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X-ray diffraction (XRD) profile analysis of pure ECAPannealing Nickel samples

Ahadi Damar Prasetya', Muhammad Rifai', Mujamilah' and Hiroyuki Miyamoto'

¹Center for Science and Technology of Advanced Material, National Nuclear Energy Agency, Serpong, Tangerang Selatan, Banten, Indonesia ²Department Science and Engineering, Doshisha University, Kyotanabe Campus, Kyoto, Japan

E-mail: iamrifai@yahoo.com

Abstract. X-Ray Diffraction (XRD) profile of pure equal channel angular pressing (ECAP)annealing nickel samples has been thoroughly investigated for studying the material structures changes that imply to the mechanical behavior. Nickel-based material can be used for several applications such as biomaterial, gear, and some part of the instrument at nuclear facilities, which require high-grade standard material properties. ECAP is one kind of severe plastic deformation (SPD) techniques to obtain excellent mechanical properties without adding another element. However, the ECAP process generates metastable structures due to some mismatch structure and inhomogeneous stress within the material. This problem can usually be resolved by annealing after the ECAP process. In this article, pure Nickel was processed by ECAP at 423 K for two passes. The post-ECAP annealed will be carried out at the temperature range from 298 K until 1373 K. The microhardness test results indicate that the ECAP process increases the microhardness significantly, which remains stable after annealing until 773 K. At higher annealing temperature, the mechanical properties will drop suddenly and reach the microhardness value of pure pre-ECAP Nickel. This behavior could be explained clearly by the XRD data analysis result, which shows similar behavior structure changes. XRD data initially show peak shifting to lower 2θ value, which indicates an expansion to a higher lattice parameter, then at the higher annealing temperature, the diffraction peaks split gradually. This peak splitting could be indexed as pure pre-ECAP Ni peaks, which could be related to the drop of the microhardness value.

1. Introduction

Nickel (Ni) has been used widely in many areas, especially for biomedical applications as an implant and in the nuclear facility. Usually, Ni is combined with Ti to form nitinol that can be used in the medical industry. Nitinol has the 'superelastic' properties that can be useful for avoiding invasive procedures that demanded non-conventional materials [1]. In nuclear applications, the nickel-based alloy has been used as the structural material of the molten-salt nuclear reactor (MSRE) [2].

Severe plastic deformation (SPD) commonly has been used for producing nanostructured and submicrocrystalline materials in bulk form. This method is promising because it produces nanostructured metals and alloys with high physical and mechanical properties such as fatigue properties, water resistance, corrosion resistance, superelasticity (at lower temperatures and larger strain rates than conventional superelastic conditions). This method can lead to a new generation of construction and

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functional materials. There are many types of SPD techniques, and the most widely used is equalchannel angular pressing (ECAP) [3, 4].

In the ECAP method, the metal material is pressed through channels at certain angles; as a result, there will be a uniform shift (shear) throughout the entire material [4]. Many studies have shown that the ECAP process increases the hardness of the metal due to changes in the microstructure of the metal. However, the ECAP process or more general SPD techniques generate metastable structures and high internal stresses as a result; usually, the material has low ductility, low impact toughness and limited thermal stability [5]. This problem usually can be resolved by annealing the material after ECAP.

Annealing is a heat treatment process in which the alloy is heated up to the prescribed annealing temperature for a prescribed time, followed by controlled cooling to softening the alloy. By annealing the material, strains can be relieved, and the strength can be reduced. However, annealing also accompanied by an undesirable grain growth that can reduce the strength gained from SPD [5].

X-Ray Diffraction (XRD) is a powerful technique for analyzing materials structure. Various aspects of materials microstructure can be analyzed using XRD, e. g.: texture, residual stress, crystallite size, character and density of defects in particular dislocations, and lattice micro-strain [6], [7]. Although the measurement of grain size by XRD can be corroborated by TEM, the determination of the lattice micro-strain often relies only on XRD. The Scherrer formula can be used to evaluate the crystallite size directly if the lattice micro-strain is not present. Besides, the Stokes–Wilson equation can be used to determine the lattice micro-strain if the crystallite size is adequately large. However, if the contribution of the broadening peak comes from both fine crystallites and lattice micro-strains, Williamson–Hall method is often applied. This method is a simplified approach that differentiates between size-induced and strain-induced peak broadenings by considering the peak width as a function of 20 [7]. The purpose of this study is to investigate the cause of mechanical behavior change of pure Ni metal caused by structure changes using XRD.

