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Multipass cold rolling and resultant mechanical properties of a Cr-containing nickel aluminide

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The difficulties encountered in fabricating Ni_3Al-based intermetallic alloys into final structural components, due to their limited workability as a result of their inherent high yield strength and low ductility at elevated temperatures, are an important issue that have restricted the commercial applications of these materials. The Osprey spray deposition process is capable of delivering near-net-shape preforms, thereby avoiding the technical problems related to the hot working of these materials, e.g. hot rolling of slabs. The present work concerns an investigation of the cold rollability of a chromium-containing Ni_3Al intermetallic alloy produced with the Osprey process. The sliced preform with a thickness of 7 mm was successfully cold rolled through multipasses into sheets with a thickness of 0.7 mm and a good surface finish. The material has been found to have a high working hardening rate at room temperature. The maximum total reduction permissible without resulting in rolling defects is 30%. Thus, for larger reductions, intermediate annealing between rolling passes is necessary and it has been optimized to be at 1100 °C for one hour. The repeated cold rolling and the recrystallization occurring during intermediate annealing change the initial microstructural features and grain size of the Osprey-spray-deposited material. The cold-worked and annealed intermetallic sheets with a thickness of 0.7 mm have a yield strength of 570 and 730 MPa and a elongation value of 33 and 7%, at room temperature and at 700 °C, respectively. Fractography shows a transition from the transgranular fracture mode at low temperatures to the intergranular fracture mode at temperatures above 650 °C.

1. Introduction

The Ni_3Al intermetallic alloys containing boron, zirconium and chromium with improved ductility at room and elevated temperatures in an oxidative atmosphere are considered promising materials for high-temperature structural applications [1, 2]. The components in gas turbine engines and turbochargers are among those placed at the top of the application lists for these materials. However, viable techniques to convert these materials into the final products must be developed, before machine designers can accept these materials. In the gas turbine engines, for example, the materials must be in the form of sheet and is then joined to form a ready-to-use assembly such as a combustion chamber or an exhaust unit. At present, cost-effective fabrication of intermetallics into structural components with desired properties remains a challenge. Difficulties arise from the fact that the characteristic properties of these materials in resisting temperature and deformation work against the ease with which they can be processed. Their low ductility and being prone to brittle fracture, in particular, impose serious restrictions on usable deformation methods and process parameters. Moreover, because these materials do not derive strength from precipitation through ageing or from martensitic or bainitic transformation through quenching, but an ordered structure, narrow margins are left to enlarge the window of deformation processing by means of heat treatment.

It has been found that the conventionally cast Ni_3Al-based intermetallic alloys often do not meet strict requirements for demanding applications, because of severe segregation. While powder metallurgy can overcome the problem of macrosegregation and improve the properties and the consistency of the properties of the intermetallics [3, 4], it suffers from such shortcomings as multi-step processing involving the handling of fine powders which presents the potential of surface contaminations. Spray deposition, known as the Osprey process, can avoid the problems associated with both casting and powder metallurgy [5, 6]. An additional advantage of the Osprey process is its capability to produce almost densified preforms with required shapes and dimensions, which significantly lessens the need for subsequent processing to the final product. This advantage is of particular significance for the intermetallic

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alloys that inherently have high yield strength and low ductility over the usual temperature range of hot working. For instance, flat preforms produced with the Osprey process do not need to be hot rolled from slabs. They can be directly used as blanks for cold rolling into sheets with a needed thickness, which is a giant step closer to the final product.

In the present work, the cold rollability of an Osprey-spray-deposited Ni_3Al-based intermetallic alloy was investigated and the parameters of intermediate annealing for large reductions were optimized. Furthermore, the rolled material was characterized in terms of microstructural development and resultant mechanical properties as a function of testing temperature.

