Solidification of undercooled Al-Fe-Nd alloy

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Abstract. *Rapid Solidification Processing, of metals and alloys, is establish by increasing of the undercooling applying high cooling rates (10^2 - 10^6 K/s) or by reduce nucleation sites using low cooling rates (1 K/s). Melt undercooling opens new solidification pathways for new non-equilibrium phases and unusual microstructures. Several techniques have been developed to reduce nucleation sites and produce increased undercooling in metals and alloys including the fluxing technique. In this work, an $Al_{96}Fe_2Nd_2$ alloy was solidified, by fluxing technique, and its microstructure and microhardness investigated as a function of the undercooling level. The increasing undercooling level from 30 K to 109 K promotes change in morphology and on the microhardness of the $Al_{96}Fe_2Nd_2$ alloy.*

Keywords: Al-Fe-Nd alloy, undercooling, microstructure, microhardness.

1. Introduction

Rapid solidification processing provides a typical case of nonequilibrium solidification, therefore microstructure refinement, solid solubility extension, and metastable phases formation will take place under this condition (Lu, Cao and Wei, 2001). Traditionally, rapid solidification is achieved by employing rapid quenching techniques, such as splat quenching, melt spinning, laser surface melting, and gas atomization, etc. The principle of these methods is the rapid removal of molten melt heat; the disadvantage is that the specimen is usually small in one dimension, leading to the difficulty in directly observation of the nucleation and growth phenomena. Therefore, researchers have paid their attention to others ways for rapid solidification (Perepezko and Uttormark, 1995). The others ways for rapid solidification are the techniques that are used for high undercoolings under low cooling rates (1 K/s), in order to reduce nucleation and to produce high undercoolings for metals and alloys, for example fluxing technique. In this technique the liquid is immersed in a material that isolates it from contact with the crucible walls and atmosphere, dissolving impurities or changing structures to make them less active. It also doesn't provide heterogeneous nucleation, which can increase the undercooling degree (Kelton, 1991).

The finding in 1988 (Inoue *et al.*, 1988) of aluminum based amorphous alloys with an Al content above 80 at.%, which exhibited tensile strengths of over 800 MPa as well as good ductility, generated great interest in the development of other similar systems. Since then, Al amorphous alloys have been produced by rapid solidification in the Al–RE, Al–ETM–LTM and Al–RE–TM systems (RE=Rare Earth, ETM and LTM=Early and Late Transition Metals, respectively). From a technological point of view, the interest is focused on the development of these high glass-forming-ability Al alloys in a bulk form that could be obtained by mass production techniques as inert gas atomization followed by a conventional consolidation process such as hot extrusion. On the other hand, from a scientific point of view, the interest in these Al-base alloys is to reach a better understanding of their glass forming ability as well as the mechanisms and kinetics which control the microstructure development upon heating, and which may give rise to the formation of nanocrystals, quasicrystals and other unknown metastable and stable phases (Cardoso *et al.*, 2001).

The aim of this research was to verify the influence of the increasing undercooling level on the solidification microstructure and microhardness of $Al_{96}Fe_2Nd_2$ alloy using the by fluxing technique.

2. Experimental Procedure

Samples of the $Al_{96}Fe_2Nd_2$ alloy were prepared from pure Al, Fe and Nd elements by arc melting under argon atmosphere. The as-prepared samples were characterised by scanning electron microscopy (SEM) with energy dispersive spectroscopy (EDS), differential scanning calorimetry (DSC). By using B_2O_3 as fluxing agent, the undercooling experiments were performed in 2 g sample in a quartz crucible with an apparatus which was described in more details elsewhere (Castro *et al.*, 2001).

A cromel-alumel thermocouple was used to record the cooling curve and detect the starting point of solidification. Cooling curves were recorded by a computerized data acquisition system. After experiments, the solidified samples were mounted in epoxy resin, sectioned, polished according to standard metallographic procedure. The samples were characterised by X-ray diffraction (XRD) and scanning electron microscopy (SEM) by back scattering (BSE). Vickers microhardness of the samples were measured on polished sample cross-sections.

