THE EFFECT OF TEMPERATURE AND AGING TIME ON THE MICRO STRUCTURE AND HARDNESS OF Ni-Al-Ti-Ge ALLOY

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Abstract
Intermetallic alloy is a combination that consists of two metal compounds or more. One of intermetallic alloy that can be used is nickel. Nickel is a corrosion resistant metal and withstands high temperature. Intermetallic alloy can be used for application on aircraft components such as turbin gas engine that operate in high temperature and extreme environment. This study is conducted to find out the changes of micro structure and hardness in NiAlTi material with addition of 1.5% germanium as a doping. In this study, heat treatment process was carried out with variation of temperature and aging time. The heat treatment included solution treatment and aging process. Solution treatment process was carried out at 1000°C using tube furnace with holding temperature time for 2 hours. Aging process was carried out at 700°C, 800°C, 870°C for 1, 2 and 3 hours for each temperature. Based on the hardness result, solution treatment heat treatment increase the grain size and also hardness of NiAlTi+1.5%Ge due to intermetallic compound formed in the alloy. Aging treatment increase the grain size and also hardness value. Maximum hardness take place at 870°C for 3 hours with hardness of 45 HRc. Characterization by optical microscope and SEM showed that the alloy contain dendritic and interdendritic microstructure. Examination by XRD and EDS shows that intermetallic compound formed in all NiAlTi+1.5%Ge specimens are dominated by intermetallic of Ni₃Al (Ɣ’).

Keywords : intermetallic alloy, Aging, Ni-Al-Ti,Ge
INTRODUCTION

Human needs on material application which resistant to high temperature is the most demanded. High temperature resistant material is material that can maintain its nature or does not experience any decline properties at high temperature. It can be a combination of certain alloy elements in order to get the needed characteristics at high operation temperature (Basuki, 2016). Nickel-based super alloys are called “creep-brITTLE” materials because of their superior tensile strength and low creep ductility (Gupta et al., 2012). Nickel-based super alloys have been widely used in different applications, where require high strengths at high temperatures (Azadi et al., 2018).

Intermetallic alloy is chemical combination with certain composition of atom. Intrinsically, intermetallic compound is strong and has high modulus of elasticity. Phase of intermetallic has some primacy such as high strength at high temperature, corrosion resistant and high temperature oxidation resistant. It also has high melting point and low density. One of intermetallic alloy is Ni-Al. Nickel is a corrosion and high temperature resistant metal (Mitra, 2017). Phase formed in the combination of Ni-Al is γ(Ni,Al) (Sims and Hagel, 1972). The application on phase Ni,Al is for Turbine blades and Jet engine turbine vanes (Sanusi et al., 2014; Jozwik, Polkowski and Bojar, 2015).

Intermetallic alloy Ni-Ti has many applications in science and industry. NiTi is the most widely used in engineering and medical applications. Compounds of titanium nickel (Ni-Ti) has many interesting characteristics namely shape memory effect, high melting temperature, super elastics (Hu, Xue and Shi, 2017; Li et al., 2017). Germanium is an important semiconductor material. Zone-refining techniques produce crystal germanium for semiconductor with extremely high purity (Zhang and Ni, 2016). Nickel aluminum bronze (NAB) is copper aluminum alloy with addition of nickel and iron elements (Qin et al., 2004). The addition of alloying elements (Al and Ti) resulting in the emergence of phase γ[Ni (Al, Ti)] coherent so as to provide reinforcement effect (Li et al., 2017).

The purpose of this study is to find out changes of micro structure and mechanical characteristics on Ni-Al-Ti material doped by 1,5% of Ge element. Solution treatment process was carried out on 1000°C temperature for 2 hours. This process aims to homogenize tested material in order to produce more stable phase. After that, quenching process with water coolant media was conducted. Then, aging process was carried out. Aging process in this study was conducted at temperature of 700°C, 800°C, and 870°C with each temperature for 1, 2 and 3 hours.

METODOLOGY

Material

The used raw material are nickel ingots in the amount of 99.99%, aluminium ingots 99.99%, titanium 99.99%, germanium 99.99% being weighed with total weight of 20 gram each sample. The added composition of germanium compound element was 1,5%. The weight was intentionally made the same with melting by 20 gram. Table 1. Showed the composition of alloy that is used in this study (% weight).