2. Experimental

The intermetallic alloy under investigation was based on the Ni_3Al intermetallic and contained chromium, boron and zirconium for the improvements of ductility, oxidation resistance and strength at room and elevated temperatures. The material in the form of preform was prepared by the Osprey Metals in Neath, UK, through remelting the ingots under argon, atomizing the melt with nitrogen and depositing the preform on a rotating substrate in a nitrogen atmosphere. The chemical composition of the as-received preform was analyzed and the results are given in Table I. The preform had a porosity level of 0.25%. It was sliced into plates with a thickness of 7 mm, a width of 20 mm and a length of 60 mm for multipass cold rolling experiments.

The sliced plates were incrementally cold rolled at a thickness reduction of about 0.35 mm per pass with a lab-scale Kerma rolling mill having a roll diameter of 86 mm. The response of the material to cold deformation was characterized by measuring its microhardness at different accumulated reductions so as to determine its work-hardening behaviour. It was further evaluated through tensile testing, on the assumption that the yield stress of the material was not sensitive to strain rate and that the stress-strain relationship in tensile testing corresponded to that in compression testing. The obtained tensile stress-strain curve was converted to the true stress-strain curve in order to obtain the mean yield stress in plain strain conditions using the von Mises criterion.

Intermediate annealing of the material rolled to the total thickness reductions of 12, 30 and 55% was performed at temperatures between 600 and 1200 °C for one hour in an argon atmosphere to search for an optimum annealing temperature. Softening as a result of recrystallization during annealing was measured with microhardness at a load of 200 g (1.98 N). With an optimized annealing temperature obtained, multipass rolling experiments were carried out till a final thickness of 0.7 mm was reached. The total reduction prior to each annealing was in the range of 27–32%. To avoid skidding of the material during rolling with the rolls having the small diameter, which would limit the reduction allowed per pass, no lubricant was applied. The maximum draught was determined in order to estimate the friction coefficient between the rolls and the material being rolled.

The microstructures of the as-spray-deposited preform and the rolled sheets were examined with a Neophote-2 optical microscope. The micrographic specimens were etched with a solution consisting of 86% acetic acid, 9% HNO_3 and 5% HF. Tensile properties of the rolled and annealed sheet material with a thickness of 0.7 mm were determined in the direction of rolling. The tests were carried out at both room and elevated temperatures (450–950 °C), in an ambient atmosphere and at an initial strain rate of $8.3 \times 10^{-4}\ s^{-1}$. Fracture surfaces of the tensile specimens were observed with a Joel JXA-50A scanning electron microscope.

3. Results and discussion

It was found that the spray-deposited intermetallic alloy indeed had a considerably high rate of work hardening. Fig. 1 shows the variation of the microhardness of the material with the total thickness reductions applied in multipass cold rolling. The maximum total reduction applied was 55% in the experiments. However, at total reductions above 30%, slight, local cracking on a micro scale already occurred and such a defect will be discussed later on in this communication. Because of work hardening, the material had an increasingly high yield strength and a reduced ductility, further cold working became more and more difficult, eventually impossible without resulting in cracks. When a workpiece with a

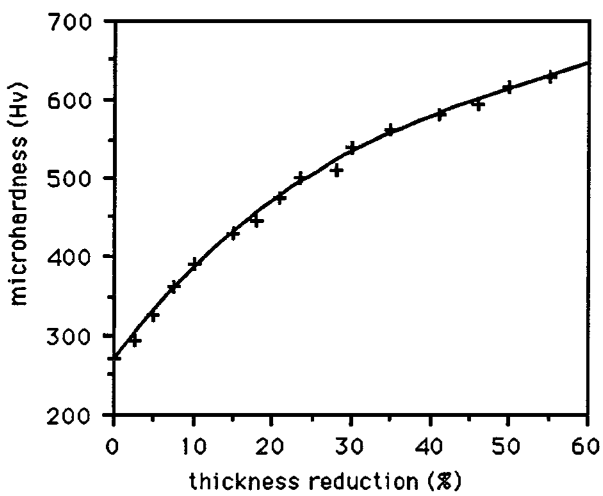

Figure 1 Microhardness as a function of total reduction applied during the cold rolling of the Osprey-spray-deposited Ni_3Al-Cr alloy.

