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The analysis of effect of heat treatment temperature on micro structure, crystal structure and hardness material on alloy $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$

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Abstract. The effect of heat treatment temperature (in 500 ^{0}C, 600 ^{0}C, and 700 ^{0}C) from $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$ as fuel cladding material candidate's reactor nuclear power plant; for microstructure, crystal structure, and hardness has been carried out. Several characteristic was conducted by using an optical microscope, x-ray diffractometer and Vickers method. The result showed the crystalline characteristic peaks by a tendency to a single crystal formation and microstructure is getting better with less precipitation and the hardness of the alloy is 329.6 4.5 HVN after the homogenization process.

1. Introduction

The increasing of the global energy demand causes the need of more sustainable energy sources including the nuclear energy. In order to developed the nuclear technology, it required a new design and the construction of nuclear fuel cladding. As a current material, commercial zirconium alloy was expensive and unable to self produced (limited to patent right). Thus, the new material must have a characteristic such as strength, corrosion resistance, fabrication capabilities, neutron adsorption cross section, and cheap [1].

In recent years, the research of zirconium alloys with Nb, Sn, Fe, Mo and Ge has been developed. In order to make the new material cheaper, the cladding material must use zirconium element that weight at least 95% [3.7] and must made domestically. One of the properties of the material that needed is the resistance in high temperatures and corrosion. In accidental condition with a lot of loss of coolant (LOCA), the integrity of the nuclear plant need to be maintained. The good material should be forming a protective layer from continues oxidation process due to high temperature oxidation that occurred on a microscopic scale. The candidate of this nuclear fuel cladding was $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$ alloy materials.

2. Theory

The heat treatment is a process that can control the material's properties, especially metal; it also can alter the material's structure. The effects of this process can make the material harder, malleable, increase toughness and to refine the metal grain size.

There are several processes of heat treatment which are annealing, sintering, calcination, tempering, normalization and homogenization. Annealing was a heating process that can reduce dislocation density and growth grain by recrystallization mechanisms. Sintering was a heating process of pellet's powder at its recrystallization temperature for material's strengthening and porosity elimination. Calcination is a process which powder's is heated at 1000 ^{0}C to remove the carbonates and water [4]. Tempering is a special heating process for softening the steel [5]. Homogenization is process heat treatment at high temperature for reducing unwanted chemical elements by diffusion and to get ordered crystal structure and grain. Normalization is a heat treatment at recrystallization temperature and cooled down at room temperature. Factors that can affect the process of heat treatment [4,5]: heating temperature, restraining temperature and cooling rate.

2.1 Atomic Diffusion

The atoms will be on a stable position at 0 K (-273 C). In this temperature, they are at the lowest energy. If the temperature is increased, the atom’s energy would be increased too and affect the atom’s movement. This process called diffusion. Diffusion mechanism was classified based on displacement and position atoms which is:

a) Empty mechanism
   There is always a blank space in crystal structure configuration. This happens by hopping of the atom to the vacant position. When one atom jumped to fill the blank space and the other will replaced it, there will be a continuity process.
b) Interstitials mechanism
   If there was a different size between 2 atoms, it can be happen interstitial mechanism. For example: the diffusion of atom nitrogen to metal surface.
c) Interchange mechanism
   Another name was ring mechanism, an atom movement such as cycle. It has a large distortion, so need a big energy.

2.2 Zirconium Alloys

Zirconium is a very hard metal grey which have melting point temperature 1860^{o}C, and a low neutron absorption 0.18 barn. Zirconium can be easily corroded [6,7]. Therefore, if we wanted some good mechanical properties we must made alloy of Zirconium (Zr-Nb-Fe-Sn) as a nuclear fuel cladding. The development of power reactors are still on going by the goal energy efficiency and reducing of waste and cost. Power efficiency can easily achieved at 400-600^{0}C [9].

3. Research Methodology

3.1 Materials and Equipment

<u>Material</u> : alloy material was made by smelting techniques in arc melting furnace, the composition were 96.2% Zr, 2.3% Sn,1.1% Nb, and 0.4% Fe. Metallographic material and complete etching materials.