3. Results and Discussion

Under equilibrium conditions, the $Al_{96}Fe_2Nd_2$ alloy transformation L $\rightarrow$ L + τ_3 ($Al_{10}Fe_2Nd$) begin at 1073 K, next transformation L + τ_3 ($Al_{10}Fe_2Nd$) $\rightarrow$ L + fccAl + τ_3 ($Al_{10}Fe_2Nd$) at 923 K and last eutectic transformation L + τ_3 ($Al_{10}Fe_2Nd$) + fccAl $\rightarrow$ τ_3 ($Al_{10}Fe_2Nd$) + fccAl + eutectic (Al + $\alpha Al_{11}Nd_3$) takes place at a temperature of 903 K. The balance phases belonging to the field of the $Al_{96}Fe_2Nd_2$ alloy composition are the ternary compound τ_3 ($Al_{10}Fe_2Nd$), fcc-Al phase and eutectic (Al + $\alpha Al_{11}Nd_3$) (Hu, 1988). The sample undercooled of 30 K presented microstructure under equilibrium conditions. However, the sample undercooled of 109 K was found the $Al_{13}Fe_4$ phase, as shown the Fig. 1. The X-ray diffraction patterns of the as-casting $Al_{96}Fe_2Nd_2$ alloy presented equilibrium conditions phases, the compound τ_3 ($Al_{10}Fe_2Nd$), fcc-Al phase and $\alpha Al_{11}Nd_3$ as show the Fig. 2. The X-ray diffraction patterns (Fig. 3) undercooled sample of 109 K confirmed the presence of the $Al_{13}Fe_4$ phase. This phase not belongs to the field of the $Al_{96}Fe_2Nd_2$ alloy composition then this phase is in equilibrium metastable with the others phases of the field alloy in study, due rapid solidification induced by increasing of the undercooling. Normally, thermodynamic driving force for crystallization is different at different undercoolings and the crystal growth kinetics under large undercooling condition is far different from that of equilibrium state, and consequently, the microstructure should show different phases (Wang and Wei, 2001). Rapid solidification induced by increasing of the undercooling provoked an extension of the field where the $Al_{13}Fe_4$ phase is in equilibrium metastable with the others phases of the field alloy in study (Massalski, 1989).

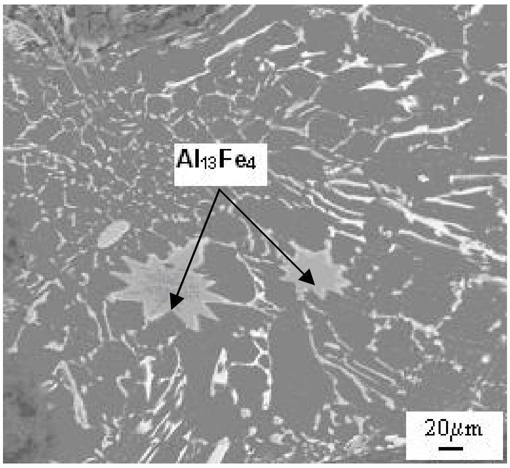


Figure 1- Microstructures presenting the $Al_{13}Fe_4$ phase with undercooling from 109 K.

