Cellular materials made of stacked tubes: influence of the manufacturing process on the dynamic behavior of the constitutive material. part I: microstructure

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Part I: microstructure

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Abstract
Cellular materials are very promising as lightweight aeronautical frames thanks to their superior specific mechanical properties such as impact resistance. However, because of the processing routes and heat treatments used in the production process, the material within the cell walls may behave differently from the bulk, and therefore the in situ mechanical properties are often unknown. The present work aims at investigating the link that exists between the processing of cellular architectures and the mechanical properties of their constitutive material. The cellular material studied is an Inconel® 600 tube stacking brazed together using a nickel-phosphorus alloy. The experimental works have been conducted in order to analyse the microstructures of Inconel® 600 tube stacking resulting from brazing and annealing heat treatment. In order to discuss the influence of the manufacturing process of tube stackings on the mechanical properties of their constitutive material, it has been proposed to perform uni-axial tensile tests on tubular specimens. Different material configurations (with or without heat treatments, with or without nickel-phosphorus coating) have been thus characterised and compared, involving both tensile tests from quasi-static to dynamic loads and electron back-scattered diffraction analyses (EBSD) performed on post-mortem specimens.

1. Introduction

High-temperature Ni-based brazing alloys was used for year in aerospace industry to bond aircraft structural parts, acoustical tail pipes, thrust reversers, turbines blades and seals. They provide good resistance to chemical corrosion and oxidation coupled with high strength at elevated temperatures. Vacuum brazing or controlled atmosphere brazing are the most common heating methods used. Typical temperatures of brazing are in the range of 890-1170°C. Alloys are available in electrochemical coating, powder, paste form, and melt spun foils or tape. A technic for fabricating a cellular structure closed from the honeycombs one consists of stacking and brazing hollow tubes in a compact arrangement. Such a structure made in metallic hollow tubes stacking is being considered for applications where multifunctional properties are required including structural properties. In the field of aeronautic industry, this kind of lightweight structure could provide solution for vibration damping, heat dissipation and storage, and crash energy absorption. The integration of metallic hollow tubes stacking as an engine component that will be routinely subjected to operating temperatures up to 700°C involved to achieve the brazing at high temperature in order to avoid the melting of the joints in use. As the behaviour of the material is governed by the amount of cold work, grain size and chemical composition, the purpose of this of this work is to study the impact of the brazing heat treatment on the microstructure of the tubes, and consequently, on their final mechanical properties.

2. Cellular material's processing route

Cellular materials of interest in the present work are brazed tube stackings. They are manufactured using tubes made of Inconel® 600 which are prior coated with a 50 µm thick Nickel-Phosphorus (NiP) alloy layer for the brazing. Inconel® 600 alloy is a standard nickel-based superalloy used for applications which require resistance to corrosion at high temperatures. Being not precipitation hardenable, this alloy can be hardened and strengthened only by cold work. Tubes, having external and internal diameters of 5 mm and 4 mm respectively, are stacked up regularly between two metallic skins to produce the desired structures (e.g. sandwiches with either a square stacking core or a hexagonal one as shown in Figure 1). Skins are made of Inconel® 600 too and their thickness equals 1 mm. A low-melting coating (nickel phosphorous alloy) deposited on the surface of each tubes by electroless plating was used for brazing. An electroless nickel bath which contains nickel sulfate as a Ni ion source and sodium hypophosphite as a reducing agent was used to coat a 50µm thick nickel-phosphorous NiP layer on the surface of each tubes. Before immersion in the electroless NiP deposition bath, the tubes were etched in nitric acid to remove the native oxide layer. This bath provides NiP coating with a medium phosphorous content closed to the eutectic concentration, namely 19 at.% P. As the tubes are not hermetically sealed at the ends, the coating occurs in the inner surface of the tubes. The tubes were stacked in a graphite die lined with inconel® plates positioned at the top and at the bottom of the stacking in order to provide a sandwich structure. A close-packed arrangement of tubes is naturally created and provides “hexagonal stacking”. A square stacking can also be obtained by tilting the mould during brazing. The bonding is achieved by applying a thermal treatment in a vacuum furnace. According to the Ni-P phase diagram, a medium phosphorous content alloy would be liquid above 1000 °C. As the liquid concentrates at the contact lines by capillarity, metallurgical joints are created between the tubes. In this study, a “soft” brazing heat treatment has been performed in order to avoid microstructural evolutions of the Inconel® 600 tubes and in particular grain growth. It consists in a progressive heating under vacuum (with a heating rate of 100°C/min) until 1000°C. Then the heating temperature is kept constant during 15mn. The heat treatment ends by a natural cooling down to room temperature.