<u>Equipment</u>: Arc furnace melting, grinding, polishing, cutting machine, optical microscope, Vickers hardness tester method, and an X-ray diffractometer.

3.2 Procedures

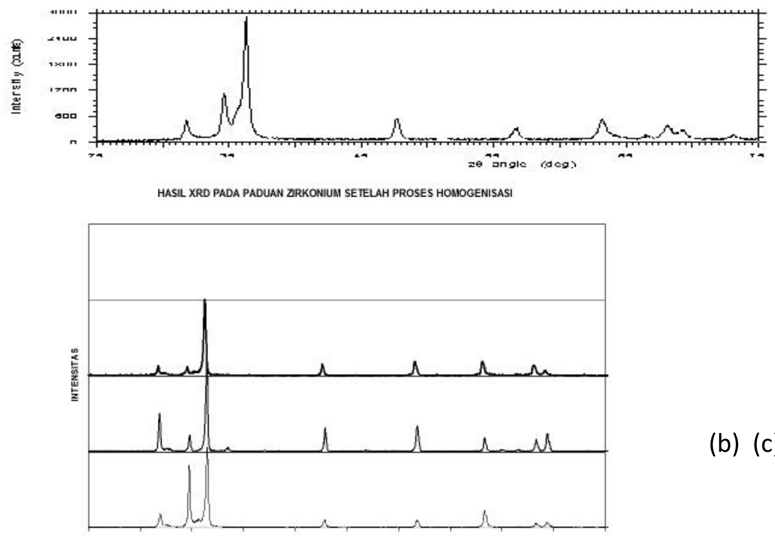
- Weigh Zr, Sn, Nb, and Fe and placed them in the crucible to be made $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$ alloy material. Furnaced them by arc melting furnace in argon gas condition to avoid them oxidation.
- Melted and cooled them then cut into 2x2 by diamond blade cutter type JMQ-12 at low speed to ensure a neat piece surface and clean of impurities.
- Normalized them by heated at 1000 ° C.
- Homogenized them by heat treatment at variation temperatures: 500 ° C, 600 ° C, 700 ° C and held for 2 hours.

- Polished them with sandpaper in roughness consecutive sequence : 120, 400, 800, 1200, to 2000.
- Topped them at velvet pabric samples on diamond paste 0.25 lm.
- Etched their surface with 3% Nital solution in 15 seconds until the surface shiny like mirror. Nital solution was made from a mixture between 3 ml HNO_3 with 97 ml methanol technical.
- Dried them with blow dryer at room temperature until completely dry.
- Tested them for microstructure by optical microscope, for crystal structure and grain size by X-ray diffractometer and for hardness by Vickers test method.

The x-ray diffractometer with a Cu-K wavelength ( ) at approximately 1,5405Å belongs to central laboratory UIN Ciputat, Tangerang. optic microscopy tool and hardnes tester Vickers method at engineering Laboratory FT, UKI, Jakarta.

4. Results and discussion

4.1a. Analysis crystal structure and grain size of the alloy material Zr $_{96,2}$ Sn $_{2,3}$Nb$_{1.1}$ Fe$_{0,4}$
Ingot alloy materials visually formed a solid alloy with a little oxidation at the surface and homogeneous melted. Smelting alloy synthesis decreased Gibbs free energy of each element for made a particular phase at alloy. In this condition, both elements Nb and Fe has melted at 1800^0C and has diffused to zirconium matrix formed phase / two new compounds Zr $(Nb,Fe)_2$ and $(Zr, Nb)_3Fe$ [9].



(a)

(b) (c) (d)

Figure 4.1 diffractograms of alloy materials Zr $_{96,2}$ Sn $_{2,3}$Nb$_{1.1}$ Fe$_{0,4}$ a) Without heat treatment and after heat treatment at temperature (b). 700°C, (c) 600°C, (d). 500°C.

