Deformation and recrystallization behavior of super high-purity niobium for SRF cavity

Y Yamaguchi¹, H Doryo¹, M Yuasa², H Miyamoto² and M Yamanaka³
¹ Graduate School of Science and Engineering, Doshisha University, Japan.
² Department of Mechanical Engineering, Doshisha University, Japan.
³ Mechanical Engineering Center, High Energy Accelerator Research Organization (KEK), Japan.

E-mail: duq0576@mail4.doshisha.ac.jp

Abstract. Deformation and recrystallization behavior of pure niobium was investigated in order to clarify the origin of its low hydro-formability despite of its high ductility comparable with pure iron. It was found that pure niobium exhibits lower strain hardening in cold rolling compared with pure iron. Furthermore, in post-deformation annealing, the hardness of niobium decreased monotonously with an increase of temperature, and the typical sharp drop by recrystallization was not evident. This softening behavior was contrasted with the high-purity iron. It is suggested that niobium exhibit the so-called in-situ recrystallization possibly because of low elastic modulus and low accumulative plastic strain energy in spite of high melting temperature. The low hydro-formability of pure niobium sheets or tubes is caused by its low strain hardening and its unique plastic anisotropy which is associated with this recovered residual rolled texture.

Keywords: texture, in-situ recrystallization, elastic modulus

1. Introduction
Fabrication of superconducting radio frequency (SRF) cavities of high-purity niobium have been tried by hydroforming using a seamless tube for the International Linear Collider (ILC) project[1]. While the conventional fabrication methods of accelerator cavity consist of press forming of half cells from sheets and coupling of an electron beam welding, the hydroforming has advantage in high productivity. However the low strain hardening and related low yield ratio with about 30% is the main reason of insufficient formability for bulging formation. In order to commercialize the system by hydroforming, it is necessary to improve the formability of the niobium materials. The insufficient formability of niobium is due not only to low strain hardening but also to low plastic anisotropy related with non-uniform microstructure and texture. The non-uniform texture is also responsible for low r-value and ridging[2-5]. Therefore, it is necessary to improve the non-uniform microstructure and texture for uniform strain distribution for hydroforming to cover the disadvantage of low strain hardening, but there are little studies on this viewpoint. In this study, we therefore aimed to investigate the origin of low strain hardening and non-uniform microstructure and texture formation of high-purity niobium after the cold rolling and annealing focusing on dislocation structure and recrystallization behavior.
2. Experimental Procedure
A high-purity niobium of RRR 464 was used as a test material. In addition, we used a high-purity iron with 99.99% purity as a comparison material. We performed cold rolling process using a two-stage rolling mill for these materials. Maximum rolling reduction was to 95%. After cold rolling, we annealed these specimens in a vacuum using an image furnace equipment (MILA3000UHV ; ADVANCE RIKO, Inc.). After these processes, we conducted hardness test using a micro Vickers hardness tester (HMV-1 ; SHIMADZU Co.) to examine the relationship between rolling reduction and hardness and the recrystallization temperature. Subsequently, we observed the microstructure using an optical microscope (OPTIPHOT-100 ; NIKON Co.) after rolling and annealing. We measure crystal orientation by electron back scatter diffraction (EBSD) equipped in a scanning electron microscope (SEM, JSM-7001FD ; JEOL Ltd.) and INCA Crystal (Oxford Instuments). Then, we conducted X-ray diffraction (XRD) using a X-ray diffractometer (SMARTLAB ; Rigaku Co.) to evaluate the dislocation density of these specimens.

3. Result and Discussion
3.1 Evaluation of work hardening rate
Figure 1 shows the relationship between the rolling reduction and the Vickers hardness of cold rolled specimens. Both of the cold rolled specimens became harder as the rolling reduction increased, which suggested that work hardening due to cold rolling occurred. In addition, comparing the slope of the hardening curves, the inclination of the high-purity niobium was smaller than that of the high-purity iron. This result suggests that work hardenability of pure niobium is lower than that of pure iron.

