Heterogeneous microstructure of low-carbon lath martensite with continuous yielding behavior in Fe-C-Mn alloys

M Sugiyama\textsuperscript{1}, G Sawa\textsuperscript{1,2}, K Hata\textsuperscript{2}, N Maruyama\textsuperscript{2}

\textsuperscript{1} Department of Materials Science, Osaka University, Nippon Steel Research Alliance Labs., 2-1 Yamadaoka, Suita 565-0871, JAPAN
\textsuperscript{2} Advanced Technology Research laboratories, Nippon Steel Corporation, 20-1 Shintomi, Futtsu 293-8511, JAPAN
e-mail: sugiyama@mat.eng.osaka-u.ac.jp

Abstract. High strength steels have been rapidly developed for several kinds of application fields such as light weight automobile bodies, multistory buildings, and oil well tubulars. With an increase of strength, the use of the low-carbon lath martensite phase puts this component as the main microstructure, or a part of the dual phase one. However, the martensite is a non-equilibrium phase, and the microstructure is also a heterogeneous composite-like one including of fine carbides and retained austenite. In order to understand the hierarchical structure, multi-scale microscopic techniques have been utilized. Internal twins in a lath and some large lath domains with fine carbides are revealed by scanning electron microscope (SEM) and transmission electron microscope (TEM) observations, although a conventional low-carbon martensite is known to include mainly dislocations in the lath structure. The heterogeneous microstructure is discussed based on a model of the local difference of transformation temperature of each lath block segment during cooling. On the plasticity of low-carbon lath martensite, the continuous yielding behavior is one characteristic property, and the elastic limit is obtained by the cyclic tensile test. The heterogeneous deformation behavior is detected as a periodic strain partitioning among lath blocks in a grain by digital imaging correlation analysis using \textit{in situ} SEM tensile testing.

1. Introduction
The majority of steels for automobile bodies and undercarriage components have been replaced by high strength steels with more than twice the strength as previous ones [1]. With further increasing demand of high strength steels in the market, the practical use of martensite phase is expected in material design of high strength microstructure. Although the martensite phase is already utilized in a dual phase and transformation-induced plasticity steels, and others, most products including the martensite phase are used after a tempering treatment, and it is still difficult to control the mechanical properties of fresh martensite quenched in low carbon steels.

From the viewpoint of industrial application, the low carbon lath martensite is most attractive microstructure with high strength and good weldability, however, the ductility is not sufficient. Fundamental knowledge of low carbon lath martensite is required to improve mechanical properties such as the yielding characteristics and elongation. It is well known that the morphology and internal structure of martensite are changed with carbon content in Fe-C alloys [2-3], and the crystal orientation relationship between the matrix and lath are made clear under several transformation conditions [4-6]. When the structure of the low carbon steels is considered, the microstructure may be categorized by three different strategies. The first factor is the mixing of the different phases, composed of the martensite, retained austenite, proeutectoid ferrite, and pearlite, with dependence of cooling velocity
from austenite phase, because the Ms temperature of lath martensite is relatively high, e.g. around 673K. The second factor is the composite structure of the lath microstructure and carbides including carbon clusters, because the Mf temperature is higher than room temperature, and carbon atoms can move even at the room temperature. The third factor is the hierarchical microstructure of lath martensite itself, such as packets, blocks, sub-blocks, and the ‘laths’.

Recently, the morphology and crystallography of low carbon lath martensite have been studied using 2D and 3D EBSD analysis, and it is made clear using variant analysis that some of characteristic variants are formed in a same block and packet, based on the K-S orientation relationship [7-10]. It is further revealed that the sub-block boundaries are formed in a block, in addition to the conventional block regions. On the strengthening mechanism, both block and sub-block boundaries act as a barrier for dislocation motion as seen in uniaxial micro-tensile tests [11]. It is also reported in Fe-0.13C-5.1Ni(wt%) that the lath martensite microstructures are composed of coarse laths of various sizes embedded in a matrix of conventional thin laths. Judging from the morphology, dislocation density and the growth area connected to the prior austenite grain boundary, it has been proposed that the coarse lath domain is firstly transformed at around the Ms temperature from the soft austenite matrix, which is reasonable regarding the intrinsic martensitic transformation sequence on the heterogeneity [12].