TABLE I Chemical composition of the spray-deposited preform (wt %)

Element	Al	Ni	Zr	Cr	B	Si	N	C	S
Concentration	9.34	bal.	0.89	7.38	0.029	0.014	0.0120	≤0.005	≤0.005

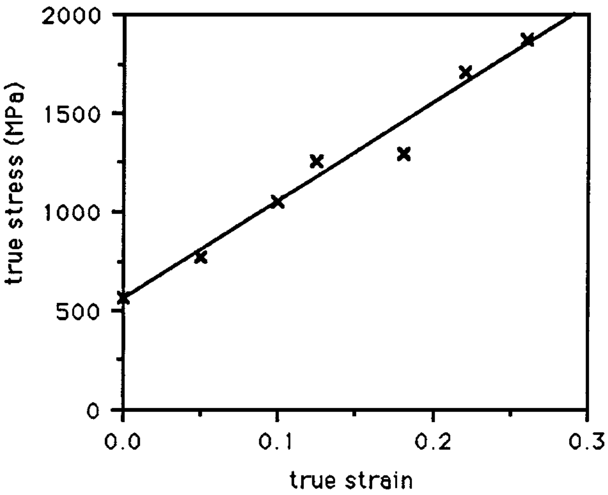

Figure 2 Relationship between the true stress and true strain obtained from tensile testing.

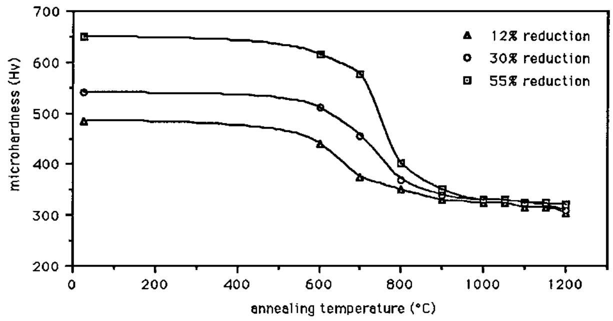

Figure 3 Variation of the microhardness of the material cold-rolled to 12, 30 and 55% total reductions with annealing temperature to derive an optimum temperature for intermediate annealing (annealing time: one hour, and atmosphere: argon).

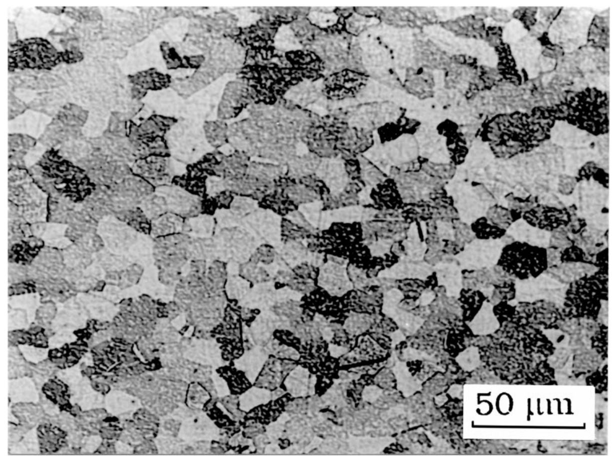

Figure 4 Microstructure of the material cold-rolled to a 30% total reduction and annealed at 1150 °C for one hour, showing the recrystallized, equiaxed grains.

thickness of 2 mm entered the last pass of rolling with a draught of 0.35 mm before reaching the final 30% total reduction, its stress would be about 1630 MPa, see Fig. 2. Thus, its mean yield stress in plane strain conditions would be as high as 1875 MPa. With the roll diameter being 86 mm, the roll pressure per unit width of workpiece could be estimated to be about 9.0 kN/mm by using the equation:

$$P_a/w = 1.2\sigma(R\Delta h)^{1/2}$$

where P_a is the roll pressure, w the workpiece width, σ the mean yield stress, R the roll radius and Δh the thickness reduction. If the correction for roll flattening [7] was considered, the roll pressure per unit width needed for the reduction would amount to 11 kN/mm. With such a high material yield stress, the minimum thickness T_{min} achievable would be limited to 0.27 mm under the present rolling conditions, as estimated by the equation:

$$T_{min} = 8\sigma\mu R/E$$

where μ is the friction coefficient between the roll and the workpiece, and E the Young's modulus of the rolls (which is about 210 GPa for steel rolls). Apparently, the pressure requirement to roll the intermetallic is much higher than that to roll other engineering materials such as aluminium alloys and steels, because the intermetallic has a higher yield stress and also a higher work-hardening rate. It must be noted that Fig. 2 is derived from tensile testing, which can only give an indication about the response of the material to deformation during rolling. Nevertheless, it can be seen that with a 10% strain the stress of the material will be doubled. With a 26% strain corresponding to a 30% reduction in rolling, its stress will be tripled. Therefore, for any further deformation beyond the total reduction of 30%, intermediate annealing is necessary.

After isothermal annealing of the rolled material at the temperatures of 600 and 700 °C for one hour, only marginal softening occurred. However, at the temperature of 800 °C or higher, the material became significantly softened. At annealing temperatures over 900 °C, the hardness of the 12, 30 and 55% rolled material converged at 330 Hv from the initial hardness values of 650, 540 and 490 Hv, respectively, as shown in Fig. 3. An annealing temperature higher than 1100 °C could hardly bring the hardness of the material further down.

Micrography showed that at the annealing temperature of 900 °C or higher, the material was completely recrystallized with the formation of homogenous, equiaxed grains, as shown in Fig. 4. However, with the increase of annealing temperature, a tendency of grain growth was observed, which was obviously not desirable for further deformation. Considering both the softening effect and grain size, the annealing temperature of the material was optimized at 1100 °C for one hour. In comparison with the annealing temperatures of other alloys, it is rather high, indicating a low grain boundary mobility of the material due to constrains imposed by its long-range ordered crystalline structure.

With a roll diameter being 86 mm, the maximum draught at which the rolls could accept the workpiece with no need to be pushed in was found to be 0.30 mm.

By using the equation:

$$\mu = (\Delta h/R)^{1/2}$$

the coefficient of friction at the interface between the rolls and the workpiece could be estimated to be 0.084.

This value falls in the range of friction coefficient from 0.04 to 0.4, corresponding to the variation of the rolling condition from well lubricated high-speed rolling to dry, rough rolling [8].

In cold rolling, shape factor h/L is an important index for deformation homogeneity (where h is the average workpiece thickness and L the length of the projected arc of the contact between the rolls and the workpiece). When the shape factor is smaller than unity, deformation is considered homogeneous across the section of a workpiece. When it is larger than unity, there exist local secondary tensile stresses on the workpiece in addition to the compressive stresses in the workpiece. Deformation inhomogeneity may lead to internal and external defects. Obviously, for a given roll diameter and a given draught, the shape factor is a function of workpiece thickness. Over the present range of the workpiece thicknesses from initially 7 mm to finally 1 mm, the shape factor varied from 1.42 to 0.17. According to the criterion of cold-rolling deformation homogeneity, it appeared that at the early stages of rolling (first passes) when the thicknesses of the workpieces were larger than 5 mm, rolling deformation was not homogeneous, since in this case the shape factor was larger than unity. Indeed, deformation inhomogeneity resulted in double barrelling on the cross section. During the subsequent rolling passes, the overhanging material was not compressed directly but forced to elongate by the neighbouring material close to the centre. The tensile stresses generated would lead to edge cracking if intermediate annealing was not applied at the intervals between rolling passes.

Another rolling defect, single barrelling at the edge of rolled sheets, was observed to occur, when workpieces were thinner than 1.5 mm, which was caused by lateral inhomogeneity in deformation. This is because when heavy deformation occurred across the thickness of a thin workpiece its centre expanded laterally more than its surface. When barrelling was severe, fine edge cracking took place. The edge cracks had a 45° angle relative to the thickness direction, indicating the tensile stresses in the longitudinal direction were the cause for cracking.

