International Journal of Chemical, Materials and Biomolecular Sciences
ISSN: 2415-6620
Vol:8, No:5, 2014

Effect of Al Addition on Microstructure and Physical Properties of Fe-36Ni Invar Alloy

Seok Hong Min, Tae Kwon Ha

Abstract**—**High strength Fe-36Ni-base Invar alloys containing Al contents up to 0.3 weight percent were cast into ingots and thermodynamic equilibrium during solidification has been investigated in this study. From the thermodynamic simulation using Thermo-Calc®, it has been revealed that equilibrium phases which can be formed are two kinds of MC-type precipitates, MoC, and M_2C carbides. The mu phase was also expected to form by addition of aluminum. Microstructure observation revealed the coarse precipitates in the as-cast ingots, which was non-equilibrium phase and could be resolved by the successive heat treatment. With increasing Al contents up to 0.3 wt.%, tensile strength of Invar alloy increased as 1400MPa after cold rolling and thermal expansion coefficient increased significantly. Cold rolling appeared to dramatically decrease thermal expansion coefficient.

Keywords**—**Invar alloy, Aluminum, Phase equilibrium, thermal expansion coefficient, microstructure, tensile properties.

I. Introduction

INVAR effect was discovered by Guillaume in 1897 [1], which has since been found in various ordered and random alloys and even in amorphous materials [2]. Other physical properties of Invar systems, such as atomic volume, elastic modulus, heat capacity, magnetization and Curie (or Neel) temperature, also show anomalous behavior. It has long been realized that the effect is related to magnetism [3], but a full understanding is still lacking. Since its development, the mechanical behavior of Invar alloy has been virtually ignored. The mechanical behavior of Invar is particularly important for appropriate applications. Stresses resulting from operation, the weight of the component itself and the loading of mechanical fasteners can all result in an unwanted change in shape for precision structures. Thus, the attributes of macroyield strength, microyield strength, and microcreep strength are key concern to designers and fabricators of precision instrumentation. The scarcity of mechanical properties data has prompted an investigation to determine metallurgical strengthening mechanisms for Invar. Conceptually, the strength of Invar can be viewed in classic metallurgical terms. Potential strengthening mechanisms include interstitial solid solution strengthening, substitutional solid solution strengthening, cold working, alloying to allow for precipitation strengthening, or compositing. Of these methods, cold working cannot be applied to Invar because it results in dimensional instability as internal stresses associated with cold work relax with time. Substitutional solid solutions are not desired because all alloy additives adversely affect low expansivity that is achieved with the Fe-36Ni composition. Commercially available low expansion alloys having very high strength are achieved by precipitation hardening – although their thermal expansion suffers since a strengthening precipitate will invariably have a high thermal expansivity.

Invar alloys are used in instruments, such as hair spring in watches and thermostatic bimetals, and also used for the construction of liquid natural gas (LNG) tanks. The most recent application of Invar is for the manufacture of high strength power-transmission wire. Due to the increase in power demand, there has been an increasing need for a core wire with a high strength and a low coefficient of thermal expansion. It is well known that Fe-36Ni based Invar alloy has a small coefficient of thermal expansion [4]. However, this alloy has a low tensile strength even after being heavily deformed. High strength Invar alloys have been strongly recommended and are partially used for this purpose [5].

The high-strength and low-thermal-expansion Invar alloy has a composition of Ni, or Ni and Co around 36%, interstitial type solid solution strengthening elements such as C and N, several kinds of substitution type solid solution elements such as Cr and Mo, and several kinds of precipitation hardening elements such as Ti, Nb, and V in such a range as not to deteriorate the low thermal expansion property, and the balance of Fe [6].In the present study, precipitation of a commercial high-strength Fe-36Ni based Invar alloy have been investigated by thermodynamic simulation using Thermo-Calc® and experimental works, such as microstructure observation, hot rolling, tensile test, cold rolling, and thermal expansion coefficient measurement.

II. Experimental Procedures

The ingots of Fe-36Ni base alloy with various Al contents up to 5wt.% were cast by vacuum induction melting. The chemical compositions of the ingots were summarized in Table I. The weight of the ingot was 50kg and the dimensions were 140mm x 140mm x 400mm.

TABLE I
Chemical Compositions of the Ingots Produced in This Study. (wt.%)

Ingot	Ni	Mo	Cr	C	Mn	Si	Nb	V	Al	Fe
A0	36	2.8	0.8	0.3	0.2	0.1	0.1	0.1	-	Bal.
A1	36	2.8	0.8	0.3	0.2	0.1	0.1	0.1	1.0	Bal.
A3	36	2.8	0.8	0.3	0.2	0.1	0.1	0.1	3.0	Bal.
A5	36	2.8	0.8	0.3	0.2	0.1	0.	0.1	5.0	Bal.