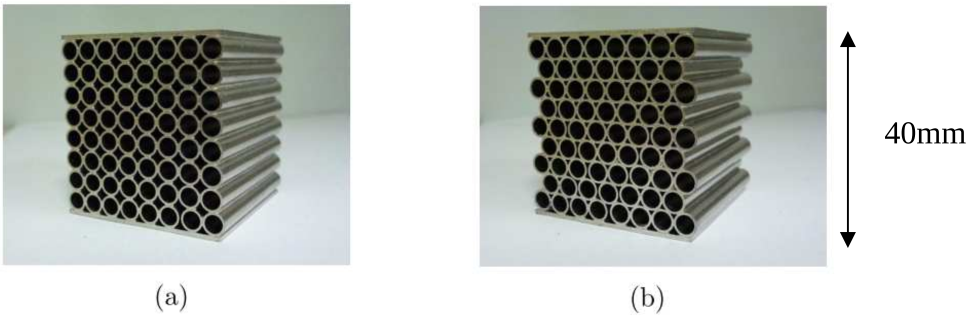

Figure 1: Arrangement of hollow tubes for fabricating cellular structure (a) square and (b) hexagonal stacking.

In order to provide a strong bond between the tubes, and between tubes and the sheet at the top and at the bottom of the sandwich structure, it is often recommended to increase the brazing duration to enhance the diffusion phenomenon and to improve the braze metal penetration into base metal. Some samples have been annealed under vacuum during 16 hours at 1050°C. A detailed scanning electron microscopy (SEM)-based microstructural and chemical characterization of the sandwich structure has been carried out to observe the effect of this annealing heat treatment. For these eight tubes wide and eight tubes high sandwich structures, the brazed joint width was around 1.5 mm. Braze joints was composed of a P-rich central area corresponding to the phase that remains liquid at the brazing temperature. During the cooling down to room temperature, the complete solidification of the remaining liquid phase results in the formation of the particular microstructure which consists of two distinct phases: a Ni-rich one embedded in an intermetallic matrix Ni_3P. At the interface of the joints and the tubes, some Ni-rich grains resulting from a local depletion of phosphorus in the liquid phase appeared. The whole grains formed a layer, appearing in figure 2 (a), and whose thickness and composition depend on the heat treatment. Moreover, the grains in the tubes are visible thanks to electron channeling phenomenon. The electron channeling contrast imaging makes use of the strong dependence of the backscatter electron (BSE) signal on the orientation of the crystal lattice planes with respect to the incident electron beam due to the electron channeling mechanism.

(a) (b)

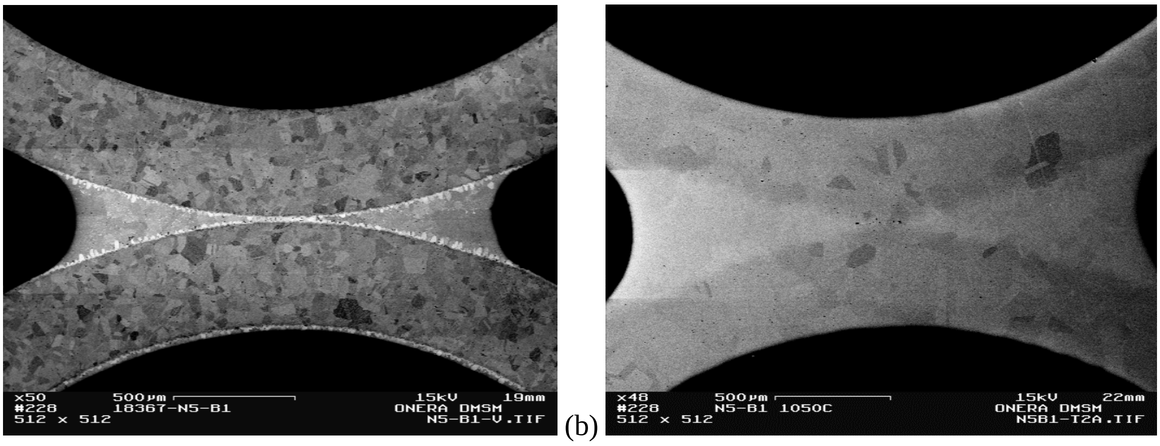

Figure 2: SEM of a brazed joint between two neighbouring tubes; (a) without annealing treatment; (b) with annealing treatment.