In addition, alligatoring happened to the material that had undergone a 35% total reduction and was given a further 5% reduction without intermediate annealing, as shown in Fig. 5. The large crack at the front edge of the rolled sheet could be attributed to too heavy prior deformation. Deformation inhomogeneity across the thickness, due to tensile stresses generated on the surface and compressive stresses in the centre, formed a torque couple, resulting in cracking. To prevent this defect from occurring, rolls with a larger diameter could be used, leading to a lower value of the shape factor so that homoge neous deformation could be created at the early rolling passes. For example, by replacing the present ϕ 86 mm rolls with ϕ 200 mm rolls, the shape factor would be lowered from 1.2 to 0.8 when a 5 mm thick workpiece was given a 7% reduction. On the other hand, a larger roll diameter would restrict the achievable minimum thickness of rolled sheets. In the experiments, auxiliary edge-restraining rolls to increase the degree of hydrostatic compression on the workpiece were found to be effective in preventing edge cracking from occurring. The sheets rolled in such a way were all bright, defect-free and of high surface quality.

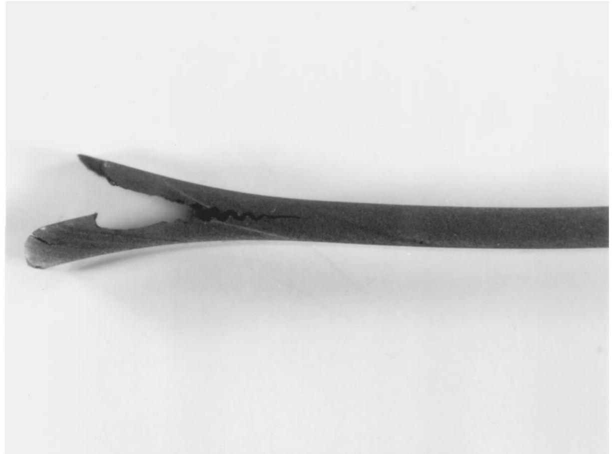

Figure 5 An example of alligatoring happening to the material which has undergone too heavy deformation without being annealed.

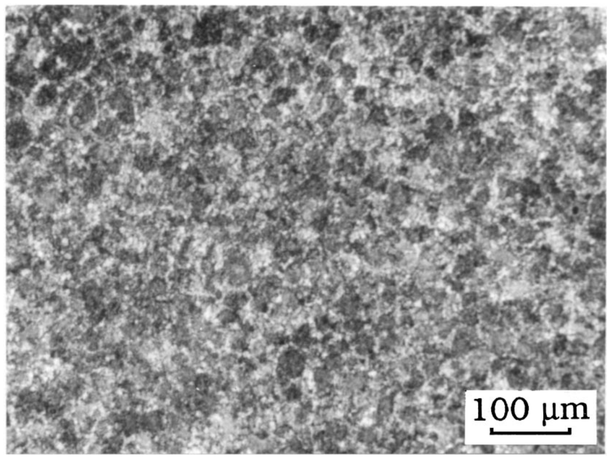

Figure 6 Optical microstructure of the as-spray-deposited intermetallic alloy showing equiaxed grains and almost full densification.

The as-spray-deposited preform had equiaxed grains with an average size of 40 μm, as shown in Fig. 6. Pores remaining in the preform were very fine and had a typical size of 5 μm. After the intermetallic underwent repeated rolling with the reduction of thickness from initially 7 mm to finally 0.7 mm (i.e. a 90% total reduction for which 18 passes were needed) plus intermediate annealing at 1100 $^\circ$C for one hour, the pores were fully closed. The microstructure of the rolled and annealed material is shown in Fig. 7. The microstructural features of the preform resulting from rapid solidification involved in the Osprey process, namely a bimodal distribution of the γ/γ' network structure with individual daisy-like flower patterns [6], disappeared. The ordered γ' phase and the disordered γ phase became uniformly distributed, see Fig. 7. The final sheet material had an average grain size of about 18 μm.

