Effect of aluminum additions on microstructures and mechanical properties of Ti micro-alloyed steels containing Ni subjected to simulating HAZ thermal cycle

Amer Eid Amer[1*], Min Y. Koo[2], Soon H. Hong[2]

[1]*Faculty of Engineering, Beni-Suef University, Beni Suef, 62511, Egypt*
[2]*Composite material Dept., KAIST, Daejon, 34141, Korea*

[*]Corresponding author, E-mail: dramer.eid@gmail.com; Tel: (+2) 01283228922

Received: 31 March 2016, Revised: 02 August 2016 and Accepted: 30 November 2016

DOI: 10.5185/amp.2017/110
www.vbripress.com/amp

Abstract

This work reports the results of an investigation of micro-alloyed steel plates subjected to high heat input (4.5 kJ/mm), simulating the thermal cycle of heat affected zone (HAZ) conducted by induction heating in a computer controlled weld thermal cycle simulator. Three samples of steels having Carbon and Nickel content of 0.08 and 0.8 wt. %, respectively, as well as different Aluminum contents of 0.004, 0.026, and 0.057 wt. %, were investigated. The variation in microstructure, hardness and crack tip opening displacement (CTOD) fracture toughness properties with Al contents were evaluated, and compared with another set of the three samples of steel having the same chemical compositions but with neglected amount of Ni content. The fracture toughness tests revealed the decrease in their CTOD values due to increasing Al contents. In contrast, the hardness measuring revealed the increase of the hardness due to increasing Al wt. %. On the other hand, it was found that the presence of 0.8 wt. % Ni promotes the formation of finer microstructure at a high heat input of 4.5kJ/mm, which leads to higher fracture toughness CTOD value(δ =1.05), which is two times the value of those which have traces of Ni content(δ =0.52). However, the hardness test results showed similar values in each case regardless the amount of Ni contents. Meanwhile, the aspect ratio of the nucleated acicular ferrite was found to be increasing together with the refinement of its grain size due to Ni addition. Hence, it can be concluded that such development and the specific additions of Aluminum positively affects the microstructures and mechanical properties of the investigated steels and their used as marine structural steel. Copyright © 2016 VBRI Press.

Keywords: Micro-alloyed steel, simulated HAZ microstructure, acicular ferrite, Ti and Al deoxidized, CTOD fracture toughness.

Introduction

The major impetuses for developments in the low carbon micro-alloyed steels have been provided by the need to improve weldability of these steels. In particular, steels that can be welded with high heat input are usually preferred since they represent a potential for reduced fabrication costs. However, the current trend towards the use of stronger steels and heavier sections has led to an increased emphasis on the HAZ toughness which tends to deteriorate at high heat inputs [**1**]. Accordingly, a new class of low carbon micro-alloyed steels with Aluminum and Nickel additions, which utilizes the concept of intergranular nucleation of acicular ferrite as means to improve the HAZ toughness in the as-welded condition, has emerged over the last years [**2-4**]. These steels are not only aluminum-killed, but are instead deoxidized with titanium to produce a relatively coarse distribution of complex Ti-oxide inclusions within the base metal. During the high heat input welding (> 4 kJ mm^{-1}), the oxides will not retard austenite grain growth, as compared to TiN, but instead act as favorable nucleation sites for acicular ferrite within the interior of the austenite grains [**5**], and thereby refine the effective grain size.

Fortunately, the intragranular acicular ferrite (IAF) could be formed by the dispersed fine nonmetallic inclusions within austenite grains, which has a chaotic crystallographic orientation, resulting in a retardation of the propagation path for a cleavage crack in steel. Therefore, the approach for obtaining more IAF grains has long been used to refine the microstructure, especially in the heat-affected zone (HAZ) of welded structures refinement [**6**]. However, when the inclusion number density is quite low (< 104 particles per mm^3), the microstructure is significantly coarser than that normally observed in the steel weld deposits [7-16]. Nevertheless, their presence contributes to suppress the formation of Widmanstätten ferrite and upper bainite within the grain coarsened HAZ, which is a common problem with the traditional micro-alloyed steels [1, 5].