The main purpose of the present study is to make clear the real lath microstructure in Fe-C-Mn alloys, especially in the low carbon content region, based on the microstructure reported using lath martensite having a low Ms temperature below room temperature in Fe-Ni-C alloys. The defect structure of the lath martensite has been investigated by TEM observations. The heterogeneous microstructure of lath martensite is further discussed with region showing the gradual yielding behavior.

2. Experimental procedure
An ingot of 50kg in weight was melted in a laboratory vacuum furnace. After hot forming, a homogenizing heat treatment was carried out at 1373K for 1hr, and the slab roughly hot-rolled from 30 mm to 3.4 mm in thickness. After removing the surface decarburized layer, the plate was cold rolled to 1.2 mm in thickness. Two kinds of model plates with compositions Fe-0.07C-1.0Mn(wt%) and Fe-1.0C-2.0Mn(wt%) alloys were prepared by almost similar procedures. The small amount of additional Mn has an effect to control the ferrite nose in the TTT diagram. Each sample plate are given different heat treatments and cooling conditions to obtain the lath martensite microstructure.

Two kinds of different sized samples for tensile tests were cut by electric discharge machine, one for the tensile tests in an Instron-type universal testing machine and the other for the in situ tensile testing in a SEM. A suitable sample surface was prepared by electron polishing using 95% acetic acid and 5% perchloric acid for SEM observations. To obtain the strain mapping, the sample surface was etched in 1% nital, and in situ SEM observation carried out. The strain mapping with a change of displacement position of each surface point (digital imaging correlation method) has been carried out in SEM. The SEM used is a Hitachi S-5000 equipped with an EBSD detector for crystal orientation analysis. The TEM observation has been done using thin foil samples after electron polishing by conventional FE-TEM with an accelerating voltage of 200kV.

3. Results
The tensile strength of the samples used is about 1100MPa, and the elongation is 5~6%. There is no clear yield point on the stress-strain curves obtained from all samples, resulting in a continuous hardening behavior on the curves under tensile testing at room temperature. With a cyclic tensile test by 20MPa each, it is confirmed in Fe-0.07C-1Mn(wt%) alloy that the initial deformation is elastic and the elastic limit appears at around 180~200MPa, as seen in figure 1(a)(b). Each stress-strain curve is superimposed in the same graph of figure 1(a), and the stress and strain profiles are plotted independently as a function of test time in figure 1(b). Although the negative value of the stress is artefact in figure 1(b), the stress value goes back to zero until stress condition of 160MPa when it is unloaded. Then, the plastic strain is residual more than the 180MPa stress condition, as described in figure 1(b).
The microstructure of lath martensite has been investigated by SEM and TEM observations. There is a slight amount of ferrite grains at the prior austenite boundary in Fe-0.07C-1Mn (wt%), but the main part of the microstructure consists of the hierarchical structure such as packets, blocks and laths in the prior austenite grain. Figure 2 is a typical back scattering electron (BSE) image taken by SEM; showing packets and blocks in a prior austenite grain. The prior austenite grain size is about 80 µm, the block and its inner lath microstructure are observed in region A and B, and large lath domains with more than 5µm in width are observed in region C.

Figure 3 is an enlarged BSE image of area A in figure 2. As indicated by the scale bar of 0.5 µm, a characteristic lath structure of 0.2 µm in width is shown in blocks indicated by P and Q. It is noted that the different variant of R is observed in the area adjacent to Q, although Q and R is considered as a same block region. The width of regions S and T is estimated at about 2～4 µm, and there is no lath structure, where small precipitates such as carbides are observed. In the narrow region of U, there is a fine stripe contrast showing an internal twin, which is clearly identified by the following observations.