![Figure 1. Relationship between rolling reduction and Vickers hardness of the specimens.](image)

3.2 Recrystallization behavior and change of texture
In order to evaluate the influence of the cold rolling process on the recrystallization behavior, we annealed the specimens after cold rolling with reduction of 60% and 95% at various temperatures. Figure 2 shows the hardness of the annealed specimens after heat treatment at each temperature. When the rolling reduction was 95%, the high-purity iron rapidly softened at 773K and softening of the high-purity niobium was completed at 1123K. In other words, the high-purity niobium softened at a higher temperature than the high-purity iron. It is suggested that this difference of softening temperature is due to the melting point. It is known that a recrystallization temperature of a metal material is approximately 0.4T_m to 0.5T_m, where T_m is a melting point of a material expressed in absolute temperature. The melting points of niobium and iron are 2741K and 1809K, respectively and the recrystallization temperatures are expected to be 1096K to 1371K and 724K to 905K, respectively. Because the recrystallization temperature deduced from the softening curve in figure 2 is generally within this range, the difference of the softening temperature is mainly decided by the melting point. When the rolling reduction was 60%, the high-purity iron and the high-purity niobium softened at 873K...
and 1173K respectively. In this case also, there was the difference of softening temperature depending on the melting point. However, the hardness of niobium decreased monotonously with an increase of temperature, and the typical sharp drop by recrystallization was not evident. This softening behavior was contrasted with the high-purity iron. It was suggested that in-situ recrystallization of the high-purity niobium occurred during the annealing process due to factors other than the melting point. Then we observed the texture of the annealed specimens. Figure 3 shows the texture of the specimens after cold rolling with reduction of 60% in the heat treatment process. It was found that the recrystalized microstructure of high-purity iron was equi-axial crystals over the softening temperature. On the other hand, the texture of the high-purity niobium remained elongated in the annealing process. At the completion of softening grain growth of crystals was observed. This suggests that the occurrence of in-situ recrystallization prevents recrystallization of the high-purity niobium.

3.3 Texture after cold rolling and annealing
Figure 4 shows orientation maps and inverse pole figures of the high-purity niobium and the high-purity iron in cold rolling and heat treatment process. The dominant orientation of both cold rolled specimens was the \{100\} \text{<110> texture, the \{111\} <110> texture and the \{111\} <112> texture. It was found that the rolling texture of niobium and iron is consistent. Next, we focused on the annealed specimens. From the invese pole figures, it was found that the high-purity niobium has less orientation
variation and <100>//ND texture dominated compared to the high-purity iron. Furthermore, in the orientation image map of the high-purity niobium after annealing, elongated <100>//ND texture was observed. It was suggested that <100>//ND texture formed by cold rolling remained after annealing, which caused in-situ recrystallization of the high-purity niobium.

3.4 Evaluation of dislocation density
Dislocation energy, becomes driving force for recovery and recrystallization. Thus, it is presumed that the recrystallization temperature is influenced by the pile-up structures of dislocations. Dislocation densities of the cold rolled specimens were studied by the following two methods. Dislocation densities of the cold rolled specimens were calculated by the peak value obtained from XRD and the Williamson-Hall method[6]. It is known that the diffraction angle $\theta_B$ and the half-value width $\Delta 2\theta$ in XRD have the following relationship.

$$\Delta 2\theta \cdot \cos \theta_B / \lambda = ks / D + 2\varepsilon \cdot \sin \theta_B / \lambda$$

(1)

$\lambda$ is the wavelength of the X-ray, $ks$ is the Schuler constant, $D$ is the crystallite diameter, $\varepsilon$ is the lattice strain. $D$ and $\varepsilon$ can be obtained from the intercept and slope of the Williamson-Hall plot. Dislocation density is calculated using the following equation.

$$\rho = \sqrt{4.32\varepsilon / Db}$$

(2)

$b$ is the Burgers vector. Figure 5 (a) shows the calculated dislocation densities of the cold rolled specimens.

In addition, the dislocation densities were calculated using the Bailey-Hirsch equation[7] shown below.

$$\Delta \tau = \alpha \mu b \sqrt{\rho}$$

(3)

$\alpha$ is the constant (0.5), $\mu$ is the rigidity ratio (niobium: $\mu = 37$ GPa, iron: $\mu = 80$ GPa), $b$ is the Burgers vector size (niobium: $b = 0.29$ nm, iron: $b = 0.25$ nm), $\rho$ is the dislocation density. $\Delta \tau$ is calculated by the following equation.

$$\Delta \sigma = M \Delta \tau, \quad \Delta \sigma = 3 \Delta HV$$

(4)

$M$ is the Taylor factor (3.3), and $\Delta HV$ is calculated from the difference in Vickers hardness when changing the rolling rate from Figure 1. The calculated dislocation densities of the cold rolled specimens were shown in Figure 5 (b).