It was reported that an increase in the AF content in the microstructure leads to significant improvements in mechanical properties. They attributed these improvements to different propagation paths of cleavage cracks in the presence of acicular ferrite [7]. Furthermore, it is now well established that the nucleation of AF is mainly affected by steel composition, cooling rate, austenite grain size and the presence of non-metallic inclusions [7].

Therefore, in this investigation, six micro-alloyed steel samples (containing Ti) have been investigated. Three of them have different Al contents of 0.026, 0.004, and 0.057 wt. %. The other three are similar but containing 0.8 wt. % Ni, as well.

The main objectives of this study are: - to apply variation in Al contents, to study the effect of this variation in the microstructure evaluation, and hence emphasizing the relationship of HAZs toughness as well as their hardness due to Al content variation.

Experimental

Materials

In this investigation low carbon Nb-Ti micro-alloyed steels were delivered by Pohang steel company POSCO, South Korea. The chemical compositions of the investigated steels are shown in **Table. 1** and equivalent carbon content was calculated according to the formula below:

$$Ceq = C + \frac{(Mn + Si)}{6} + \frac{(Ni + Cu)}{15} + \frac{(Cr + Mo + V)}{5}$$

Table 1. Chemical composition of the investigated alloys.

Alloy	C %	Si %	Mn %	Ni %	Sol. Al%	Ti %	Nb %	N ppm	C eq%
1	0.082	0.16	1.44	0.35	0.006	0.009	0.012	39	0.362
2	0.078	0.15	1.45	0.35	0.034	0.013	0.012	37	0.359
3	0.080	0.15	1.45	0.35	0.063	0.014	0.012	36	0.362
4	0.078	0.15	1.58	0.80	0.004	0.008	0.011	35	0.406
5	0.080	0.14	1.55	0.81	0.026	0.012	0.011	33	0.413
6	0.078	0.16	1.54	0.80	0.057	0.014	0.012	32	0.404

Characterization

The steels had simulated HAZ double-stages thermal cycle. During the experiments, thermal cycles with low and high rate net heat inputs ranging between 0.7 and 4.5 kJ/mm were applied, which is the most widely applicable range in welding of HSLA steel grades.

Optical metallographic samples, prepared by conventional grinding and polishing techniques and etched with 2% Nital solution, were examined under a light microscope. To reveal their overall microstructure and to evaluate their different micro-constituents by using image analyzer and Scanning electron microscopy (SEM) were used as well. Thin foils for transmission electron microscopy (TEM) were prepared by twin jet polishing in an electrolyte of 90% acetic acid and 10% perchloric acid. Thin foil samples were examined in a Transmission Electron Microscope (Philips CM200 with EDAX) at 200 kV operating voltage. Hardness testing was carried out by Vickers hardness machine AKASHI/ Mitutoyo, at a load of 1 kg.

The CTOD test was used to determine the fracture toughness properties of the investigated steel samples. The test was carried out at -10^{o}C. To prepare a specimen for CTOD test according to ASTM E1920, a notch was machined in the center of the specimen and then an actual fatigue crack is carefully induced at the sharp point of the notch. Initially, average 2.5 KN with 2 KN amplitude was loaded from 3.2mm to 3.5mm and then average 1.1 KN with 0.9 KN amplitude was loaded about 1mm ~ 2mm as a fatigue crack. The actual test was performed by placing the specimen in 3-point bending and accurately measuring the amount of the crack opens. For this purpose, a strain gauge was employed, mounted to a clip between two precisely placed knife edges at the mouth of the machined notch. The CTOD fractured samples' surfaces were examined by means of SEM to clarify the different morphologies and fracture aspects.

Results and discussion

CTOD & Hv testings

Results of CTOD & Hv tests are illustrated in **Table. 2**, in the case of high heat input of 4.5 Kj/mm, which corresponds to the rate of cooling of $\Delta t_{8/5} = 61s$.

The results showed that CTOD values of steel samples 1 and 3 decreased from δ = 0.52 to δ = 0.17 mm by increasing the Al contents from 0.006 to 0.063 %, respectively. However, CTOD values of Ni-containing steel samples 4 and 6 decreased from δ = 1.05 to δ = 0.76 mm, when the Al contents increased from 0.004 to 0.057 %, respectively. On the other hand, Hardness Vickers (Hv) was found to change in different mode. As the hardness values increased by increasing Al contents in steel samples 1, 2 and 3 from 183, 200 and 202 Hv, respectively, it was found that the hardness results were observed to be virtually constant in the case of Ni-containing steels, even though different ratios of Al contents.

