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Kinetics and dynamics of planar abnormal grain growth in nanocrystalline nickel

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ABSTRACT

Growth of abnormally large grains has been observed previously in nanocrystalline Nickel both indirectly by electron imaging after heat treatment and by *in-situ* resistivity measurement. However the form and rate of the growth has been derived indirectly. This work makes use of *in-situ* heat treatment combined with back scattered electron imaging illustrates that the abnormal micro-planar faceted grains grow with a dependence not only on the surrounding but also as the orientation of the growing facet. This observation has shown that the grains grow in a semi-constrained manner such that high index faces such as {211} grow faster than stable {111} faces.

1. Introduction

Nanocrystalline (nc) materials, including metals and alloys are recognised to be important because of their observed and potential superior mechanical, physical and chemical properties compared with the equivalent at the micro-scale material [1–3]. As described by Pineau et al. [4] there have been several reviews focusing upon the underlying mechanisms that allow extreme strengths to be obtained together with factors that control the associated fracture [5–10]. In general an important contribution to the improved properties is related to the large volume fraction of grain boundary compared with a similar material where the grains are of a micro-scale dimension [11,12]. However if the mechanical properties are to be retained at higher temperatures the stability of these nc materials against grain growth and recrystallisation is a key issue [13–15]. Nc materials show thermal instability however the mechanisms and kinetics of grain coarsening and growth differ compared with the corresponding material at the micro-scale. Grain growth at temperature in micro-scale polycrystalline metals and alloys has been addressed extensively [16–21]. In the simplest case it can be considered to be driven by the reduction in grain boundary area, A, which if all grain boundaries are assigned the same energy, γ, and this is independent of time at temperature, the total energy, E, of the polycrystalline material is reduced so that

$$\delta E = \gamma \sum_{i=1}^{N} \delta A_i \tag{1}$$

Thus for microscale grain size material the reduction in energy is achieved by migration of individual grain boundaries so that larger grains grow at the expense of smaller grains by the reduction in the size and removal [21]. In smaller grain size materials, nc materials, shorter length scale sub-micrometre grain growth has an effect because energy reduction is more complex in such cases total energy includes the variation from boundary to boundary [21]. This grain boundary energy γ_b can vary significantly dependent on the specific boundary misorientation. Therefore the total energy reduction is achieved by either a decrease in boundary area, energy or a combination of these two factors so that

$$\delta E = \sum_{t=1}^{N} \gamma_b \delta A_i + \sum_{t=1}^{N} A_i \delta \gamma_b \tag{2}$$

Banmak et al. [22] showed for grains within thin polycrystalline films of material grain growth is also driven by a reduction in total grain boundary energy. However complexities arise from the two dimensional form so that triple points (junctions) promote curvature driven growth of the grain. In the case of nc polycrystalline metals and alloys the relative grain volumes, grain boundary areas, grain edges and grain nodes have the potential to modify the grain

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growth kinetics. Experimentally a relative stagnation in growth has been observed in both thin films and nc polycrystalline nickel when a certain grain size is achieved during coarsening with time at temperature suggesting different kinetics are obeyed for smaller grain sizes [22,23].

Accelerated thermally induced grain growth in nc materials is due to additional growth modes; rotation and coalescence of adjacent grains as well as normal grain boundary movement [5,12]. Certainly grain coarsening in nc materials can be inhibited by the presence of impurity atoms. In such cases interaction of impurity atoms with grain boundaries is complex so that some boundaries can be pinned by some types of impurity atoms whereas other elements increase mobility leading to grain growth [5,12,24]. As a consequence this approach to grain boundary pinning has been adopted to control grain size in nc materials. Recently for the specific case of a ncNi containing 280 ppm S with an initial grain size of ~50 nm, Darnbrough and Flewitt [23] have shown that at temperatures above 300 °C grains coarsen to a mean size of ~550 nm with an activation energy of 72.9±13.4 $kJmol^{-1}$. At this stage normal growth stops and the process stagnates. The stagnation was attributed to grain boundary movement being pinned by the presence a critical concentration of the impurity S atoms. This stable microstructure is retained at temperatures up to 485 °C. Above this temperature the grains remain at a constant size of ~500 nm but there is the additional process of the formation of distributed large abnormal micro-planar boundary grains. The planar boundaries in this fcc material form with non-close packed crystal facets. To quantify the changes associated with the early stages of ncNi growth and the emergence of the abnormal grains *in-situ* measurements were carried out by electrical resistivity. This technique allowed the evaluation of the evolution of grain boundary volume fraction [25].