Figure 4.1 showed a diffractogram pattern of alloy materials Zr $_{96,2}$ Sn $_{2,3}$Nb$_{1.1}$ Fe$_{0,4}$; the measurement results used x-ray diffractometer with a Cu-K wavelength ( ) around 1,5405Å. Adequate X-ray beam penetration power can displayed a best profile peak to background ratio. Diffraction peak intensity very clearly difference and sharply visible in Figure 4.1 (a) - 1(d), like needles. At the scattered start peak looked widen because of the influence of fluorescence material. The whole pattern of diffraction peaks dominated by the phase zirconium cladding as the primary matrix material. Three main first peak in dominant phase were (10 0), (0002) and (10 1) looked at the angle 2 = 32.080^0 , 34.840^0 and 36.650^0. The highest intensity for 2010 counts / second (cps) was saw at field (10 1) belongs to zirconium phase. The diffraction pattern showed two predominant phase which formed before heat treatment were treated as follows: phase zirconium (Zr) in hexagonal structure, new phases or compounds Zr $(Nb, Fe)_2$ and $(Zr, Nb)_3Fe$ in tetragonal structure. Some peak intensity shrinkaged, even there was only one dominant peak at 700 ° C. it was caused by atom diffusion of alloy material $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$.

Table 4.1 Results matter of the grain size of the alloy material $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$

Temperature (°C)	2 (deg)	d(Å)	FWHM (deg)	The Grain Size (Å)
Original (yet H.T)	36,251	2,4759	0,448	2.989
500	36,465	2,4620	0,200	7.299
600	36,455	2,4728	0,180	8.117
700	36,285	2,4627	0,160	9.354

Table 4.1 showed the heating process made the grain size becomes larger, the higher temperature the larger size grain. It can be happen because of atom diffusion at boundaries grain towards grain when it heated. There was a striking changeable depend on heat treatment temperatures: 500 ° C, 600 ° C and 700 ° C, the change of crystal structure direct to the single crystal formation in the plane *hkl* (101). There were some shrinkage peak intensity when the temperatures changed from 500 ° C to 600 ° C, and at 700 ° C there was a dominant peak with the angle θ 36.285^0. It indicated the material direct to homogeneous single crystal structure [6]. It become zirconium alloy would be better able as fuel cladding at high temperature. At high temperature the atoms will be retained their crystal structure, so there was no creep or fractures [5].

4.2 Analysis of the microstructure of alloy material $Zr_{96,2}Sn_{2,3}Nb_{1,1}Fe_{0,4}$

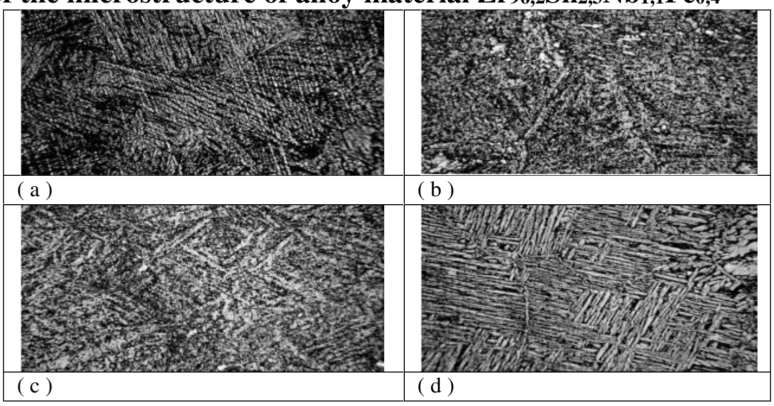


Figure 4.2 Photogram $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$ alloy material surface with magnification of 500 X: (a) the original sample (not heat treatment), after the heat treatment (b) 500 ° C, (c) 600 ° C, and (d) 700 ° C.