S. H. Min is with the Department of Advanced Metal and Materials Engineering, Gangneung-Wonju National University, 120 Gangneung-daehangno, Gangneung, Gangwon 210-702, South Korea.
T. K. Hais with the Department of Advanced Metal and Materials Engineering, Gangneung-Wonju National University, 120 Gangneung-daehangno, Gangneung, Gangwon 210-702, South Korea (phone: 82-10-2367-1989; e-mail: tkha@gwnu.ac.kr).

Microstructures of the Invar alloy were observed by optical microscopy using the etchant called Glyceregia consisting of 3 parts HCl, 1 part HNO_3, and 3 parts Glycerin. Morphology, composition and distribution of precipitates were investigated using scanning electron microscopy under the BS (back-scattered) image mode. To predict the equilibrium phases, their chemical compositions and equilibrium weight fraction, and the forming temperatures, Thermo-Calc®, commercial thermodynamic simulation software was used in this study.

Hot rolling was conducted on the ingots at 1200°C to obtain plates with thickness of 14mm, from which specimens for tensile test and thermal expansion coefficient measurement were machined. The dimensions of tensile test specimen were 27mm in gauge length and 6.4mm in diameter and those for thermal analysis were 20mm in length and 5mm in diameter, respectively. Thermal expansion coefficients were measured at temperatures ranging from room temperature to 900°C with heating rate of 2°C/min, using NETZSCH DIL402C. Cold rolling was also conducted on the hot-rolled Invar alloys at room temperature, after machining plates with thickness of 6 mm, to obtain sheets with various thicknesses from 2 to 3mm. On these sheet specimens, tensile tests were carried out at room temperature under the strain rate of 5×10^{-4} s^{-1}.

III. Results and Discussion

A typical microstructure of as-cast Invar alloy (A0) is given in Fig. 1, revealing coarse precipitates along grain boundaries. Back-scattered SEM image of a coarse precipitate is also given in the figure together with chemical composition analyses. It is interesting to note that the coarse precipitate is not carbide but a Mo-rich solid solution. Instead, very fine particles uniformly distributed in the matrix are carbides of Mo and Nb as shown in TEM observation given Fig. 2.

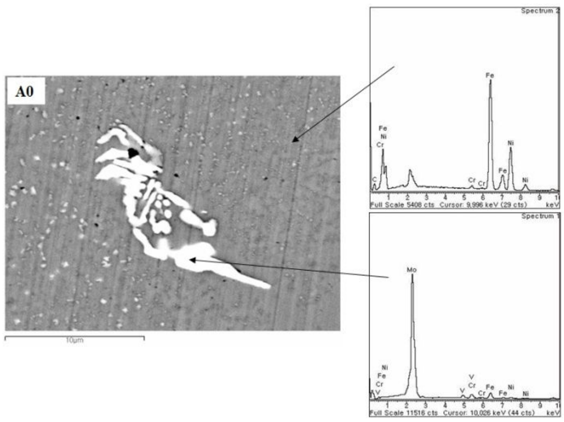

Fig. 1 Typical microstructure of as-cast Invar alloy ingot A0 showing back-scattered electron image of a coarse particles and its chemical compositions

Calculated phase distribution diagram of Fe-36Ni base Invar alloy is given in Fig. 3, in which the equilibrium phases, their equilibrium weight fraction, and the forming temperatures are given. There are two types of carbides in this alloy, i.e. $M_{23}C_6$ and Mo_2C, which are expected to contribute to the precipitation hardening in the lights of weight fraction and temperature.

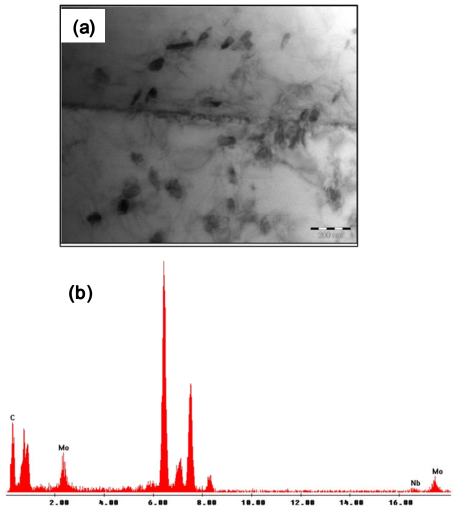

Fig. 2 TEM micrograph (a) of fine precipitates of ingot A0 and its chemical compositions (b)