2. Research Method

2.1. Materials

The materials used in this study are pure Ni metal, a high-temperature lubricant made from fluorine and buff paper with alumina suspension (PRESI) 9 μ m, 3 μ m, and 1 μ m, OP-S suspension (Streuers).

The tools used in this study include an ECAP machine, infrared furnace (UVLAC MILA5000), and X-Ray Diffractometer (SmartLab, Rigaku), Vickers hardness testing machine.

2.2. Method

Initial coarse grain specimens were formed with dimensions of $8 \times 8 \times 120$ mm. The sample then processed by ECAP, two passes at 423 K via Bc route using a split die with two channels intersecting at an angle of 90°, the specimen was then lubricated with high-temperature fluorine-based lubricants. The samples then annealed from 298 K to 1373 K in a vacuum for one hour. The initial sample (as-received) was characterized using Vickers microhardness tester. While ECAPed and post-annealed ECAP samples were characterized by X-ray Diffractometer (XRD), and Vickers microhardness.

The microhardness tests were performed on a Vickers hardness testing machine under a load for 15 s dwell time after for each annealing. Hardness testing was carried out for ten times per each sample.

X-ray diffraction (XRD) test of ECAPed and post-annealed samples were carried out by SmartLab, Rigaku. XRD sample surfaces were buffed by an automatic polisher. The SmartLab X-ray diffractometer used CuK α , 40 kV, and 0.2 A from 30° until 130° with continuous scanning type.

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3. Result and Discussion

3.1. Result

The initial sample has a microhardness value of 75 Hv, and the ECAP process increases it to 275.7 Hv. The value of microhardness compared to the. Annealing temperature remains stable until 773 K. The microhardness value started to drop from 773 K until 873 K and then remains stable again until the end of measurement at 1273 K. The microhardness falls slightly at the end of annealing (1373 K) with the value of 73.75 Hv. Overall, the microhardness curve from Figure 1. exhibited softening with three-stage, such as recovery, recrystallization, and grain growth [4].



Figure 1. Microhardness profile of pure ECAPed Ni after annealing

The XRD is shown in Figure 2. The XRD data show five peaks appear, around 43° , 50° , 75° , 92° , and 97° , indicating peaks from Ni with FCC structure. However, all the peaks shifted to lower 2θ , and the peaks broaden (for lower annealing temperature) if compared to the standard Ni, and peaks are splitting started from annealing temperature 973 K until 1373 K. This XRD profile could be present because of the presence of uniform strain (tension) that shifted the peaks and non-uniform strain (compression) that makes the peaks broaden [7]. This phenomenon can be an indication of structural change that comes from the ECAP and annealing process of the sample. The structure change can be the source of the mechanical behavior (microhardness) change of the samples.

Williamson-Hall (W-H) analysis with a uniform deformation model (UDM) was used to calculate the micro-strain. UDM assumed that the lattice strain present in the material is uniform in all crystallographic directions [8]. The Williamson-Hall equation used in this calculation is presented in Eq. (2), the equation is derived from Eq. (1). Eq. (1) assumes that the peak broadening occurred simultaneously due to both size (BD) and strain effects (B ϵ).

$$\beta = \beta_D + \beta_\varepsilon = \frac{K\lambda}{D\cos\theta} + 4\varepsilon\tan\theta \tag{1}$$

$$\beta \cos \theta = \frac{K\lambda}{D} + 4 \varepsilon \sin \theta$$
⁽²⁾

Where D is the crystallite size (in nm), ε is the lattice strain, K is the crystallite-shape factor (taken as 0.9), λ is the wavelength of the incident X-rays (0.154 nm for Cu K α radiation), β is the full-width at half-maximum (FWHM) of the diffraction peak (in radians), and θ is the Bragg angle (in degrees). From the plot of $\beta \cos \theta$ compared to the 4sin θ , a straight line that facilitates the extraction of lattice strain (ε) from its slope and crystallite size (D) can be obtained. The lattice strain is the slope, and the crystallite size can be calculated from the intercept.