X-ray analyses were performed in order to have access to chemical distributions (figure 3). The phosphorous distribution shows clearly the location of the Ni_3P phase in the joint area. During heat treatment, the tubes are progressively enriched in phosphorus coming from the liquid phase. But our observation did not reveal a deep penetration of phosphorus element into the tubes walls, even with annealing treatment (figure 3 (a)). Indeed, phosphorus diffuses up to its limit of solubility in nickel around 0.32% (atomic percent) at 1000°C according to the binary Ni-P diagram. As this solubility is very low, phosphorous appears clearly in the Ni_3P phase where its concentration is around 33% (atomic percent). During brazing and annealing heat treatments, the diffusion of alloy elements of Inconel® 600 such as chromium and iron took place from the tubes to the joint area. Elemental distribution shows a heterogeneous composition of these two elements in the joint area. Indeed chromium and iron diffusion took place in the interfacial Ni-rich layer and decreased in the Ni_3P phasis as highlighted by low chromium and iron contents in the Ni_3P phase. According to the homogeneity of composition, especially in the case of annealing treatment, the diffusion of iron element into the interfacial Ni-rich layer seems to be faster than the one of chromium element. The comparison of the element distributions with and without annealing treatment (figure 3 (a) and (b)) shows that the annealing treatment allows grain growth of the interfacial Ni-rich layer and creates a Ni-based junction between the tubes. The interfacial Ni-rich layer appears thicker and richer in chromium content after annealing treatment. It creates a strong Ni-based junction which could avoid premature joints failure into the brittle Ni_3P phase.

(a)

(b)

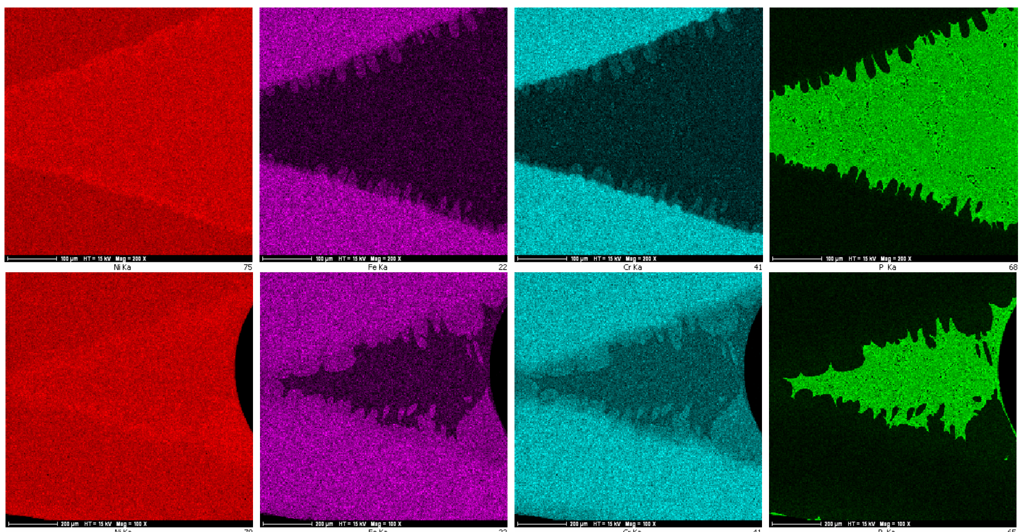

Figure 3: Elemental distributions according to the X-ray mapping (Ni, Fe, Cr, P); (a) without and (b) with annealing treatment.

3. Tensile tubular specimens

In order to analyse the influence of the manufacturing process on the mechanical properties of the Inconel® 600 base material, it is proposed to compare the mechanical behaviour of tensile tubular specimens under uni-axial tensile loading at different rates of strain for different material configurations. The machined specimen in Figure 4 has been developed to avoid plastic strain location and failure near the gripping areas and to make the analysis of the tube material behaviour possible under a wide range of average strains and strain rates. Tubular samples are made from 75mm length tubes with external and internal diameters equals to 6mm and 4mm respectively produced by TEXTPART using extrusion and cold-drawing. The external diameter in the center of the specimen corresponds to the external diameter of the tubes implemented in the cellular materials in Figures 1 and 2 ie 5mm. The table 2 gives the process steps of the different analysed specimens.