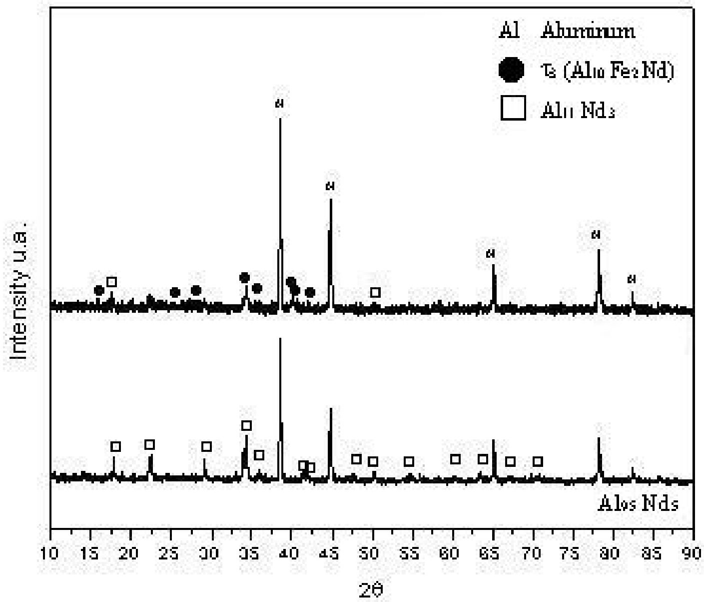


Figure 2- X-ray diffraction patterns of the as-casting $Al_{96}Fe_2Nd_2$ alloy.

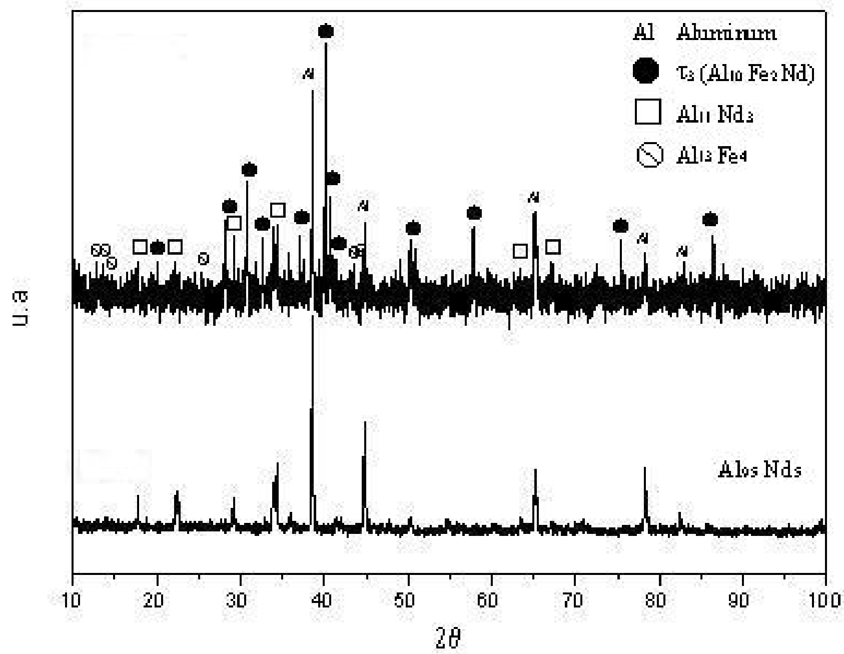
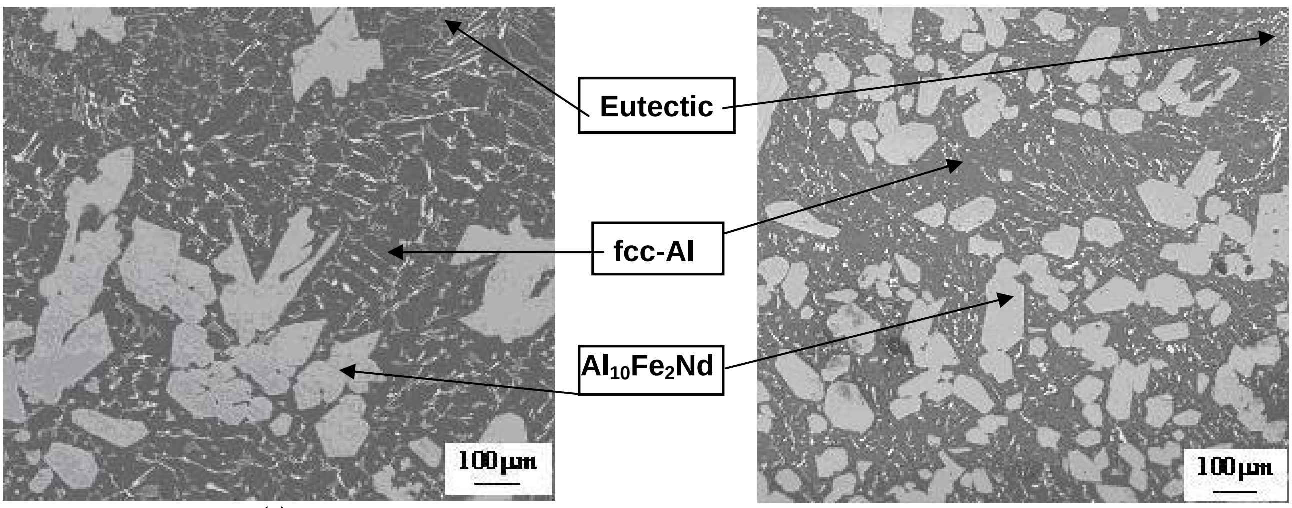