The results of the W-H analysis are presented in Table I, while the plots are presented in Figure 3. The results showed that annealing decrease the micro-strain in the samples, annealing more than 973 K drops the micro-strain significantly. However, the results after annealing more than 973 K showed that the micro-strain value is negative, and the slopes of the W-H plots become negative, indicating that strain broadening must be small and the strain comes from compressive strain [8-10]. Before annealing more than 973 K, the micro-strain value is positive, and the slopes of the W-H plots are positive, indicating the presence of tensile strain [8,9].



Figure 2. (a) XRD profile from 30° until 130°; (b) XRD profile from 42° until 45.5°, the peak start to split after annealing more than 973 K; (c) XRD profile from 50° until 52°, the peak start to split after annealing more than 973 K

Samples	Process	A (nm)	Microstrain (%)
Post-ECAP	Recovery	0.356144795	0.002104
Annealing 473 K		0.355859891	0.002237
Annealing 573 K		0.355760542	0.002108
Annealing 698 K		0.356060520	0.001967
Annealing 773 K		0.355983793	0.001943
Annealing 798 K	Recrystallization	0.356079922	0.001887
Annealing 823 K		0.356127128	0.001834
Annealing 873 K		0.356245592	0.001835
Annealing 973K		0.356225033	-0.000544
Annealing 1073 K	Grain growth	0.356016772	-0.000117
Annealing 1273 K		0.356321338	-0.000029
Annealing 1373 K		0.356140702	-0.000262

Table 1. Lattice parameter and microstrain value in each annealing temperature



Figure 3. Williamson-Hall plot of each sample

3.2. Discussion

The comparison between the microhardness and micro-strain is shown in Figure 4. It shows that the trends of both almost similar, indicating that microhardness depends linearly with the micro-strain. Both affected by the annealing temperature, annealing at the recrystallization temperature will decrease the micro-strain in the material, which will affect the microhardness.

Figure 5 explains the comparison between the lattice parameter and the micro-strain. Although the lattice parameter data are not good enough, it can be shown that there is a trend of lattice parameter decrease until annealing at 573 K and then started to increase again in the higher temperature. First, annealing will decrease the tensile strain on the material also decrease the lattice parameter, so the

material becomes denser. This explains in the recovery process, and the microhardness remains stable with a slight increase.

Furthermore, on the higher temperature (recrystallization dan grain growth) the lattice parameter started to increase in line with the tensile strain release, then only compression strain is present due to the negative value of the micro-strains. It indicates that the rearrangement of atoms occurs, forming a more stable structure. Annealing at a higher temperature will trigger thermal expansion, so the distance between atoms become more stretchable. So, not only tensile strain release and defects annihilation that occurs, but the forming of a new structure also occurs. The splitting peaks from the XRD data after 1073 K also indicate the forming of a new structure. The new structure can be Ni crystal on the standard condition, where this is explained by the split formed at a higher 2θ , approaching the 2θ of Ni in standard condition. At last, this new structure has a lower microhardness value than the post-ECAP.



Figure 4. Comparison between microhardness and microstrain in the same range of temperature



Figure 5. Comparison between micro-strain and lattice parameter in the same range of temperature

4. Conclusion

Annealing process of pure ECAP nickel could affect the microhardness due to structural change and stress release; it can be seen that the micro-strain value trends in line with the microhardness value at the same annealing temperature. This structure change phenomenon could be described clearly using XRD data, in the recovery stage (473 until 773 K), the lattice strain decreases in line with the lattice parameter, while in the recrystallization and grain growth stage (798 until 1373 K), the lattice strain started to increase, the tensile strain release occurs, and at the end, only compression strain is left. Higher annealing temperature will direct the structural change towards the pure Nickel in standard condition, indicated by XRD peaks splitting and peaks shifting toward the standard pure nickel XRD peaks position.

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