In this paper we explore the coarsening of grains in two nc polycrystalline nickels, one containing 280 ppm and the other less than 120 ppm impurity S using a hot stage within a scanning electron microscope. This is to allow in-situ observation of the growth of individual grains from the initial nanograin size through stagnation to the formation and growth of abnormal grains. The kinetics are compared with previous observations and inparticular the rate of growth of specific planar crystallographic facets associated with the abnormal grains are measured.

2. Materials and experimental methods

2.1. Nanocrystalline nickel

For this investigation two sources of commercially available polycrystalline (nc) nickel were selected. The first in the form of a 50 mm × 50 mm × 0.5 mm thick sheet produced by electrodeposition obtained from Intgran Technologies Ltd (Canada). The second was purchased from Goodfellows Ltd (UK) in the form of a layer of electrodeposited nc nickel of 0.1 mm thickness on a copper substrate. The former contained 280 ppm impurity sulphur as measured by combustion infrared analysis. The sulphur content of the latter was calculated via comparison of the ncNi plus Cu and the Cu substrate. By considering the relative volume of the two samples a value of between 30 and 120 ppm sulphur was evaluated for the nc nickel. The grain size of these two materials was measured using dark field transmission electron microscopy (JEOL 2000) and found to be 50.6± 7.3 nm and 46.4±2.6 nm respectively [23,25–28]. Both materials were found to be fully dense and subsurface interrogation through focused ion beam (FIB) milling and scanning electron microscopy (SEM) using the FEI Helios NanoLab 600i workstation showed no voids on a microscopic scale. In order to prepare the specimens for investigation in the hot stage SEM a series of mechanical polishes were undertaken with decreasing grit sizes and finished with a colloidal silica vibropolish for 2 h.

2.2. Hot stage scanning electron imaging

This work considers the temporal development of grain structure with heat treatment at 500 °C within a Zeiss SEM. The heat treatments comprised of a 600s ramping phase followed by a dwell at 500 °C for 12 h (43,200s) while grain microstructure images were recorded every 80s. This was achieved using back scattered scanning electron imaging (BS-SEM) with the specimen held at temperature using a hot stage (Gatan/Deben Microtest 5000 W). The specimen was spot-welded to a plate of stainless steel which was held by the heating grips. The two ends of the stainless steel plate were heated directly so that the plate, as well as the specimen, were heated by thermal conduction. The temperature at the centre of the plate (the location of the specimen) was measured by an additional thermocouple spot-welded to the stainless steel plate and logged as the heat treatment of the specimen. The need to spot-weld the specimen and thermocouple restricted the choice of support plate material to stainless steel. This is because nickel (or copper) have high thermal conductivity which does not allow the local heating required to spot weld the sample and thermocouple. During temperature ramping, the jaws were gradually opened to compensate the thermal expansion of the stainless steel plate ensuring no load was applied. The temperature control was achieved manually to select a responsive ramping rate and negate/reduce overshoot.

The observations made *in-situ* were of the 2D grain structure on a surface produced via polishing and as such are a slice through the 3D microstructure. Hence the observations are not directly illustrative of the growth within the material. Post heat treatment electron backscatter diffraction (EBSD) orientation mapping (Zeiss Sigma FEG-SEM with EDAX camera) gave the crystallography of the facets of grains at the surface. Hence the index of the growing facet can be found, from the subsurface angle between it and the grown face, observed through FIB sectioning and imaging [23]. This gives insight into the link between growing and static faces with crystallographic reference. Given the observed growth on the surface and a known angle to the growing facet it can be labeled with a crystal direction and the growth velocity normal to the crystallographic facet can for compared with models and observation of growth of crystals in free systems.

2.3. Data analysis

Investigation using predominately imaging techniques requires careful consideration when extracting quantitative information. In this work a number of algorithms where developed within Matlab 1 to identify grain structure. First and foremost image noise was addressed using a moving modal average kernel. Grain structure was isolated through backscatter image contrast, regions of neighbouring pixels with similar contrast are labeled as a grain. Growth of abnormal grains relative to the static surround microstructure can be observed through subtraction of the first normalised image from those following and looking at the regions that have the highest absolute contrast as they are most different from the original microstructure allowing abnormal grain growth to be followed.

3. Results

Prior to undertaking the hot stage experiments in the SEM heat treatments were conducted over a range of temperatures and times. The two materials under investigation show distinct evolution of microstructure under heat treatment leading to; in the low