The internal defects of lath martensite in low carbon steels have been studied by TEM observations. The typical microstructure of the lath martensite contains a high density of dislocations and tempered fine carbides in both of the Fe-0.07C-1Mn and the Fe-0.1-2Mn alloys. In both samples, however, internal twins are also observed together with dislocations, as seen in figure 4(a)(b). The thickness of internal twins is several tens of nm, and some of them are penetrated from end to end in a lath domain.
as shown in bottom area of figure 4(a), and others are introduced inside a lath structure, as seen in figure 4(b). Although it is difficult to estimate the amount of internal twins quantitatively, the internal twins are sometimes observed even in the Fe-0.07%C-1%Mn alloy. In comparison with the amount of internal twinning in samples quenched with 500°C/sec and 1500°C/sec cooling rate, the amount of internal twins tends to increase in the sample quenched by faster cooling speed. With slight composition difference between Fe-0.07%C-1%Mn and Fe-0.1%C-2%Mn alloys, the internal twins in lath martensite are observed more in Fe-0.1%C-2%Mn. With increasing Mn content, the ferrite nose leads to the delayed direction on the phase transformation, which gives a similar effect to increasing the cooling speed.

The nature of the internal twins is investigated by conventional diffraction analysis under TEM observations, as seen in figure 5. Figure 5(a) is a dislocation structure of a lath martensite, in which the dashed line indicates the lath boundary. When the sample is tilted in the same area, the internal twins appear in the lath microstructure, as shown in figure 5(b). The internal twins are always coexistent with the dislocations in the low lath martensite. The tilting experiment is important to confirm the internal twins under TEM observation. By the diffraction analysis shown in figure 5(c)(d), the twin plane is
identified to be (112), in which the twin plane is satisfied with the edge-on condition with an incident beam direction of [-311]. The thickness of the twins is very thin such as several tenth of nm, resulting in the strong streaks on the diffraction pattern of figure 5(c).

As already seen in figure 1, the clear yielding point on the stress-strain curve is not observed for low carbon lath martensite. That indicates a heterogeneous deformation process in the early stage of deformation. The initial deformation behavior is considered to be very sensitive to the microstructure. The digital imaging correlation is a powerful technique to identify the morphological change in lath microstructure. The markers for measurement of small displacements uses directly the surface roughness of lath martensite after a nital etching treatment. An initial microstructure of lath martensite has been observed by SEM in Fe-0.07wt%C-1.0wt%Mn alloy, and the sample is deformed using a tensile test stage in the SEM until 0.4% strain, then the secondary electron image is taken from the same area, as indicated in figure 6(a). Based on the two micrographs, the strain mapping is calculated as seen in figure 6(b). It is made clear by the mapping data that the color change showing initial strain is distributed by the unit dimension of block noted by yellow arrows in lath microstructure.

4. Discussion

It is characteristic that the internal twins are observed even in a low carbon lath martensite with less than 0.1wt%C in Fe-C-Mn alloys. In Fe-C alloy systems, the martensite microstructure is of lath type in the carbon range of less than 0.6wt%, and the lens and thin plate martensite having internal twin structure appear in Fe-C(>0.6wt%), whose Ms temperature is lower than 623K. The density of twins changes from a partial to a whole region of lens martensite on decreasing of the Ms temperature, the internal twins are introduced instead of the dislocation, as a result of the lattice invariant shear of (101)[10-1]. Thus, the internal twins of lath martensite in the low carbon martensite are considered to be introduced as the lattice invariant shear in the area transformed at low temperature below 623K. Since the Ms temperatures of Fe-0.07wt%C-1wt%Mn and Fe-0.1%C-2%Mn( wt%) are estimated to be 739K and 689K by the following equation; Ms=546exp(-1.362C)-30.4Mn-11Si [13], the Mf temperatures are expected to be lower than 623K. If the lath martensite with internal twins is transformed at low temperature in comparison with surrounding lath martensite area, the austenite matrix will be plastically deformed by the dilation component of the surrounding transformation. This is the reason why the internal twins in low carbon martensite are always coexistent with dislocations. On the other hand, there are lots of carbide in the large lath martensite, and it may be considered that the large lath area is transformed at high temperature, as proposed by Morsodrof [12]. This will be further studied in detail.