Table 2. CTOD and Hardness Hv Results.

Sample	CTOD	Vicker Hv
1H	0.52	183
2H	0.44	200
3H	0.17	202
4H	1.05	185
5H	0.95	195
6H	0.76	192

Alloys Chemical Design

Since the main aim of this investigation is to emphasis on the HAZ toughness which could be deteriorated at high heat inputs, the new developed class of low carbon micro alloyed steels, which utilizes the concept of intergranular nucleation of acicular ferrite as a means to improve the HAZ toughness in the as-weld condition, has been designed to serve that purpose. These steels are not only aluminum-killed, but also deoxidized with titanium to produce Ti-oxides, to act as favorable nucleation sites for acicular ferrite within the interior of the austenite grains, and thereby refine the effective grain size. Furthermore, the effect of different Al contents in such steel, as 0.004, 0.034, and 0.063%wt, were proposed to investigate the possible developments of the microstructure, as well as understanding the role which can be played by Al in influencing the strength of ferrite. In coincidence, the medium Al ratio (0.03%wt.) makes the amount reported as the content of traditional steel Al deoxidized, and the others are lower and higher than this ratio. The fracture toughness tests revealed the decrease of its CTOD values by increasing Al contents. In contrast, the hardness measuring presented the increase of the hardness by increasing Al wt%. Eventually, as it had been reported [**18**], one of the main factors which affect the formation of integranular acicular ferrite is the hardenability, three of these steel samples that contained 0.8 %wt. Ni (for the reason of hardenability effect) had been studied and compared with those having traces of Ni content. The results demonstrated that the presence of 0.8 nickel promotes the formation of finer microstructure at high heat input of 4.5kJ/mm, which leads to a higher fracture toughness CTOD value which is two times the value of those which have very small amount of Ni content. However, the hardness results showed similar values in each case regardless the amount Ni contents.

Microstructure investigation

Effect of Al on Microstructure:

Optical micrographs in **Fig. 1** show the microstructures of Steel samples S1 through S3 simulated HAZ at heat input of4.5 kJ/mm. The microstructure consists of coarse grain boundary- quasi polygonal ferrite formed along prior austenite grain boundaries, intergranularly nucleated acicular ferrite, and upper bainite or widmanstatten ferrite side plate structure with small amount of martensite. When Al content is relatively low, i.e., 0.006 wt pct, the interwoven structure of acicular ferrite within the austenite grains are found. This acicular ferrite structure is believed to be induced by the inoculation of fine titanium oxide particles dispersed in the steel. In details; as the amount of acicular ferrite decreases with increasing Al content, the microstructure of steel S3 with the highest Al content exhibits an overall side plate ferrite and/or upper bainite structure, as given in **Fig. 1(c)**.

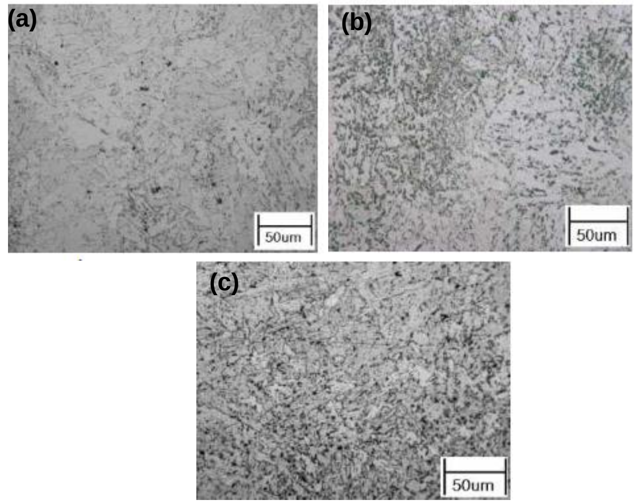

Fig. 1. Microstructure observation of a) steel S1, b) steel S2, and C) steel S3.