Figure 3- X-ray diffraction patterns of the sample undercooled of 109 K.

A maximum undercooling of 109 K was obtained for $Al_{96}Fe_2Nd_2$ alloy and its solidification microstructures at different undercoolings are illustrated in Fig. 3. Observed that when undercooling is small, the sizes of the compound τ_3 ($Al_{10}Fe_2Nd$) and eutectic region are large, as shown in Fig. 3(a). With increasing of the undercooling, both the compound τ_3 ($Al_{10}Fe_2Nd$) size and the eutectic area decrease sharply, as Fig. 3(b) presents. This means microstructure refinement occurs with the enhancement of undercooling (Kattamis, 1970). It is well known that the driving force for crystallization of undercooled melt is Gibbs energy difference between solid and liquid, which is a function of undercooling. Therefore, the microstructures of solidified alloys often change with undercooling. When undercooling is small, solidification proceeds in a manner close to equilibrium condition, leading to slow speed of both nucleation and crystal growth. When undercooling becomes large, the crystal growth velocity usually increases, provoked changes morphology in primary phase and eutectic (Wang and Wei, 2001).

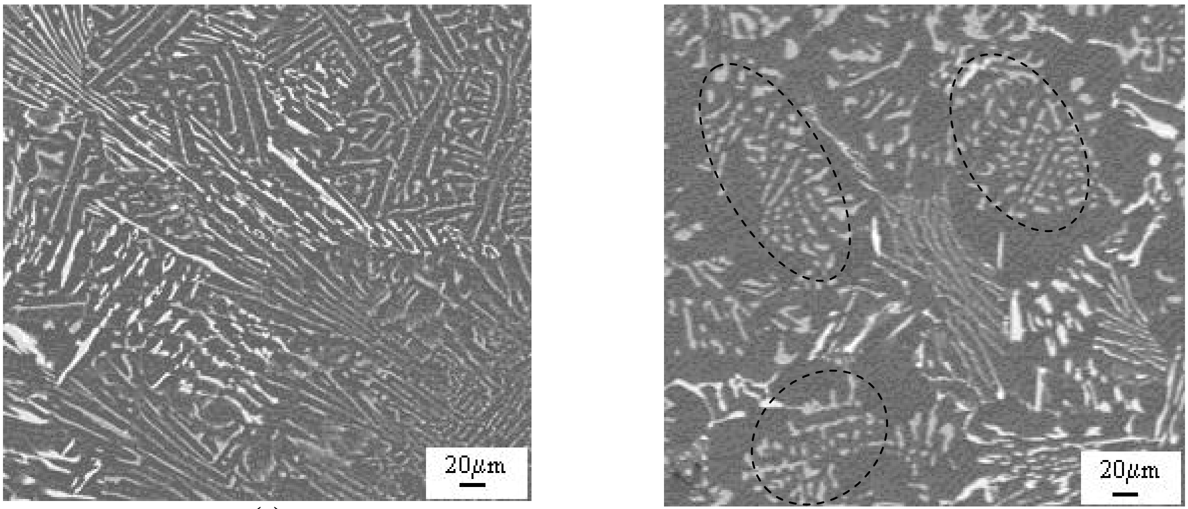


(a) (b)