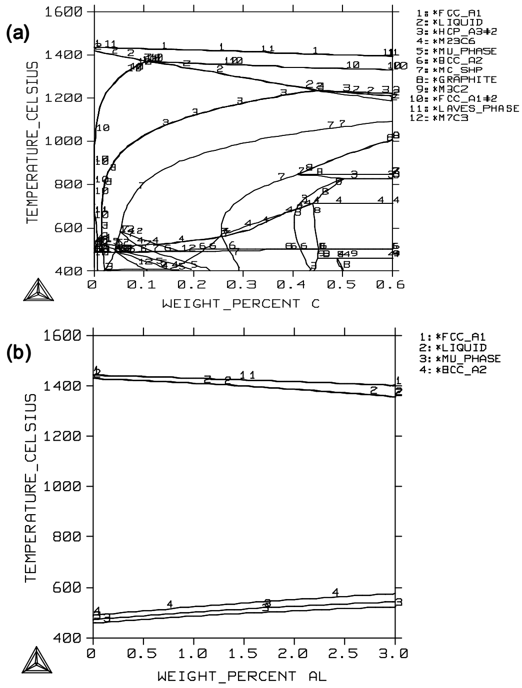

Fig. 3 Phase distribution calculated on Fe-36Ni base Invar alloy with variation of C content (a) and Al contents (b), respectively

The Mo_2C phase appears from about 1200°C with its maximum weight fraction of 3% near 900°C. Interestingly, the equilibrium weight fraction of Mo_2Cphase is reduced from 900°C with formation of $M_{23}C_6$ phase. It is apparent from the figure that the coarse phases formed during casting process as illustrated in Figs. 1 and 2 are thought to be Mo-rich solid solution and can be dissolved at temperature above 1200°C. It is noted that MoC carbide exists at temperatures from 900°C to 700°C but its equilibrium weight fraction is very low. Addition of aluminum appears to stabilize BCC phase but the effect is very trivial.

Fig. 4 shows microstructures of the ingots after hot rolling and it is obvious that hot rolling at 1200°C reduced almost large precipitates observed in Fig. 1. With contents of aluminum increased, the amount of remained precipitates appeared to increase.

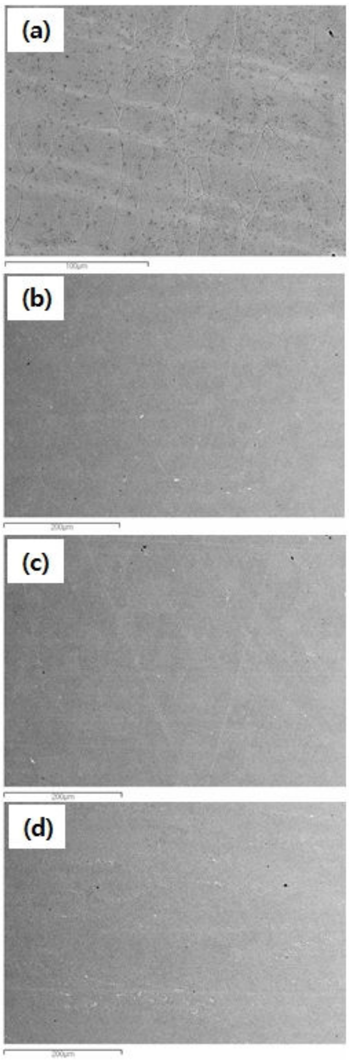

Fig. 4 Microstructures observed after hot rolling at 1200°C conducted on the ingots of A0 (a), A1 (b), A3 (c), and A5 (d), respectively

It is well known that the precipitation of Ni base intermetallic compounds such as Ni_3Al (γ') depletes the matrix in Ni, which raise coefficient of thermal expansion (CTE). The composition of the residual matrix depends on the Ni content of the alloy [7], on the solubility limits of Al at the aging temperature, and on the exact composition of the γ', which can also vary with the temperature and time of aging. Results of CTE measurement on the ingots are summarized in Fig. 5 in the temperature range from room temperature to 800°C. Addition of aluminum seriously deteriorates CTE, especially at the ambient temperature range where almost applications of Invar alloy are conducted. Interestingly, addition of Al by 1wt.% shows very similar values of CTE to those of the ingot without Al. In this regard, the Invar alloy with A1 composition is expected to be used high strength applications such as overhead power cable cores if several strengthening mechanisms were combined to obtain the high strength of 1200MPa.