Figure 5 showed that in both of the high-purity niobium and the high-purity iron, the pile-up of dislocations increased as the rolling reduction increased. It was suggested that the lowering of the softening temperature accompanying the increase in the rolling reduction shown in FIG. 2 is due to the pile-up of dislocations. Moreover, it was also found that the dislocation density of the cold rolled high-purity niobium is higher than that of the high-purity iron. In other words, dislocations of the high-purity niobium tended to accumulate. This result showed that the difficulty of recrystallization of the high-purity niobium doesn’t result from dislocation density.

3.5 Factors impeding recrystallization
It was suggested that dislocation density of the high-purity niobium is higher than that of the high-purity iron. We speculated that this may be due to the low modulus of elasticity of niobium. Iron with high modulus of elasticity has dislocations with strong repulsive force and causes cross slip. Therefore, it is conceivable that dislocation density decreases due to annihilation of dislocations.
Recrystallization depends on the dislocation density as well as the dislocation energy. Since dislocation density is high in high purity niobium, it is presumed that dislocation energy is affected. The equation of dislocation energy is shown below.

\[ E = \frac{1}{2} \mu b^2 \]  

(5)

Since the elastic modulus of niobium is lower than that of iron, the dislocation energy of niobium is also lower than that of iron. It is suggested that the residual of rolled texture of niobium after annealing results from the difficulty of recrystallization due to low dislocation energy.

![Figure 4. Orientation image maps and invers pole figures of the high-purity niobium and the high-purity iron in cold rolling and heat treatment process.](image)

![Figure 5. Dislocation densities by (a) Williamson-Hall method and (b) Bailey-Hirsch equation of the specimens after cold rolling with reduction of 60% and 95%.](image)

4. Conclusion

As a result of investigating the texture of the high-purity niobium and the high-purity iron after cold rolling and annealing, the following was clarified. From the rolling hardening curves and softening curves, it was confirmed that high-purity niobium was harder to work hardening than high-purity iron. It was suggested that the cold rolled material of high purity niobium causes in-situ recrystallization due
to residual rolling texture composed of $<100>/\langle ND$ texture even after annealing. Comparing the dislocation densities of the high-purity niobium and the high-purity iron after cold rolling calculated by the Williamson-Hall method and Bailey-Hirsch equation, it was confirmed that the pure niobium has higher dislocation density than pure iron. It was suggested that the residual of rolled texture of niobium after annealing result from the difficulty of recrystallization due to low dislocation energy.

**Reference**

[1] Nagata T, Abe N, Masui H, Sshinozawa S, Nagakubo J, Murakami H, Inoue H, Yamanaka M and Kano E 2015 Studies on material properties regarding plastic deformation of niobium product for superconducting accelerator cavity –aiming at production of HOM coupler and other components by press forming technology *Proceedings of the 16th KEK Mechanical Engineering Workshop* 77

[2] Jiang H, Baars D, Zamiri A, Antonie C, Bauer P, Bieler T R and Grimm T L 2007 Mechanical properties of high RRR niobium with different texture *IEEE Transactions on Applied Superconductivity* 17(2) 1291

[3] Furubayashi E 2000 *Recrystallization and material texture* (Japan: Uchidarokakuho) p16, 99-119

[4] Miyamoto H, Xiao T, Uenoya T and Hatano M 2010 Effect of simple shear deformation prior to cold rolling on texture and ridging of 16% Cr ferritic stainless steel sheets *ISIJ International* 50(11) 1653-1659

[5] Fuiiwara H, Adachi M, Miyamoto H and Hatano M 2015 Effect of One-Pass ECAP prior to Cold Rolling on the Deformation Structure and Recrystallization in Ferritic Stainless Steel Sheets *Tetsu to Hagane-Journal of the Iron and Steel Institute of Japan* 101(11) 619-625

[6] Williamson G K and Smallman R E 1956 III Dislocation densities in some annealed and cold-worked metals from measurements on the X-ray debye-scherrer spectrum *Philosophical Magazine* 1(1) 34-46

[7] Bailey J E and Hirsch P B 1960 The dislocation distribution, flow stress, and stored energy in cold-worked polycrystalline silver *Philosophical Magazine* 5(53) 485-497

**Acknowledgement**

This work was financially supported by High Energy Accelerator Research Organization (KEK), ULVAC, Inc. and NEURON JAPAN CO.,LTD. and these supports are gratefully appreciated.