On the issue of mechanical properties, it is important whether the internal twins play a role of obstacles for dislocation motion, resulting in a negative factor to improve the ductility. The amount of
internal twins in low carbon lath martensite is however not so high as affect the mechanical properties. The existence is nevertheless valuable for consideration of the transformation mechanism of lath martensite.

The elastic limit showing round shaped yielding behavior is estimated by the cyclic tensile test in the present study. There are some softened areas in the microstructure where dislocations will start to move at low stress. It is calculated by Allen that local softening phenomenon leads to round shaped stress-strain curve [14]. The local softened area in a lath microstructure and different transformation temperature of each lath domain cause the different strength of each block domain. In the present study, the initial deformation of low carbon martensite occurs at the unit of a block dimension. Because of the resolution limit of the present DIC method, it is difficult to make clear the exact locally location of the softened region, but the heterogeneous deformation behavior takes place at each block microstructure. Since the block is a group of almost same lath orientation, it is considered that the local softening regions originated from carbon segregation around dislocations, or precipitation of carbides and/or clusters are similarly distributed among blocks on cooling and auto tempering.

5. Conclusions
The microstructure of lath martensite in Fe-lowC-Mn alloys have been investigated by several kinds of microscopy in a multi-scale dimension study, and the following conclusions are obtained.

(1) The multi-scale microscopic observations reveal that the microstructure of lath martensite in low carbon steels consists of heterogeneous lattice defects such as dislocations, internal twins, and different types of carbides, although the internal defect content of lath martensite is considered to be mainly dislocations in Fe-C-Mn alloys.

(2) A large size of lath martensite is often observed under the SEM observations, which is of similar dimension to the conventional block region defined as the hierarchical structure of low carbon lath martensite.

(3) The characteristic heterogeneous microstructure is related to the continuous yielding behavior on the stress-strain curve. It is confirmed by the DIC method that the initial deformation under tensile test in SEM is occurred by the different magnitude among each block regions.

Acknowledgement
A part of this work was supported by “Advanced Characterization Nanotechnology Platform, Nanotechnology Platform Program of the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan” at the Research Center for Ultra-High Voltage Electron Microscopy (Nanotechnology Open Facilities) in Osaka University.

References
[1] Senuma T 2001 ISIJ Int. 41 520-532
[2] Krauss G 1999 Mat. Sci. and Eng. 40-57
[3] Hutchinson B, Hagnost J, Karlsson O, Lindell D, Tornberg M, Lindberg F and Thuander M 2011 Acta Mater. 59 5845-5858
[4] Wakasa K and Wayman C M 1981 Acta Metall. 29 973-990
[5] Sandvik B P J and Wayman C M 1983 Metall Trans. A14 809-822
[6] Kelly P M, Jostsons A and Blake R G 1990 Acta Metall. Mater. 38 1075-1081
[7] Morito S, Tanaka H, Konishi R, Furuhara T and Maki T 2003 Acta Mater. 51 1789-1799
[8] Kitahara H, Ueji R, Tsuji N and Minamino Y 2006 Acta Metall. 54 1279-1288
[9] Morito S, Huang X, Furuhara T, Maki T and Hansen N 2006 Acta Metall. 54 5323-5331
[10] Kinney C C, Pytlewski K R, Khachaturyan A G and Morris Jr. J W 2014 Acta Mater. 69 372-385
[11] Mine Y, Takashima H, Matsuda M and Takashima K 2013 Mater. Sci. Eng. A 560 535-544
[12] Morsdorf L, Tasan C C, Ponge D and Raabe D 2015 Acta Mater. 95 366-377
[13] Perlade A, Bouaziz O and Furnemont Q 2003 Mater. Sci. and Eng. A356 145-152
[14] Allain S, Bouaziz O and Takahashi M 2012 ISIJ Int. 52 717-722