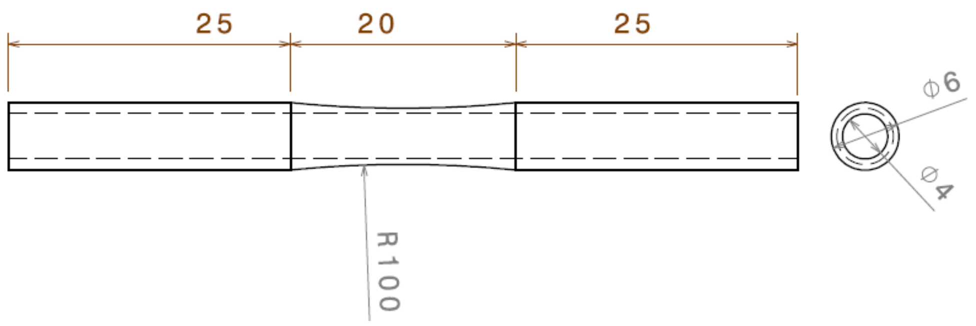

Figure 4: Dimensions of the tubular specimen for tensile tests (in mm)

Sample	Machining	NiP coating	Brazing heat treatment	Annealing	Loading
Inco600M	X				
Inco600BM	X				X
Inco600HT	X		X		X
Inco600NiP	X	X	X		X
Inco600HT-A	X		X	X	X
Inco600NiP-A	X	X	X	X	X

Table 2: Process steps of the tensile tubular specimens for EBSD Analysis.

This is particularly important that the tensile tubular specimens and the tubes within the cellular materials have a similar diffusion and penetration of brazing metal (Ni-P alloy layer) into the Inconel® 600 base material. The set of Inco600NiP specimens has been coated by 15µm Ni-P alloy layer. This thickness has been calibrated in order to produce a similar diffusion and penetration of the brazing metal into the base metal, compared to the actual manufacturing process. As the cellular materials are being subjected to possible dynamic loads (i.e., fan blade off application), the mechanical behaviour of the tube materials is also studied under different displacement rates to analyse possible strain rate effects.

4. Analysis of post-mortem specimens

4.1. Experimental procedure

In order to investigate the evolution of the microstructure resulting from dynamic load, Electron Backscattered Diffraction (EBSD) Spectroscopy analysis was carried out on post-mortem samples. All samples were cut mechanically on the longitudinal section of the tube, in a plane that passes through the tube axis. Then the cut samples were cold-mounted in epoxy and polished. The material configurations were analysed in order to study the effect of heat treatement and phosphorus on the mechanical properties of Inconel® 600 base material. The microstructure of low-speed loaded (6 $mm.min^{-1}$) and high-speed loaded ($1 m.s^{-1}$) post-mortem samples were also compared (Table 1).

Material configuration	Load conditions	Name of the post-mortem specimen
Inco600BM	6 $mm.min^{-1}$	Inco600BM-6
	$1m.s^{-1}$	Inco600BM-1
Inco600HT	6 $mm.min^{-1}$	Inco600HT-6
	$1m.s^{-1}$	Inco600 HT-1
Inco600NiP	6 $mm.min^{-1}$	Inco600NiP-1
	$1m.s^{-1}$	Inco600NiP-6
Inco600HT-A	$1m.s^{-1}$	Inco600HT-A-1
Inco600NiP-A	$1m.s^{-1}$	Inco600NiP-A-1

Table 1: Material configurations and load conditions of the analysed post-mortem specimens

Two areas of interest were compared: the fracture area (A), and a non-loaded area (B) (figure 5). EBSD patterns were collected from a Scanning Electron Microscope (SEM) Zeiss DSM 960 to map the crystallographic orientations distribution. The scanning was performed in order to put Transversal Direction (TD) parallel to the load direction, ie. the tubular specimen axis. The Normal Direction (ND) is normal to the specimen's surface. Two categories of Bravais unit cells are present: the wall in Inconel®600 and the Ni-rich grains in the brazing layer crystallized in a cubic unit cell (Inconel® 600 has a face-centered cubic structure as pure nickel), while Ni_3P matrix of the brazing layer crystallizes in a quadratic system. A large area of 500 x 500 μm^2 with 0.5 µm steps at the center of the tube wall was investigated.