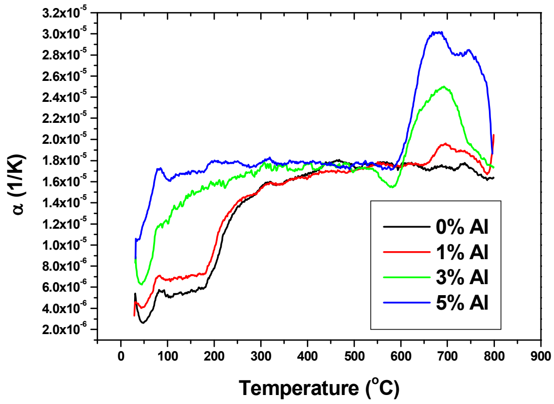

Fig. 5 Thermal expansion coefficient measured on the Invar alloys with various aluminum contents

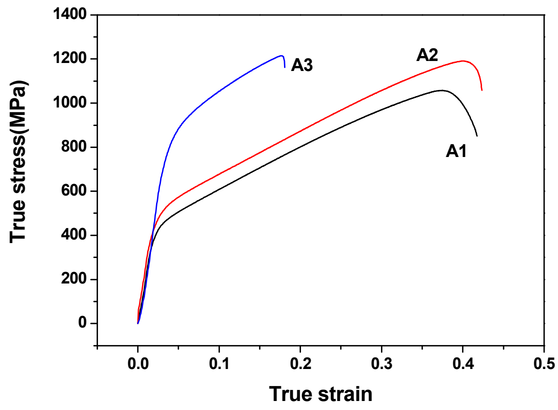

Fig. 6 True stress-true strain curves of Invar alloys with various aluminum contents obtained after hot rolling process

The results of tensile test conducted at room temperature on the specimens with various Al contents after hot rolling are given in Fig. 6. Yield and tensile strength significantly increased with increasing Al content. High tensile strength above 1000 MPa was obtained in all cases and high strength of 1200MPa was obtained in the specimen with Al content of 5wt.%. It is, however, obvious that the alloy with high Al content of 5wt.% shows increased yield and tensile strength but significantly low uniform elongation, which will be a barrier to practical applications. During hotrolling process, coarse precipitates were presumably resolved as expected from phase equilibrium simulation and elongation was improved in the cases of ingots A1 and A3.

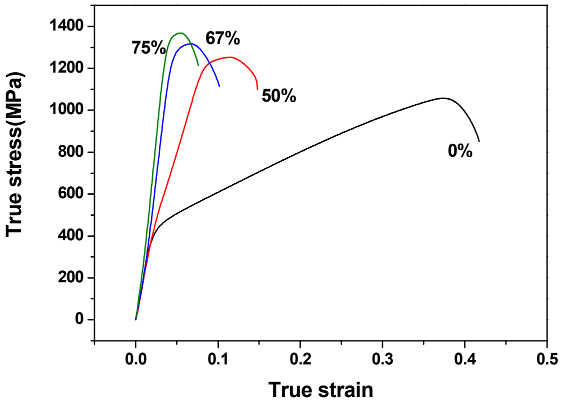

Fig. 7 True stress-true strain curves of the ingot A1 with various cold reduction ratios obtained after cold rolling process

Fig. 7 shows true stress-true strain curves of A1 ingot after cold rolling with various cold reduction ratios. By cold deformation, yield and tensile strength of A1 alloy increases dramatically. Very high strength of 1400 MPa was obtained by cold rolling. The result of CTE measurement on the cold rolled Invar alloy is illustrated together with hot rolled one in Fig. 8. It is very interesting to note that thermal expansion coefficient of cold worked specimen is much lower than that of hot rolled one, decreased with temperature and reached near zero value. The reduction in CTE by cold work is well established phenomenon [8], [9] and is used in addition to adjustment of the residual matrix to attain the target values.

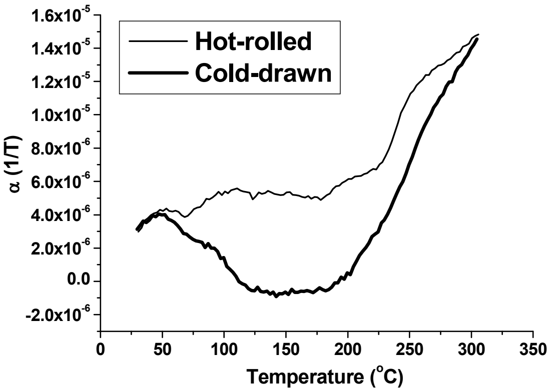

Fig. 8 Thermal expansion coefficient measured before and after cold working process of the Invar alloy

IV. Conclusions

The phase equilibrium of Fe-36Ni base Invar alloy was investigated in this study. Yield and tensile strength significantly increased with increasing Al content. High tensile strength above 1000 MPa was obtained in all cases and high strength of 1200MPa was obtained in the specimen with Al content of 5wt.%. By cold deformation, yield and tensile strength of A1 alloy increases dramatically. Very high strength of 1400 MPa was obtained by cold rolling. Addition of aluminum seriously deteriorates CTE, especially at the ambient temperature range where almost applications of Invar alloy are conducted. Interestingly, addition of Al by 1wt.% shows very similar values of CTE to those of the ingot without Al.

Acknowledgment

This research was supported by Basic Science Research Program through the National Research Foundation (NRF) funded by the Ministry of Education, Science and Technology. (No. 2011-0013839).

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