time of metalized layer sintering on the thickness and homogeneity of the joining layer, the leakage rate in alumina–copper joining zone, and also identifies the different phases formed during sintering. The samples were characterized by optical microscopy (OM), scanning electron microscopy (SEM) and energy dispersion spectroscopy (EDS). Microstructure studies indicate that sintering the metalized layer with a holding time of 90 min at the temperature of 1530 °C, and with an applied layer thickness of 50 μm with proper plating and brazing stages lead to a completely homogeneous joining zone with an adequate thickness (about 33 μm). The results of leak tests on alumina–copper specimen in this condition was less than 10^{-9} Pa l s^{-1}.

Keywords: A. Joining; B. Electron microscopy; Sintered metal powder process (SMPP); Alumina

1. Introduction

During the last thousand years, man has tried different methods for producing strong and secure ceramic–metal joints [1], but developments in industrial processes were achieved in the 1980s [2].

From a theoretical point of view, a wide range of methods are applicable for ceramic–metal joining; but because of differences in the physical and chemical structures of ceramics and metals only a few methods can be used. One of the methods which results in sound joints between ceramic and metal is coating a Mo–Mn layer on alumina surface and sintering the metalized layer before brazing. This method is known as sintered metal powder process (SMPP) [3]. In first attempts, Pulfrich [4] introduced a mechanism for ceramic–metal joining in 1930. In later years scholars such as Pincus, Cole, Hynes and Denton [5], introduced some studies on phase formation during sintering and the effect of glassy phases on metalized layer adhesion. Floyd investigated the effect of particle size on the metalized layer adhesion [5].

The metalized layer formula in the Mo–Mn process consists of a mixture of Mo and Mn powders with some glassy compounds and a solvent which is applied in the form of slurry on the ceramic surface [6–9]. The main purpose of SMPP is wetting the ceramic surface for brazing. After sintering the metalized layer, in order to increase the surface wetness and avoid any attacks between the created joint of alumina and Mo–Mn layer by filler braze, metalized layer must be plated [10]. Main advantages of SMPP are applicability for large number of samples, lower cost, less sensitivity to process parameters against other brazing techniques like active metal brazing, good results in leak tests, and wide usage for alumina as most applicable ceramic. Numerous process steps is the main disadvantage of this process.

Successful ceramic–metal joining depends on different factors such as: joining zone thickness, phase formation during sintering, metalized layer morphology, conditions of plating and brazing. Thus, to have a secure joint between ceramic and metal, accurate investigation of effective factors should be taken into consideration [11].

In the present research, the effects of temperature and time of metalized layer sintering on the thickness and homogeneity of joining layer and alumina–copper joining zone leakage rate have been studied based on metalized layer applied composition.

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0272-8842/$36.00
doi:10.1016/j.ceramint.2009.10.016

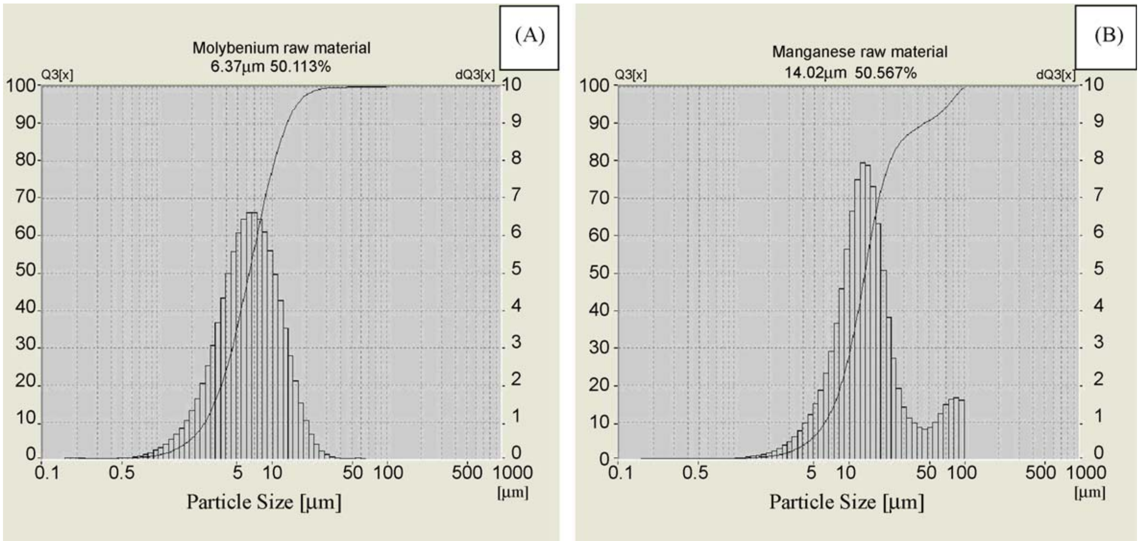

Fig. 1. Laser particle size distribution of (A) Mo and (B) Mn powder.