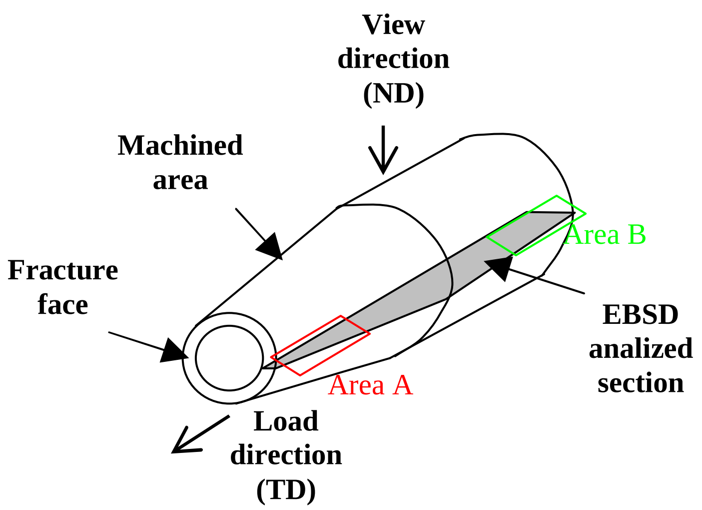

Figure 5: Schematic illustrations of the analysed plans into a cut of a post-mortem sample: areas of interest in a section of a post-mortem sample: the fracture area (A), and a non-loaded area (B).

4.2. Microstructures resulting from dynamic load

The EBSD analysis in the loaded areas of all post-mortem tubular specimens revealed typical grain morphology of high-strained material illustrated in figure 6: the grains are elongated in the loading direction, ie. in the tubular specimen axis. The deformed area spreads on few mm^2. The figure 6 shows the typical orientation mapping referring to TD and ND, and the corresponding pole figure for the Inco600BM-1 tubular specimen. Textures develop from the rotation of slip systems in grains during this plastic deformation. Comparing with that a non-loaded specimen (Figure 7), it is seen that there is a strong texture development in tube wall due to significant plastic deformation. This texture is observed both in TD and ND. The orientation mapping referring to TD shows that there are two preferential orientations of the cubic unit cell: major <111> in blue, and minor <100> in red as given by the corresponding key color. It means that, in the face-centred cubic cell, either the diagonal or the edge is parallel to TD. These two preferential orientations are also highlighted by the direct pole figures. In 001-axis and in 111-axis, the two pole figures show intensity peaks in the directions closed to TD. The orientation mapping referring to ND also reveals a texture. It appears that the grains are mostly in green: the <101> is preferentially parallel to ND. As the specimens have been cut in a plane that passes through the tube axis, and as ND is normal to the specimen surface, ND is a particular direction in the tube.

