Development of a Novel Ni-Fe-Cr-B-Si Interlayer Material for Transient Liquid Phase Bonding of Inconel 718

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Abstract. A Ni-Cr-Fe-Si-B based interlayer material was developed by mechanical alloying (MA) process in a high-energy planetary ball mill. Equiaxed alloy powders of size 12 µm was obtained after milling for 50 hours. X-ray diffraction analysis of the milled powder revealed that milling of elemental powders initially resulted in microcrystalline alloy powder having face centered cubic structure, which on subsequent milling resulted in nano-crystalline alloy powder with a crystallite size of 3.2 nm. XRD analysis also reveals formation of metastable eutectic alloys resulting in lowering of the melting point of the interlayer material to 1025 °C. IN 718 superalloy samples were joined at 1050°C using the developed interlayer. A homogeneous joint was formed by the newly developed interlayer material. Three different zones were observed at the bond (i) isothermally solidified zone, (ii) diffusion affected zone and (iii) unaffected base metal. In the diffusion-affected zone, boron was present at the grain boundaries of Ni γ matrix in bulky metal borides form. The diffusion of boron from interlayer material into the base material was mechanism of isothermal solidification and bond formation in transient liquid phase bonding of IN 718.

Keywords: Transient liquid phase bonding, interlayer, IN718, diffusion

1. Introduction

Joining of nickel based super alloys is a challenge in liquid fueled rockets, land-based gas turbines and nuclear reactors [1]. Since during application, these are subjected to severe conditions of high temperature, mechanical and thermal fatigue loading, in addition to corrosive and oxidation environment, presence of even a micro-crack in the weld zone will leads to early failure of the joint[2,3]. Transient liquid phase (TLP) bonding is an emerging process for joining super alloys where an interlayer is sandwiched between the base materials and heated to 20°C-50°C above the melting temperature of the interlayer material. The interlayer melts and the individual elements diffuse across the material and subsequently solidify isothermally [4]. The success of the TLP bond depends on the bonding temperature and composition of interlayer material [5]. Several interlayer materials have been developed, but each having its own advantages and disadvantages. Now days BNi-2 interlayer material is used to join IN 718 by TLP bonding [6, 7]. However, the melting temperature of the interlayer is very high. This requires higher bonding temperature and may affect the microstructure of the base material. Researchers have tried nickel and phosphorous-based interlayer material. However, the presence of phosphorous has detrimental effect on the mechanical properties of the joint [8]. Therefore, in this present paper, a novel Ni-Cr-Fe-B-Si based interlayer material was synthesized using mechanical alloying process for joining IN 718 super alloy and characterized by using FESEM, XRD, TEM and DSC.
2. Preparation of Interlayer Material

The interlayer material should contain major elements of the base metal and at least one melting point depressant element. Nickel, chromium and iron are the major elements of the base material. Boron and silicon has high diffusivity in nickel. Hence, the interlayer elements consisted of Ni, Cr, Fe, B and Si. In IN 718, the amount of Ni is 50-55 weight percentage. Therefore, higher composition of Ni is taken in the interlayer material. And by studying the binary Ni-B and Ni-Si phase diagram, the composition of B and Si are selected to be 3.2 and 4.5 respectively. Ni Commercial powder from LOBA CHEMIE of Ni (99.8 % pure), Cr (99 % pure), Fe (95 % pure), B (99% pure), and Si (98.5% pure) of weight percentage 80.8, 10, 1.5, 3.2, and 4.5 respectively were taken as interlayer material. Powders of required quantity were blended and milled for 50 hours in an Insmart systems planetary ball mill. Hardened steel vial of inner diameter 150 mm and balls of 7.9 mm diameter was used keeping weight of ball to powder ratio as 10:1. Powder samples (2 grams each) were taken after 5, 20, 35, and 50 hours of milling for analysis. The morphological evolution and composition analysis for each mechanical alloyed brazing filler material was examined using a Zeiss make FESEM (Model: Sigma). Thermal analysis was carried out using a computerized Netzsch make, differential scanning calorimeter (DSC), Model: STA449F3A00. For thermal analysis, the powder mixture was heated to 1200 °C at a heating rate of 15 K in an Argon atmosphere and heat flux was measured by the software. Particle size was calculated in a Laser particle size analyser. In addition, structural characterization was done in a X-ray diffraction technique using Cu Kα radiation (λ = 1.54 Å) at a tube rating of 40 kV and 30mA in a commercial powder X-ray diffract meter.