2. Experimental procedure

The slurry was made of Mo, Mn, Al_2O_3, CaO and SiO_2 99.99% pure powders. Particle size distributions of Mo and Mn powders are shown in Fig. 1. Butyl acetate, nitro cellulose and butyl oxalate used in the slurry were provided as high purity materials as well. Sintering was carried on in a 1500 °C furnace with 6 l capacity under hydrogen atmosphere.

Ceramic discs samples with $d = 25$ mm and $H = 7$ mm dimensions were cleaned in an ultrasonic set which contains F.E.P. solution, then washed in 50% pure HCl solution for 5 min. To make the required slurry, a combination of 70% Mo powder, 10% Mn powder, 5% SiO_2 powder, 3% CaO powder and 12% Al_2O_3 were mixed. Butyl acetate, nitro cellulose and butyl oxalate were added to the mixture. The prepared slurry mixture was ball milled for 48 h. The slurry was painted on the surface of alumina by a painting brush. The applied layer on the surface of sample is between 25 and 50 μm thick. The sintering of metalized specimens was conducted in a furnace under wet hydrogen atmosphere. Sintering temperatures were 1400, 1470, 1530 and 1600 °C with holding times of 40, 60 and 90 min. After primary sintering process, the metalized layer surfaces were nickel plated about 5 μm thick by electroplating method. Electroplating process was carried out using 300 g/l $NiCl_2$, 30 g/l H_3BO_3 solution with pH about 4–5 at room temperature. In order to achieve plating layer with thickness of about 5–7 μm, current density of 5.5 A/m^2 were used for 40–50 min. Samples were then sintered under wet hydrogen atmosphere, again. At the end, Ag (75%)–Cu (20%)–Pd (5%) filler with $d = 15$ mm and $H = 0.5$ mm dimensions was inserted between alumina and high free oxygen copper (HFOC) in $d = 25$ mm and $H = 5$ mm dimensions. The samples were conducted in a furnace under the hydrogen atmosphere for brazing process.

Leak test was done on all samples in order to calculate the leakage rate between alumina and copper joining zone based on ASTM F19-64 [12]. The brazed test specimen were vacuum-leak checked on a conventional helium spectrometer-type leak checker with sensitivity of 10^{-9} Pa l s^{-1} at standard temperature and pressure. The samples were subjected to an atmosphere of helium for 0.5–5 min. The analysis of the structure of inter-layers of alumina and copper and the investigations on the effects of temperature and time of sintering on the final thickness and homogeneity of the joining zone were conducted by scanning electron microscopy and energy dispersion spectroscopy (Philips, XL30 Series). To study phases' formation during metalized layer sintering, XRD test was taken on 9 same slurry solutions that were sintered in different conditions with different temperatures of 1000, 1470 and 1530?C and time durations of 40, 60 and 90 min using X-ray diffraction analysis (Philips, MPD-XPERT, λ: Cukα = 0.154 nm).

Fig. 2 indicates a diagram of the experimental procedure.

3. Results and discussion

3.1. Thickness and homogeneity

The effect of time and temperature of sintering and the thickness of applied layer on the leakage rate and the thickness of joining layer are listed in Table 1. As shown in Table 1, the layer thickness increases by increasing the holding time at each temperature with slow thickness growth rate.

When the thickness of the applied layer is 50 μm, the final thickness of the joining layer is larger than the thickness obtained with the use of the 25 μm applied layer. This is because interdiffusion can occur through 50 μm layer. Both Helgesson [4] and Takahashi and Okabe [10] mentioned that the applied layer thickness should be between 40 and 70 μm. Thin layers lead to weak joints and small width of the interface zone, and if the thickness is higher than the allowed limit, a high amount of applied layer would not get involved in the interaction with alumina, thus, a proper bonding would not occur between Mo–Mn and alumina.

Table 1 and Figs. 3–5 show the effect of temperature on the thickness of joining zone. As seen, the effect of