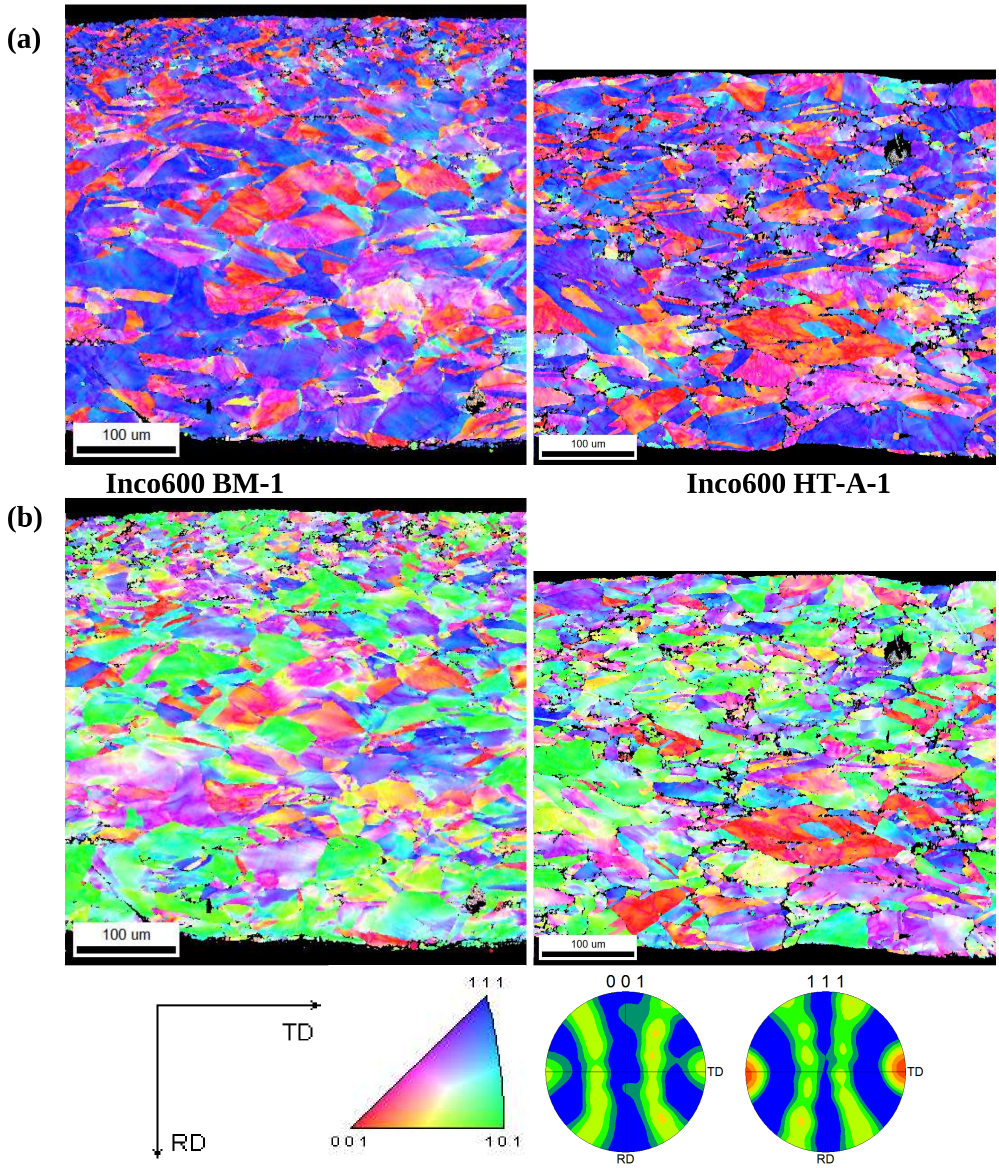

Figure 6: EBSD analysis of a Inco600 BM-1 and Inco600 HT-A-1 tubular specimens in the loaded area A; orientation mapping referring to (a) TD axis direction and (b) ND axis direction; the corresponding key color and the direct pole figure.

EBSD analysis highlighted heterogeneity of grain size in the wall thickness in the Inco600 BM-1 specimen. The figure 7 shows the orientation mapping of a non-loaded Inco600BM tubular specimen in the central area. The corresponding pole figures show that no texture is present before loading. The grains appear twinned. The analysed areas, chosen to cross the wall thicknesses, highlight that the grains are finer at the inner surface of the tubular specimens. Grain size is dependent on processing. The initial tubes were manufactured through a mechanical cold-drawing process followed by annealing step. They are drawn through a mould using a guide bar to keep the tube straight and give the inner diameter after drawing. The outer diameter and wall–thickness of the tube are reduced and a significant plastic deformation occurs in material at the end of drawing. Grain size reduces quickly through the sequence of cold drawing and annealing. A heterogeneous plastic deformation during manufacturing process could explain the heterogeneity of grain size in the wall thickness of the tubular specimens. In general, material with a fine grain structure is preferred because it has better corrosion resistance and higher tensile, fatigue and impact strength.