Figure 4.2 appeared the microstructure of alloy material $Zr_{96,2}$ $Sn_{2,3}Nb_{1.1}$ $Fe_{0,4}$ before and after heat treatment at different shape of micro structure. Atom diffusion easily occurred at the material surface because it has lower stability than atom in crystal. This is due to the coordination of surface atoms was equal to the coordination of crystal atoms. Therefore the surface atoms have a higher free energy and a less robust bond [5]. There were black spots on grain and boundaries grain. It indicated the presence of material precipitation. Previously [9.14] precipitation of alloy material can be happen if there was intermetallic compounds in material such as Zr (Nb, Fe)$_2$ and (Zr, Nb)$_3$Fe. The precipitation may be made in the process to make alloy Zikonium or when heat treatment was given either at normalization or homogenization. When precipitation processed some element constituent material will secede and made intermetallic phases. Generally precipitation has a high energy and inhibit deformation rate of the pressure received from the material [11-13].

4.3 Analysis hardness of alloy material $Zr_{96,2}Sn_{2,3}Nb_{1,1}Fe_{0,4}$

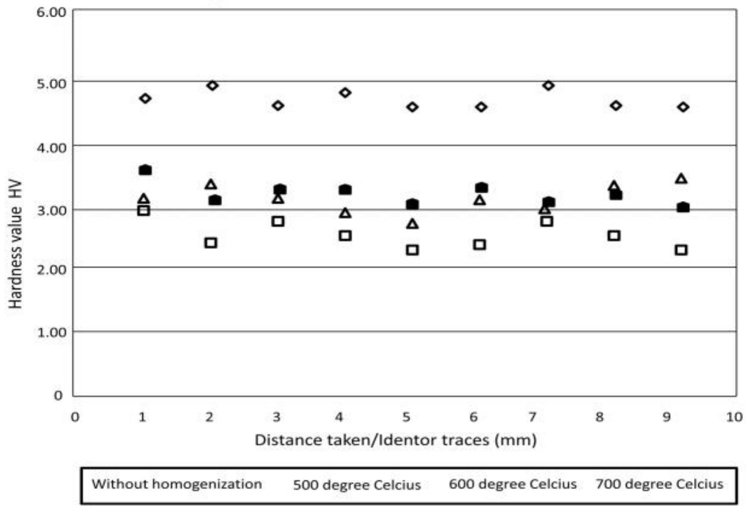


Figure 4.3 Hardness Test Results on alloy material $Zr_{96,2}Sn_{2,3}Nb_{1,1}Fe_{0,4}$

Figure 4.3 showed the higher temperature the more decreased hardness of the alloy. It was happen because of atom diffusion, there was migration from boundary to limit and make space; the consequences of violence will diminish [5]. The violence reduction as well as solid dislocation usually inhibit the creep propagation and pressure deformation was reduced. It become material hardness was reduced too. Figure 4.3 showed after homogenization, the material hardness increased, even in the surface the hardness scattered flat. If the hardness have flatten the material would be better, because the thermal expansion distribute quietly. At high temperatures dislocation of grain boundary made creep spread. Because the atoms get heat energy made the distance between atoms larger and propagated the creep [5]. Heat treatment multiplied scattering wheat field. This field made the crystals to their dominant orientation (preferred orientation); was field (10 1). Facts showed that the larger of grain size was 2989 (before heat treatment) for 9354 (after the heat treatment) caused a shift to the field (10 1). The hardness 329.6 HV after heat treatment the alloy material $Zr_{96,2}Sn_{2,3}Nb_{1,1}Fe_{0,4}$ smaller than before homogenization process 477,11HV. However it was feasible to be used as fuel cladding material, due to the hardness value 76.27 HV as fuel cladding material hardness minimum [8:13].

Conclusion

- Material alloys after heat treatment Zr96,2Sn2,3Nb1,1Fe0,4 made two dominant phases: zirconium phase hexagonal structure and intermetallic phases were Zr (Nb, Fe)$_2$ and (Zr,Nb)$_3$Fe the tetragonal structure. Also occured width grain from 2,989 (before heat treatment) to 9,354 , after heat treatment in the field (10 1).
- A violence decreased in $Zr_{96,2}Sn_{2,3}Nb_{1,1}Fe_{0,4}$ alloy material after heat treatment of 477.11 HV before heat treatment decreased to 261.44 HV at a temperature of 500^0C.

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