The tensile properties of the rolled and annealed intermetallic alloy at room temperature are given in Fig. 8 (see the value labels at the testing temperature of 25 $^\circ$C). It must be noted that the tensile elongation at fracture is as high as 33%, which is obviously contributed by the

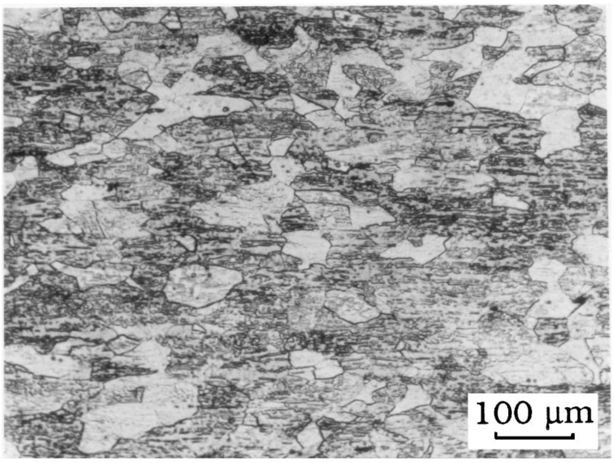

Figure 7 Microstructure of the material having been cold rolled to a 90% total reduction (in the rolling direction).

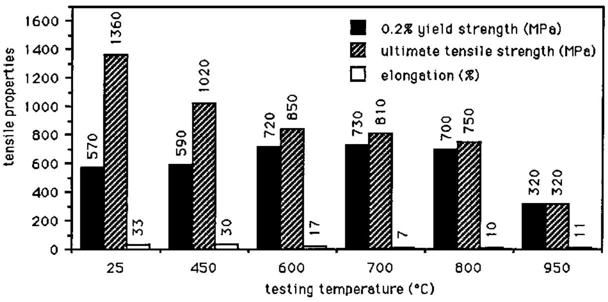

Figure 8 Tensile properties of the material having been cold rolled to a 90% total reduction and annealed.

0.03% boron addition to the base Ni_3Al alloy. Fig. 8 also gives the tensile properties of the rolled and annealed material at elevated temperatures. It is apparent that the material exhibits a characteristic correlation of yield strength against temperature; while the ultimate tensile strength monotonously decreases with rising temperature, the 0.2% yield strength increases to a peak of 730 MPa at 700 °C and then decreases. This may be attributed to a thermally activated change in the slip systems of the material resulting in additional constrains for dislocation motion [9]. The tensile elongation of the rolled material declines slightly when temperature rises to 450 °C and then hits a minimum at 700 °C. It must be noted that in the present investigation the tensile tests were performed in air with a possibility for dynamic oxygen embrittlement to occur. The 7% elongation of the material at 700 °C is actually quite high. This is obviously contributed by the 8% chromium addition to the base Ni_3Al alloy, which promotes the rapid formation of a protective chromium oxide film to prevent oxygen from entering the material along grain boundaries.

Fractography revealed a lack of grain-boundary facets on the surface of the material fractured at room temperature, suggesting that the fracture mode was mainly transgranular, as shown in Fig. 9. The fracture surface was observed to be about 45° relative to the tensile direction, corresponding to the plane of the maximum shear stress. These ductile fracture features are consistent with the measured tensile elongation value of 33% at room temperature. Such a fracture mode

Figure 9 Fracture surface of the material tested at room temperature showing the transgranular fracture mode.