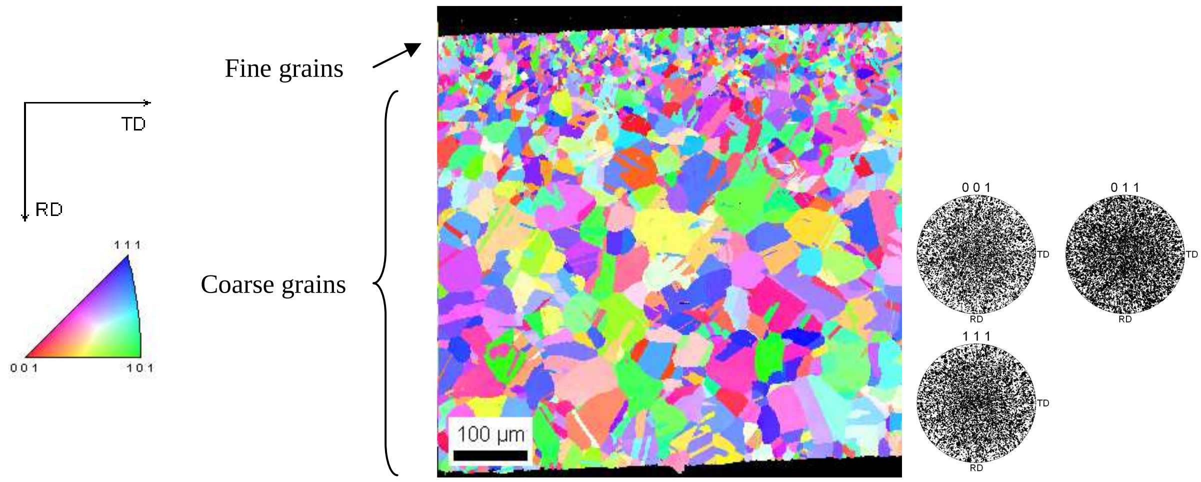

Figure 7: EBSD analysis of a Inco600BM tubular specimen in a non-loaded Inco600BM tubular specimen in the central area: Orientation mapping referring to TD axis direction and key color; the corresponding pole figure.

EBSD analysis highlighted no heterogeneity of grain size in the wall thickness in the Inco600 HT-A-1. The homogeneous grain size can be explained by grain growth of the finer grains localized at the inner part of the wall occurring during the annealing treatment. For Inconel® 600, grain growth occurs because of carbides coalescence above 980°C. At that temperature, the finely dispersed carbide particles in the alloy's microstructure, which inhibit grain growth, begin to coalesce. It means that the time and temperature of the annealing treatment (16 hours at 1050°C) is enough to dissolve the carbides completely and results in increased grain size. rate of local work hardening in inner wall of tubular sample is enough to recrystallize the microstructure during brazing step, and annealing treatment provide an homogeneous grain size into the wall of the tubular samples.

4.3. Local strain characterization using EBSD intragranular misorientation measurements

The state of residual strain in materials can be investigated by EBSD. The technique provides high spatial resolution, strain sensitive examination of plastic strain, enabling analyses that are otherwise difficult given the spatial / angular resolution and sampling limitations of XRD and TEM, respectively. The angular spreads into a grain were quantified. Figure 8a) gives the maps of grain orientation spread of some high-speed loaded tubular specimens ($1 m.s^{-1}$) in the loaded area: Inco600BM-1, Inco600NiP-1 and Inco600NiP-A-1. A grain was defined as a group of at least five neighboring indexed points having the same crystal orientation within a tolerated angular deviation of 5°. Intragranular misorientation measurements by EBSD capture strain manifested as a continuous change in local lattice orientation across grains of deformed materials, with the gradient of deformation a function of dislocation density and degree of dislocation order. Within each grain, the point with the lowest misorientation gradient (i.e. least deformed) is calculated. This is taken as a reference orientation, and the misorientations to all other points in the grain are calculated. The blue color indicates where grains have very little internal misorientation. The green, yellow, orange and red colors represent progressively increasing levels of internal misorientation. Grains are outlined with black boundaries. Such maps clearly show the most strained grains. The same colour scale, from 0 to 25° misorientation is used for the three analyses. It appears that three high-speed loaded tubular specimens are in the same state of residual strain. A majority of grains are colored in green. In the case of the specimen Inco600NiP-1, the grains colored in blue at the border of the wall belong to braze phases. As Ni_3P is brittle, the braze layer breaks during loading, and few level of residual strain is observed. For the specimens Inco600BM-1 and Inco600NiP-1, the grains located at the inner surface of the tubular samples appear in blue because of their small grain size. In fact, the plastic residual strain level is locally high as revealed by the Kernel Average Misorientation (KAM) maps given in figure 8b). The KAM is defined for a given point as the average misorientation of that point with all of its neighbors, which is calculated with the proviso that misorientations exceeding a tolerance value of 5° are excluded from averaging calculation. The magnitude of the KAM is affected by the scan step size, which was always set as 0.5 µm in the present paper. The figure 8b) show that the average misorientation in the grains located at the inner surface of the tubular samples of the specimens Inco600BM-1 and Inco600NiP-1 are higher than the rest of the wall. The maps of Kernel Average Misorientation and Grain Orientation Spread of the Inco600NiP-A-1 reveal a lower and homogeneous residual plastic strain, due to the annealing treatment.