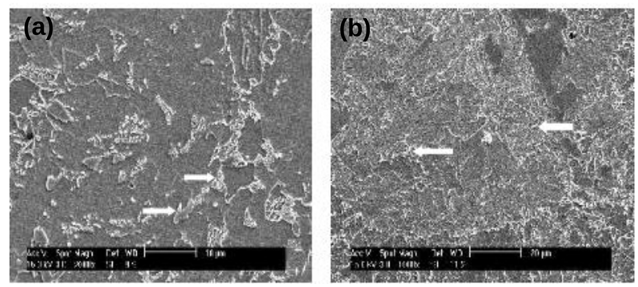

Fig. 2. SEM photos; a) for steel S1 and b) for steel 3.

Therefore, it seems that the inoculation of titanium oxide particles, which intragranularly nucleate acicular ferrite, is not effective at high Al contents. **Fig. 2** shows SEM micrographs comparing the microstructures of steels S1 with the lowest Al content and S3 with the highest, both of which were cooled at the same rate. While coarse pearlite colonies between ferrite laths are observed in steel S1, small microphase particles, instead of pearlite colonies, are observed in steel S3. The TEM with EDX work has confirmed that the microphase in steel S3 consists mainly of cementite together with a very small amount of martensite. In general, carbon atoms are distributed from transformed ferrite to remaining austenite when the austenite- ferrite transformation proceeds in a temperature range where diffusion is somewhat active. This is because austenite has larger carbon solubility than ferrite. Carbon is, consequently, concentrated in untransformed austenite regions between ferrite laths. Then, cementite will precipitate in the carbon concentrated regions during cooling. In the case of high Al content, however, the carbon concentrated regions transform into cementite or martensite because Al promotes the formation of cementite in steels.

In agreement with Lee and Pan [**19**] it was observed that the amount of acicular ferrite decreases with increasing Al content in Ti-containing low carbon steels. They proposed that a small addition of Al decreases soluble oxygen content in molten steels, owing to the deoxidization effect of Al, and then hinders the formation of titanium oxide particles simply by reducing the opportunity for titanium to combine with oxygen. They also proposed that the change in microstructure results from the decrease in the amount of titanium oxide particles by Al addition. The TEM work discloses that Ti2O3 is a dominant oxide particle in steel samples S1 through S3, regardless of Al content, even though it decreases at highest content of Al. These oxide particles are believed to be Ti and Al composed particles, covered by MnS as shown in **Fig. 3**. As it had been reported [**20-22**], most oxide particles are analyzed as mixtures of Ti2O3 and Al2O3. It seems that increasing the Al content reduces the amount of Ti in these oxide particles, i.e., Ti2O3. Because Al has a greater oxygen affinity than Ti, Al2O3 is likely to replace Ti_2O_3 at high Al contents. In this study, it has been found that oxide particles consist mainly of Al_2O_3 when the Al content is higher than about 0.004 wt pct. With increasing Al content, the amount of Ti_2O_3 decreases and that of Al_2O_3 increases. As previously mentioned, Ti_2O_3 particles absorb Mn from surrounding matrices and form $MnOTiO_2$ around them. The Mn absorption is associated with cation vacancy in Ti_2O_3 [**20**]. In other words, Mn atoms are absorbed into cation vacancy sites in Ti_2O_3. Since Al_2O_3 is, however, anion vacancy type oxide, it is not expected to absorb surrounding Mn atoms, therefore, Al_2O_3 particles might not provide the nucleation sites for acicular ferrite. As a result, it can be concluded that the transition from Ti_2O_3 to Al_2O_3 with increasing Al content leads to the change in overall microstructure, i.e., the transition from acicular ferrite to upper bainite or Widmanstatten ferrite due to a shortage of effective intergranular nucleation sites. Hence, the hardness will increase and CTOD fracture toughness tends to decrease.