<table>
<thead>
<tr>
<th>Alloy</th>
<th>% Weight</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>81,425</td>
</tr>
<tr>
<td>Al</td>
<td>6,55</td>
</tr>
<tr>
<td>Ti</td>
<td>10,525</td>
</tr>
<tr>
<td>Ge</td>
<td>1,5</td>
</tr>
</tbody>
</table>

Smelting was done using single arc melting furnace under argon gas atmosphere with ultrahigh purity argon gas. Single Arc Melting Furnace is a home made furnace which function to melt a metal or alloy. This process resulted compound of Ni-Al-Ti+1,5%Ge in the physical form as button ingot as seen in figure 1.

![Figure 1. Specimen resulted from melting.](image)

Methods

The specimen resulted from melting stage was processed in the solution treatment and aging. The solution treatment process was done on 10 specimens at 1000°C using tube furnace with holding temperature time for 2 hours under high purity argon atmosphere. After that,
quenching process was carried out using water coolant media. Then, aging process was carried out. The aging process in this study was conducted in the temperature of 700°C, 800°C and 870°C for 1, 2, 3 hours respectively. The aging process is used in the tube furnace under high purity argon atmosphere.

The curve of solution treatment and ageing process of Ni-Al-Ti + 1.5%Ge alloy is shown in figure 2.

![Figure 2. Curve of solution treatment and aging process Ni-Al-Ti + 1.5%Ge.](image)

**Characterizations**

After the ingots sample were aging, the ingots sample were observed by an optical microscopy for microstructural characterization. X-ray diffraction is used for phase present examination and SEM-EDS for characterization specimens. Hardness Rockwell C was used to evaluate the hardness of the alloys with force 0.5 kg and dwell time 10 minutes.

**RESULTS AND DISCUSSION**

**Hardness Test (HRc)**

Hardness test was carried out on specimen NiAlTi+1.5%Ge (as-cast), NiAlTi+1.5%Ge (as-quench), and NiAlTi+1.5%Ge (aging). Hardness test used rockwell scale-C method. Hardness test was conducted five times as showed in table 2.

Based on hardness test data specimen NiAlTi+1.5%Ge (as-cast) and NiAlTi+1.5%Ge (as-quench) has the same hardness value. In accordance with the equation Hall-Patch (G. E., 1962). The process of solution treatment could not influence the hardness of NiAlTi+1.5%Ge significantly. Sample in solution treatment produce the increase of hardness value compared with as-cast due to intermetallic compound in alloy.

![Table 2. Hardness of specimen NiAlTi+1.5%Ge (as-cast) and NiAlTi+1.5%Ge (as-quench).](image)

**Figure 3. Curve of specimen hardness value Comparison NiAlTi+1.5%Ge (as-cast) and NiAlTi+1.5%Ge (as-quench).**

![Table 3. Hardness of NiAlTi+1.5%Ge specimen after aging.](image)
Hardness test data was converted into curve of NiAlTi+1.5% Ge as-aging specimen as seen in figure 4.

<table>
<thead>
<tr>
<th>Effect Temperatur and Aging time on the hardness Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hardness HRC</td>
</tr>
<tr>
<td>1 jam</td>
</tr>
<tr>
<td>2 jam</td>
</tr>
<tr>
<td>3 jam</td>
</tr>
</tbody>
</table>

Figure 4. Hardness NiAlTi+1.5%Ge as aging

Based on hardness test data, it used rockwell scale-C specimen NiAlTi+1.5%Ge (aging) in temperature of 700°C, 800°C, and 870°C for 1 hour, 2 hours and 3 hours each temperature which produce an increased hardness value but not significant (Setiawan et al., 2019).

Micro Structure Examination

Metallographic test is a branch of metal science to see micro and macro structure of material in order to recognize metal structure, phases and characteristics of metal along with its alloy. It is carried out by using optical microscope OLYMPUS BX60M. The result of metallographic can be seen in figure 5 (a-k).

(a) NiAlTi+1.5%Ge (as-cast)

(b) NiAlTi+1.5%Ge (as-solution treatment)

(c) NiAlTi+1.5%Ge (aging T=700°C, t=1 hour)

(d) NiAlTi+1.5%Ge (aging T=700°C, t=2 hours)

(e) NiAlTi+1.5%Ge (aging T=700°C, t=3 hours)

(f) NiAlTi+1.5%Ge (aging T=800°C, t=1 hour)

(g) NiAlTi+1.5%Ge (aging T=800°C, t=2 hour)
NiAl Ti+1,5% Ge (aging T=800°C, t=3 hours)

NiAl Ti+1,5% Ge (aging T=870°C, t=1 hour)