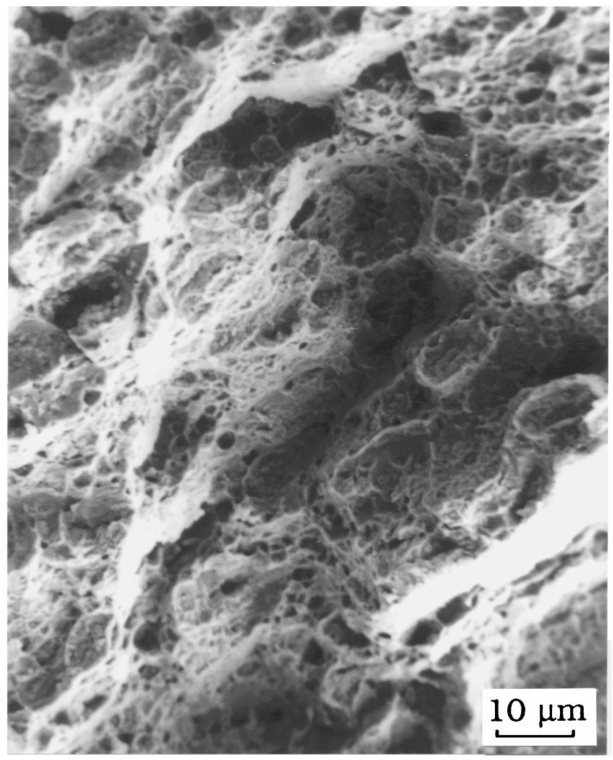

Figure 10 Fracture surface of the material tested at 600 °C showing dimples and also intergranular facets as a result of fracture in a mixed fracture mode.

persisted in the material fractured at temperatures up to 600 °C, as evidenced by equiaxed dimples on the fracture surface (Fig. 10). These dimples indicate that fracture occurred through the coalescence of transgranular microvoids, which were initiated at existing internal structural discontinuities such as prior porosity and

Figure 11 Fracture surface of the material tested at 900 °C showing the typical intergranular fracture mode.

inclusions. As local stresses increased, the microvoids grew, coalesced and finally formed a continuous fracture surface. It was also noticed that at the testing temperature of 600 °C, intergranular fracture started to take part in the fracture process, resulting in a mixed mode of fracture on the fracture surface. With tensile testing temperature increased over 700 °C, the fracture mode changed from transgranular to completely intergranular. Fig. 11 shows the fracture surface of the material tested at 950 °C. Brittle fracture along the grain boundaries is consistent with the fracture surface being perpendicular to the tensile direction and the reduced elongation value at this tensile-testing temperature.

4. Conclusions

1. The spray-deposited Ni_3Al-Cr intermetallic alloy can be cold rolled through multipasses up to a maximum total reduction of 30% without cracking. It has a high yield stress and work hardening rate. However, further cold working is still possible if the material is subjected to intermediate annealing.

2. Intermediate annealing has been optimized at 1100 °C for one hour to remove working hardening through recrystallization. A further increase of the annealing temperature results in grain growth.

3. With the optimized intermediate annealing, a workpiece can go through 18 passes of cold rolling with its thickness being reduced from initially 7 mm down to 0.7 mm (a 90% total reduction). The rolled sheets are bright, defect-free and of high surface quality. However, an inadequate annealing parameter or a too large reduction or a too high shape factor will lead to such defects as barrelling, edge cracking and alligatoring.

4. The as-spray-deposited intermetallic has homogenous, fine grains. After the material is cold rolled from 7 mm thick plates through multipasses into 0.7 mm thick sheets and annealed at 1100 °C for one hour, the initial microstructural features resulting from rapid solidification involved in the Osprey process are removed. The ordered γ' phase and the disordered γ phase become uniformly distributed. And the average grain size is reduced from 40 to 18 μm.

5. The rolled and annealed intermetallic alloy has a yield strength of 570 MPa and an elongation value of 33% at room temperature, as contributed by its homogeneous, fine-grain structure and the presence of boron. Its yield strength becomes even higher at 700 °C with a relatively high tensile ductility. Fracture mode changes from predominantly transgranular ductile fracture at low temperatures to intergranular brittle fracture at temperatures above 650 °C. Consistently, over this temperature range, the tensile ductility is decreased from 33 to 7%.

6. The spray-deposited Ni_3Al-Cr intermetallic alloy exhibits a quite reasonable cold rollability and excellent mechanical properties, which enhances its acceptability as a promising sheet material for high-temperature applications.

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