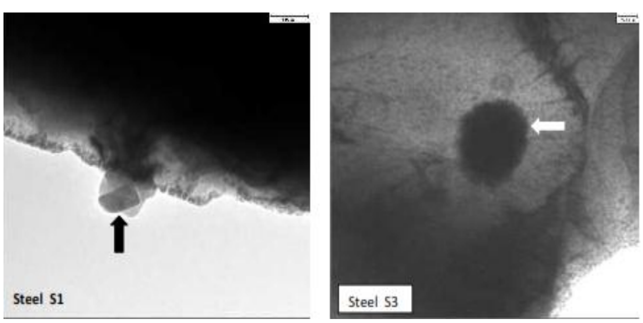

Fig. 3. TEM photo shows the Ti oxides in steel S1 and S3.

Effect of Ni on microstructure:

Adding an amount of 0.8 wt. % Ni in steel samples 4, 5, and 6 led to the refinement of their microstructures. **Fig. 4** shows the microstructure that was developed due to Ni addition in Steel S4 compared with steel S1. However, TEM observations from **Fig. 5** demonstrated that this amount of Ni increased the acicularity of acicular ferrite. Furthermore, the most important effect of Ni addition was found to be on the acicular ferrite (AF) aspect ratio (width/length).

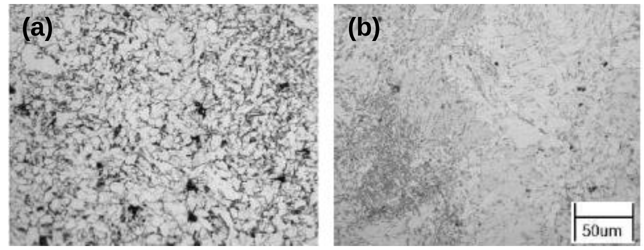

Fig. 4. Optical Microscope photos show the microstructure comparison between a) steel S4 contains 0.8% Ni, and b) S1 without Ni.

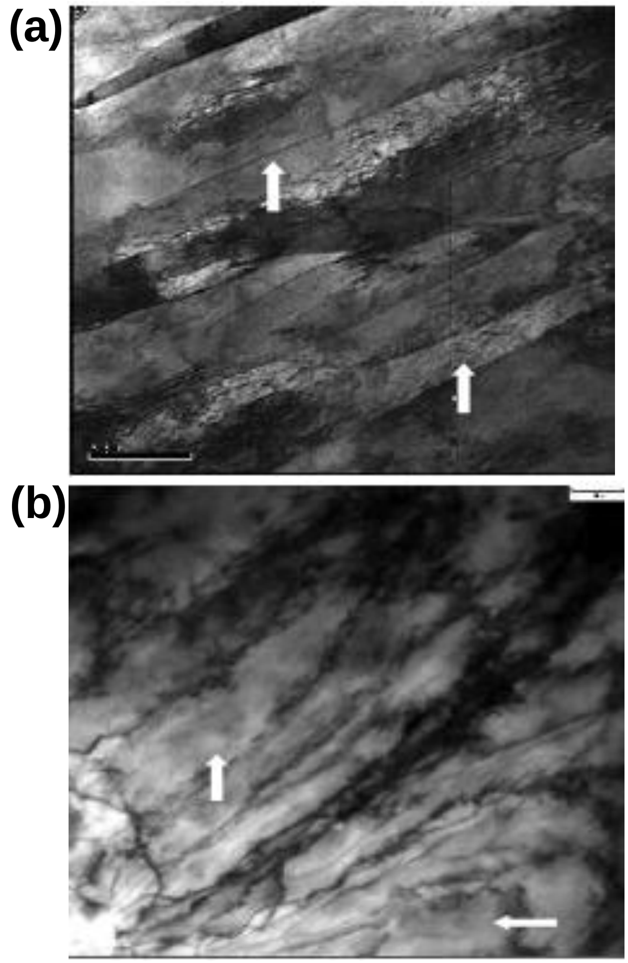

Fig. 5 TEM micrograph shows the effect of Ni additions on acicular ferrite morphology in a) steel S4 has 0.8% Ni, and b) S1 without Ni.