NiAl Ti+1,5% Ge (aging T=870°C, t=2 hours)

NiAl Ti+1,5% Ge (aging T=870°C, t=3 hours)

Figure 5. Structure of micro specimen 200x magnification

In metallographic examination of specimen NiAlTi+1,5%Ge as-cast, as-quench and aging, the results showed that the microstructure of all alloys consisted of dendritic and interdendritic. The dendritic microstructure contain of NiAl (Ɣ) phase and interdendritic of Ni3Al (Ɣ’). The Ni3Al (Ɣ’) is called as intermetallic compound alloy of NiAlTi+1,5%Ge. The strengthening of nickel-based superalloys is mainly obtained by the coherent precipitation of a large amount of Ni3Al type γ’ % phase in a nickel-based γ matrix (El-Bagoury, 2011). The microstructure of Ni3Al alloy usually consists of Ni solid solution matrix and typically cuboidal Ni3Al precipitates(Ganesh Kumar and Joseph Sahaya Anand, 2011). Ni elements will dissolve and form a precipitate compound Ni3Al, Ni3Ti and Ni3Ge. The compounds that formed it will improve the mechanical properties of the alloy, due to the precipitation strengthening mechanism (Jozwik, Polkowski and Bojar, 2015; Setiawan et al., 2019).

Grain Size

The grain size of microstructure is calculated by Jeffries method. The table 4 is the result of grain size calculation using Jeffries method and coverted in to bar chart graphic as seen in figure 6. The table 4 showed that as quenched alloy have got relatively similar grain size compare with as cast alloy.

Table 4. The grain size of specimen NiAlTi+1,5%Ge as-cast and as-quench.

<table>
<thead>
<tr>
<th>Alloys</th>
<th>Grain size in (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiAlTi+1,5%Ge (as-cast)</td>
<td>44.3</td>
</tr>
<tr>
<td>NiAlTi+1,5%Ge (as-quench)</td>
<td>49.03</td>
</tr>
</tbody>
</table>

Figure 6. Curve of grain size NiAlTi+1,5%Ge as-cast and as-quench

Data in the table 5 is the result of grain size of NiAlTi+1,5%Ge alloy after aging. Then it converted into bar chart as seen in figure 7.
Table 5. The grain size of NiAlTi+1.5%Ge alloys after aging

<table>
<thead>
<tr>
<th>Grain size (µm)</th>
<th>800°C</th>
<th>850°C</th>
<th>900°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Jam</td>
<td>51.6</td>
<td>56.6</td>
<td>59.7</td>
</tr>
<tr>
<td>2 Jam</td>
<td>61.0</td>
<td>66.6</td>
<td>69.7</td>
</tr>
<tr>
<td>3 Jam</td>
<td>70.0</td>
<td>75.1</td>
<td>78.2</td>
</tr>
</tbody>
</table>

The result showed that the rise of the temperature and time aging will increase the grain size of microstructure of the NiAlTi+1.5%Ge alloy. According to Hall and Pecht relation it can be concluded that the hardness of the NiAlTi+1.5%Ge alloy should be decreased with increases of grain size but it hardness increased after quenching and aging as seen in figure 3, and 4. The increase of hardness of the NiAlTi+1.5%Ge alloy after quenching aging is due to existence of intermetallic compound as seen in the X-ray diffraction in figure 9 and 10.

X-Ray Diffraction Test

X-Ray Diffraction (XRD) test is conducted to find out the phase present in the NiAlTi+1.5%Ge alloy and an intermetallic compound. The X-ray Diffraction examination was carried out on specimen NiAlTi+1.5%Ge as-Cast as showed in figure 8 and NiAlTi+1.5%Ge as-Quench in figure 9. Figure 10 showed the X-ray diffraction examination on NiAlTi+1.5%Ge alloy after aging at 800°C for 3 hours. Figure 8 showed that peak of XRD in the alloy dominated by nickel aluminate (Ni₃Al) and intermetallic compound NiGe and NiTi. In figure 9 showed that the phase present in the NiAlTi+1.5%Ge alloy dominated Ni₃Al as primary peak in the XRD data. Secondary peak contain intermetallic compound of NiTi and NiGe (Yuniarto etc, 2019).

Figure 7. Curve of grain size value comparison specimen NiAlTi+1.5%Ge (aging).

Figure 8. XRD as-cast Ni-Al-Ti+1.5%Ge.

Figure 9. XRD as-quench Ni-Al-Ti+1.5%Ge.