Figure 3- Microstructures at different undercoolings. (a) ΔT = 30 K; (b) ΔT = 109 K.

Some works (Wei and Herlach, 1994; Wei and Herlach, 1997; Walker, 1961; Li *et al.*, 2002) have reported refinement in undercooled samples, explaining that the most probable mechanism for this phenomenon is structure fragmentation, if the undercooling passes a certain critical undercooling ΔT^*. It means that, rapid growing primary phases and eutectic become morphologically unstable and decay with a reduction of the interface area, as well as the driving force for such a process.

The Fig. 4 shows that was observed a substantial change in the eutectic morphology with the increasing undercooling degree. The morphology indicated a transition from eutectic regular lamellar (Fig. 4a) to some regions of eutectic anomalous, (Fig. 4b).



(a) (b)

Figure 4- Morphology indicating a transition from eutectic regular lamellar (a) to some of eutectic anomalous (b) (regions surrounded). (a) ΔT = 30 K; (b) ΔT = 109 K.

Many studies (Wei and Herlach, 1994; Goetzinger, Barth and Herlach, 1998; Wang *et al.*, 1997; Abbaschian and M. D. Lipschutz, 1997) have observed transition from eutectic regular lamellar to eutectic anomalous in other eutectics systems. They have established a variety of propositions to explain the formation mechanisms of anomalous eutectic structures. Thus it is reasonable to conclude that anomalous eutectic is the product of rapid solidification whereas lamellar eutectic forms under slow solidification conditions. The most probable physical mechanism for this phenomenon is that a considerably high amount of interfacial energy is stored in the fine lamellar eutectic structure in comparison to the gain of volume energy. The reduction of the interfacial energy acts as a driving force for the on going fragmentation mechanism leading to a transformation from the lamellar eutectic microstructure to an equiaxed microstructure of the anomalous eutectic (Wang *et al.*, 1997).

The mechanical behaviour of the $Al_{96}Fe_4Nd_4$ alloy was checked through Vickers microhardness measurements (HV) on samples undercooled. The increasing undercooling level from 30 K to 109 K promotes increasing on the microhardness from 72 HV to 135 HV respectively. The refinement of the $Al_{10}Fe_2Nd$ primary phase, change eutectic morphology and presence of the $Al_{13}Fe_4$ phase can to have provoked increasing on the hardness value of the alloy. This result is interesting for probable technological applications of undercooled alloys.

4. Conclusions

Flux technique was used to undercooling of the $Al_{96}Fe_4Nd_4$ alloy. The alloy has been undercooled by up to 109 K. It was revealed by microstructural observations a presence of the $Al_{13}Fe_4$ metastable phase and that grain refinement of the primary phase and eutectic has taken place in alloy when the undercooling increasing from 30 K to 109 K. Moreover, a substantial change in the eutectic morphology with the increasing undercooling degree was observed too. The morphology indicated a transition from eutectic regular lamellar to some regions of eutectic anomalous. It was suggested that rapid growing primary phases and eutectic become morphologically unstable and decay with a reduction of the interface area, as well as the driving force for such a process.

The refinement of the $Al_{10}Fe_2Nd$ primary phase and eutectic, change eutectic morphology and presence of the metastable $Al_{13}Fe_4$ phase can to have provoked increasing on the hardness value of the alloy from 72 HV to 135 HV.

5. Acknowledgements

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