The aspect ratio of the nucleated acicular ferrite was found to increase together with the refinement of its grain size due to Ni addition. Hence, it is believed that such development in the steel structure led to the superior improvement in the fracture toughness of the steel. However, the predominance of acicular ferrite in the microstructure is not the only factor determining the high toughness of the HAZ, many authors have reported that micro phases, specifically the martensite-austenite (MA) constituents can influence strongly the toughness of welded steels. It was found that M-A volume fraction decreased frequently with the addition of 0.8 wt. % Ni in each steel samples. Optical microscopic with image analyzer was used to measure the ferrite grain size and

MA volume fraction and revealed that the addition of Ni promotes the formation of the very fine ferrite at low Al content as shown in **Fig. 4**, as well as, **Fig. 6** which showed schematically the effect of nickel additions on the ferrite grain size and martensite volume fraction as it was evaluated by image analyzer optical microscope tool.

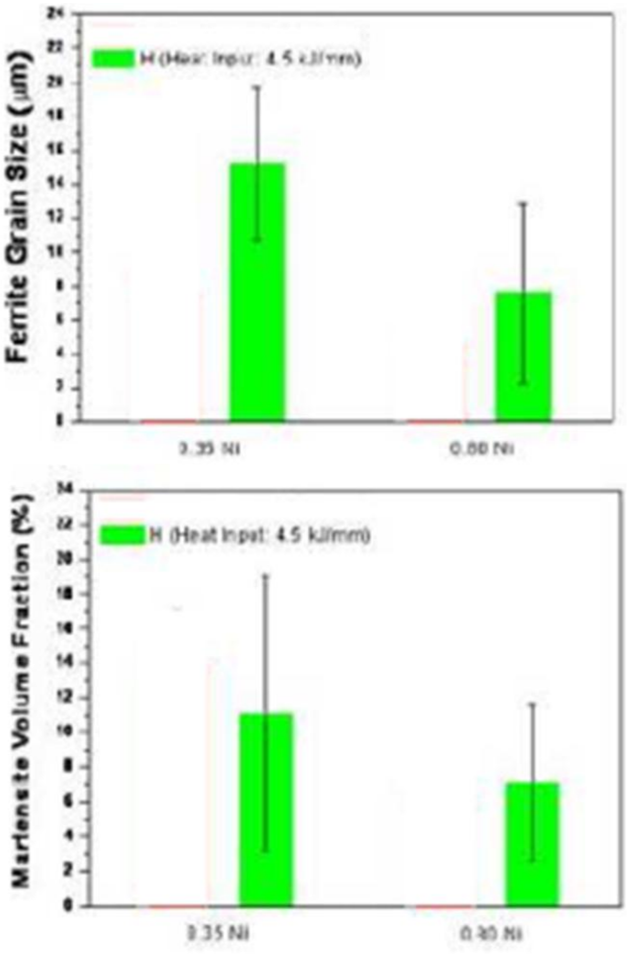

Fig. 6. Ferrite grain size and Martensite volume fraction Schematic diagram showing a comparison between steel sample that contains .8% Ni and another without Ni content.

Conclusion

As emphasized here and before, addition of titanium to the base steel improved toughness of the HAZ by refining the austenite grain size and by changing the transformation products from Widmanstatten ferrite, pearlite, and bainite to more equiaxed ferrite and pearlite microstructure. It has also been found that a small addition of Al influences greatly the nature of the oxide particles. As the Al content increases, most oxide particles change from Ti2O3 into Al2O3. This induces the change in microstructure that the upper bainite or Widmanstatten ferrite sideplate replaces acicular ferrite. Therefore, it is very important to control even minor changes of Al contents in Ti-containing steels, in order to obtain the well-developed microstructures of acicular ferrite. However, additions of (Al) to the base steel degraded the HAZ toughness, whereas the addition of Aluminum with the presence of 0.8% Ni restored the toughness to the values to become double of those in the case of the same steel free Ni, even though it contains Ti. In the case of high heat input, the addition of Al produced quasi polygonal ferrite with some martensite increases, by increasing Al contents, whereas Al plus Ni produced a more equiaxed ferrite, or acicular ferrite increase, by increasing Al contents. A steel containing 0.03% Al plus 0.8% Ni exhibited optimum combination of strength and CTOD fracture toughness.

Acknowledgements

The authors are grateful to the management of POSCO Steel for providing research materials of this investigation. In addition we are in debt to the Post Doc. Fellowship program of the KOSEF, Republic of Korea, for sponsoring the visit of Dr Amer.Eid Amer, Egypt at the Korea Advanced Institute of Science and Technology (KAIST) – Dept. of Materials Science and Engineering.