Figure 10. XRD as aging at T=800°C, t=3 hours Ni-Al-Ti+1.5%Ge.

The X-Ray Diffraction on specimen NiAlTi+1.5%Ge after aging process in the temperature of 800°C for 3 hours have been conducted. There is phase Ni₃Al(Y') on the highest peak in the figure 10 which is known as...
primary phase or matrix that has the most composition, whereas phase of Ni₄Ti₃, Ni₃Ti, NiTi, Ni₃Ge have relatively low peak which are secondary phase.

**SEM-EDS Characterization**

Scanning Electron Microscope (SEM) and Energi Dispersive Spectroscopy (EDS) were carried out to find out morphology and composition of the compounds formed in specimen Ni-Al-Ti+1.5%Ge after aging process already done in the temperature of 800°C for 3 hours. The following result of SEM-EDS specimen Ni-Al-Ti+1.5%Ge after aging in the temperature of 800°C for 3 hours depicted in Figure 11,12 and 13. Figure 11 depicted that the microstructure of NiAlTi+1.5%Ge after aging in the temperature of 800°C for 3 hours consist of dendritic and interdendritic microstructure with the matrix of Ni₃Al. The interdendritic contain of Ni₃Al and intermetallic.

**Figure 11.** SEM NiAlTi+1.5%Ge aging T=800°C, t=3 hours 3000x.

**Figure 12.** Testing area of EDS Window area 1 dan area 2 material NiAlTi+1.5%Ge aging T=800°C, t=3 hours.

**Figure 13.** EDS Window area 1 NiAlTi+1.5%Ge aging T=800°C, t=3 hours.

Table 6. Composition value of EDS result graphic Window area 1 material NiAlTi+1.5%Ge T=800°C, t=3 hours.

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight %</th>
<th>Atomic %</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiL</td>
<td>79.20</td>
<td>70.99</td>
</tr>
<tr>
<td>GeL</td>
<td>1.33</td>
<td>0.97</td>
</tr>
<tr>
<td>AlK</td>
<td>7.80</td>
<td>15.22</td>
</tr>
<tr>
<td>TiK</td>
<td>11.66</td>
<td>12.82</td>
</tr>
</tbody>
</table>

**Figure 14.** Result graphic of EDS Window area 2 material NiAlTi+1.5%Ge aging T=800°C, t=3 hours.
Table 7. Composition value of EDS graphic result Window area 1 material NiAlTi+1.5%Ge aging T=800°C, t=3 hours.

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight %</th>
<th>Atomic %</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiL</td>
<td>79.82</td>
<td>71.50</td>
</tr>
<tr>
<td>GeL</td>
<td>0.92</td>
<td>0.67</td>
</tr>
<tr>
<td>AlK</td>
<td>7.85</td>
<td>15.31</td>
</tr>
<tr>
<td>TiK</td>
<td>11.41</td>
<td>12.52</td>
</tr>
</tbody>
</table>

Table 7. Composition value of EDS graphic result Window area 1 material NiAlTi+1.5%Ge aging T=800°C, t=3 hours.

Figure 15 showed the plotting chemical composition of Ni and Ti based on EDS area 1 and 2 on the Ni-Al-Ti Ternary phase diagram .[ASM] Based on EDS analysis chemical composition of Ni and Ti on the area 1 and 2 and plotted in the ternary diagram Ni-Al-Ti the crossection on the two lines located at Ni3Al (ϒ’). The Ni3Al (ϒ’) is the main phase is confirmed bymicrostructural and X-ray diffraction examination (Pradoto etc.), and atom (%) composition.

CONCLUSION

Based on hardness result, microstructure, XRD, SEM and EDS examination, it can be concluded that solution treatment increase the grain size and also hardness of NiAlTi+1.5%Ge due to intermetallic compound formed in the alloy. The aging treatment increase the grain size and also hardness value. Maximum hardness take place at 870°C for 3 hours with hardness of 45 HRc. Characterization by optical microscope and SEM showed that the alloy contain dendritic and interdendritic microstructure. Examination by XRD and EDS shows that intermetallic compound formed in all NiAlTi+1.5%Ge specimens are dominated by Ni3Al (ϒ’).

ACKNOWLEDGEMENT

Author would like to say thank you to PSTNT-BATAN that was very helpful in the study process and to all materials and equipment and UNJANI who always give the author support and direction in accomplishing this study. Main contributor :Djoko Hadi Prajitno.

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