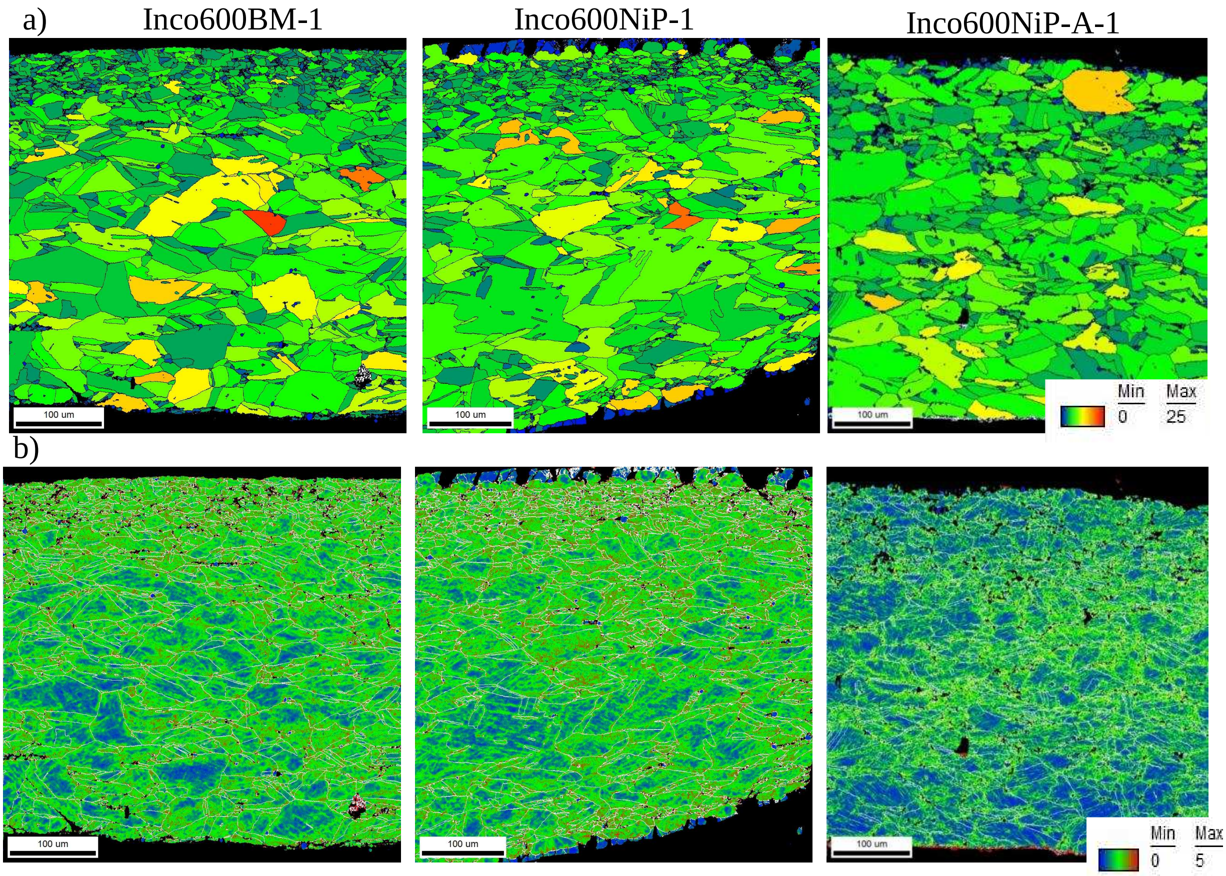

Figure 8: a) Maps of GOS (Grain Orientation Spread) of three high-speed loaded tubular specimens in the loaded area; b) Kernel Average Misorientation (KAM) maps.

5. Conclusion

A SEM analysis of tubular stacking has been conducted in order to explain the improvement of mechanical strength through annealing treatment (16h at 1050°C). During annealing, grain growth allowed to create a Ni-based junction between the tubes and to avoid premature joints failure. A systematic study of the microstructure of Inconel® 600 alloy loaded under dynamic condition has been thoroughly investigated as a function of the initial state of tubular samples with particular emphasis on local strain characterization using EBSD intragranular misorientation measurements. Based on these studies, the follow conclusions are summarized:

- Mechanical properties of the tubes depend on their manufacturing process (residual work hardening);
- A strong texture develop in tube wall due to significant plastic deformation;
- Texture and residual strain level are not affect by the initial configuration or the load speed;
- Because of its brittleness, the local strain in the braze layer is low;
- Annealing treatment provides a homogeneous grain size and residual plastic strain into the thickness of the wall of the tubular samples.