Author's contributions

Conceived the plan: Soon H. Hong, Amer Eid Amer; Performed the expeirments: Amer Eid Amer, Min Y. Koo; Data analysis: Amer Eid Amer, Min Y. Koo; Wrote the paper: Amer Eid Amer. Authors have no competing financial interests.

References

1. Aihara, S.; Shigesato, G.; Sugiyama, M.; Uemori, R.; NIPPON STEEL TECh. Report. No. 91, January **2005**,43.
2. Shigesato, G.; Sugiyama, M.; Aihara, S.; Uemori, R.; Tomita,Y. *Tetsu-to-Hagane Journal*., **2001**, 2, 93.
3. Lee, J. L.; *Acta Metall Mater.***1994**, *42(10)*, 3291.
4. Shanmugam,S.; Misra, R.D.K.; Hartmann,J.; Jansto,S.J. *Mater. Sci. Eng.A.* **2006**, *441*, 215.
5. Junhua,K.; Lin,Z.; Bin,J.; Pinghe,L.; Aihua,W.; Changsheng,X. *Mater. Des*. **2004,** *8*, 723.
6. Li,X.; Min,Y.; Liu,C.; Jiang,M. *Steel Research int.* **2015**, *86*, 999.
7. Loder, D.; Michelic, S. K.; Mayerhofer, A.; Bernhard, C.; Dippenaar,: Materials Science and Technology Conference and Exhibition 2014, MS and T, **2014**, 469.
8. Contreras, A. ; Albiter, A. ; Salazar, M. ; Perez, R. ; *Mater. Sci. Eng. A*. **2005**, *407*, 45.
9. Zhao, M.-C.; Yang, K.; Shan, Y.; *Mater. Sci. Eng. A* ,2002, *335*, 14.
10. Xiao, F-. R.; Liao, B. ; Shan, Y.-Y.; Qiao, G-.Y.; Zhong, Y.; Zhang, C.; Yang, K. *Mater. Sci.Eng.A*. **2006**, *431(1-2)*,41.
11. Kim, Y.M.; Kim, S.K.; Lim, Y.J.; Kim, N.J.; *ISIJ Int.* **2002**, *42(12)*,1571.
12. Hwang,B.; Lee, S.; Kim, Y.M.; Kim, N.J.; Yoo, J.Y.; Woo, C.S. *Mater. Sci. Eng.A*. **2004**, *368*, 18.
13. Hwang, B.; Kim, Y.M.; Lee, S.; Kim, N.J.; Ahn, S.S.; Metall. Mater. Trans.A. **2005**, *36*, 371.
14. Hwang, B.; Kim, Y.M.; Lee, S.; Kim, N.J.; Ahn, S.S.; Metall. Mater. Trans.A. **2005**, *36*, 725.
15. Zhao, M.-C.; Yang, K.; *Scripta Mater.* **2005**, *52*, 881.
16. Elwazri,A.M.; Varano,R.; Siciliano,F.; Bai,D.; Yue,S. *Metall. Trans.A*. **2005**, *36*, 2929.
17. Zhao, M.-C.; Hanamura, T.; Qui, H.; Yang, K.; *Mater. Sci. Eng.A.* **2005**, *395*, 327.
18. Takahashi, A.; Iino, M. *ISIJ Int.***1996**, *2*, 235.
19. Lee, J.L.; Pan, Y.T.; *Metall. Trans.A*. **1993**, *24*, 1399.
20. Shim, J.H. ; Byun, J.S.; Cho, Y.W.; Oh, Y.J.; Shim, J.D.; Lee, D. N. *Metall. Mater. Trans.A.* **2001**, *32*, 75.
21. Goto, H. ; Yamaguchi, K. ; Ogibayashi, S. ; Kenichi, M. *Tetsu-to-Hagane´*. **1997**, *83*, 833.
22. Zhuo , X.; Wang, X. ; Wang, W. ; Geon, L. H. *J. of Uni. of Sci. and Tech. Beijing, Min, Metall, Mater.*, **2007**, *14*, 14.