3. Results and discussions

3.1 Phase formation for MA powders

Through mechanical alloying process, well-mixed non-equilibrium phases are formed due to the high impact of the powders by the balls. This results in generation of considerable amount of residual internal energy in the powder mixture. After milling for sufficient time periods, a well homogenized powder mixture is obtained. Figure 1 shows SEM micrographs of the powder particles milled for different time periods. During MA process the average particle size of the milled powder decreases considerably with time. The average particle size after 50 h of milling was approximately 12 μm with the agglomeration of smaller particles. The dominant mechanism during mechanical milling between 0 and 50 h involves fracture and cold welding. During MA process, the particles were work hardened, fractured via a fatigue mechanism and welded by cold welding process.

Figure 2 shows the XRD results of the milled powder mixture. At the start of milling (0 hours), the sharp peaks are the reflections from planes of Ni, Cr, Fe, B and Si. With increasing milling time, the peak corresponding to reflections from Si (111), (220), (311) and B (222) disappears indicating dissolution of Si and B in the Ni lattice. This results in the formation of non-equilibrium solid solution of Ni (Cr, Fe, Si, B) phase. This was confirmed by the decrease in the lattice parameter with milling time when milled up to 5 hours. After 5 hours of milling a continuous increase in the lattice parameter was observed indicating dissolution of Cr and Fe in the nickel solid solution. As the atomic radius of Cr and Fe are higher than Ni, the lattice parameter of the solid solution increases. The powder crystallite size was estimated from the full width at half maximum of the respective XRD peaks using the Williamson–Hall method. The peak broadening is due to size effect βs and lattice imperfections (micro-strain) βε. βs of Bragg reflection (h k l) originating from the finite size of the powder follows the Scherrer equation, βs = 0.9λ/d cos θhkl. Strain induced broadening βε, is determined by the Wilson formula [9], βε =4ε tan θhkl, where d is the crystallite size, λ is the wavelength of X-ray (0.154 nm), ε is the micro strain and θhkl is the Bragg’s angle. Considering the contributions of these two effects as mutually independent, the total peak width βhkl can be expressed as:

\[ β_{hkl} = β_s + β_ε = 0.9λ/d \cos \theta_{hkl} + 4\varepsilon \tan \theta_{hkl} \]  

(1)
Rearranging the equation (1) gives,

$$\beta_{hkl} \cos \theta_{hkl} = (0.9 \lambda /d) + \varepsilon (4 \sin \theta_{hkl})$$

(2)

The $d$ and $\varepsilon$ were estimated from plots of $\beta_{hkl} \cos \theta_{hkl}$ versus $4 \sin \theta_{hkl}$ for all related peaks. Figure 3 shows the Williamson–Hall plot of the milled powders. From the curves, micro stain and the mean crystallite size have been determined for different milling time. The plots of mean crystallite size vs. milling time and micro strains vs. milling time has been shown in Figure 4 (a) and Figure 4 (b). With increase in milling time crystallite size of the MA powder decreases. After 50 hours of milling, the crystallite size reduced to 3.2 nm. The micro strain calculated by Williamson–Hall method shows increasing pattern. This signifies, with increasing milling time the imperfections in the powder particle increases. This can be beneficial for a diffusion-controlled process.

Figure 1. FESEM micrograph of milled powders (a) 5 h, (b) 20 h, (c) 35 h, and (d) 50 h

Figure 5 shows the typical bright-field TEM (BF-TEM) micrographs and selected area electron diffraction (SAED) patterns of Ni (Cr, Fe, B, Si) alloy powders milled for 50 hours. The micrograph of as-milled powder confirms the existence of nano-grain structure. TEM micrographs also reveals agglomeration of fine-grain powders generally found in mechanical alloyed powder. The SAED patterns show the polycrystalline nature of the powders with reflections corresponding to (111), (200), (220) and (311) planes of Ni (Cr, Fe, B, Si) solid solution. The $d$ spacing of the alloy calculated from the SAED pattern by Image J software is in good agreement with the value calculated by XRD analysis.
Figure 2. X-ray patterns of the alloy powders milled for various time periods

Figure 3. Williamson–Hall plot of MA powders milled for various time periods
Figure 4. shows variation of (a) crystallite size and (b) micro-strain with milling time.

Figure 5. (a) bright field TEM micrographs and (b) SAED pattern of the Ni (Cr, Fe, B, Si) alloy powders milled for 50 h.

3.2 Calculation of melting temperature

Figure 6 shows the raw DSC scans (energy flow versus temperature) of 50 h milled powder heated at 15 °C/min from ambient temperature to 1200 °C. The DSC curve shows an exothermic peak and an endothermic peak at about 800 °C and 1025 °C respectively. The exothermic peak was due to crystallization of the amorphous powder formed during milling. The endothermic peak at 1025 °C is due to melting of alloy phase formed during MA process. The high-energy ball milling causes mixing at the atomic level resulting in the formation of complex non-equilibrium phases having low melting temperature. The melting temperature of the interlayer material is 25 °C lower than the commercially available BNi-2 filler material [9].
4. TLP bonding of IN718 using newly developed interlayer material

IN 718 samples of size $14 \times 10 \times 5 \text{ mm}^3$ were cut using wire EDM and the faying surfaces were grounded using SiC papers of grit size 600 and cleaned by acetone to remove surface oxides. The newly developed interlayer of thickness 50 $\mu\text{m}$ applied between surfaces. The assembly was kept in impedance heating furnace chamber and a pressure of 1 MPa was applied to the joint. The assembly was heated to 1050 $^\circ\text{C}$ and held for 1 h. This was subsequently furnace cooled to room temperature. Sample for metallographic observation of the bonded area was sliced using wire EDM. These were polished following the standard specimen preparation techniques and studied using the optical microscope. Figure 7 (a) and (b) shows the microstructure of the base metal and TLP bonded joint, respectively. The base material microstructure shows Ni $\gamma$ solid solution matrix, blocky MC carbides. Three different zones can be seen at the TLP joint: (i) base metal zone, (ii) bonded zone (isothermal solidification zone) and (iii) the diffusion-affected zone. The isothermally solidified zone shows homogeneous microstructure. The boron in the diffusion-affected area is present at the grain boundary of Ni $\gamma$ matrix in the bulky form in the form of metal borides. The EDS analysis of the DAZ is depicted in table 1, which shows diffusion of boron from bond zone into the base material. And the diffusion of B from the molten interlayer into the base material causes compositional changes in the bond zone. This increases the melting point of the interlayer material. When the melting point reaches bonding temperature isothermal solidification starts. The diffusion of B is the main cause of isothermal solidification in the TLP bonding of IN 718 super alloy.

| Elements | Ni  | Cr  | Fe  | B   | Nb  | Ti  | Al  | Si  |
|----------|-----|-----|-----|-----|-----|-----|-----|-----|
| Amount (wt. %) | 49.4 | 14.3 | 16.8 | 12.3 | 5.4 | 0.9 | 0.7 | 0.1 |
Figure 7. shows microstructure of (a) IN 718 base metal, and (b) TLP bonded IN 718

5. Conclusions
A novel Ni-Cr-Fe-Si-B based interlayer material was developed via MA process. An f.c.c. Ni (Cr, Fe, Si, B) solid solution powder mixture was obtained by mechanical alloying. 50 hours of milling resulted in alloy powders having crystallite size of 3.2 nm. The melting temperature of the interlayer material is 1025 °C, which is 25 °C lower than the commercially available interlayer material BNi-2. IN718 plates was successfully joined by transient liquid phase bonding using the developed interlayer material. The microstructural study of the bond region revealed three distinct zones: (i) bonded zone, (ii) diffusion affected zone and (iii) base metal zone. Diffusion of boron from interlayer to base material results in isothermal solidification during TLP bonding resulting in formation of borides at the diffusion-affected zone.

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