Estimation of Recrystallized Grain Size under Continuous Annealing of Cold-rolled β Titanium Alloy Strips

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We have investigated the behavior of the recrystallization and the grain growth of a cold-rolled Ti–15V–3Cr–3Sn–3Al and proposed a method for estimating the grain size after recrystallization during a continuous annealing process. The estimation of the grain size recrystallized under continuous annealing has been carried out by sequentially integrating, step by step, each increment of the isothermal grain growth at each temperature.

The observation of the microstructure reveals that the grain size recrystallized under isothermal annealing, $D(T, \tau)/\mu\text{m}$, can be indicated by $D(T, \tau) = 0.80 \times 10^4 \tau^{0.24} \exp(-1.50 \times 10^4/RT)$, where T and τ are the annealing temperature and time, and R is the gas constant. The estimate of the grain size recrystallized under a continuous heating process is in good agreement with the actual grain size. In addition, the grain size of the Ti alloy produced on the production line also agrees well with the calculated results using the temperature and passing time period of the continuous annealing line. This estimation provides good information for controlling the grain size after continuous annealing and leads to the production of finer-grained Ti–15V–3Cr–3Sn–3Al coils than have ever been obtained before.

KEY WORDS: beta titanium alloy; recrystallization; grain growth; grain size; continuous annealing.

1. Introduction

In recent years, beta titanium alloys have been important as industrial titanium alloys because their cold formability is at a level which can never be attained by alpha–beta titanium alloys.

As for sheet products of titanium alloys, cold strips of this type of titanium alloy are the most cost-effective in terms of both primary and secondary fabrication costs. In addition, recent research has also indicated that beta titanium alloys can be highly strengthened with sufficient toughness by appropriate thermo-mechanical processings.^{1,2)}

However, inappropriate heat treatments often lead to grain coarsening and result in problems such as orange peel after bending, and low ductility. In order to get good cold formability in beta titanium alloys, it is very important to control the grain size after annealing.

For small test pieces, it is relatively easy to control recrystallization behaviors during annealing using salt baths since the heating time may be negligible. On the other hand, on a real production scale, heat treatments are usually carried out in atmospheres. In this case, it is difficult to apply the data on the relationships among cold reduction, annealing temperature, and holding time which can be obtained from the results of such ideal heat treatments. For producing fine-grained and fully recrystallized beta titanium sheets, it is necessary to take

into account the effects of heating time on recrystallization and grain growth behaviors.

The purpose of this study was to develop a method for estimating recrystallized beta grain sizes of beta titanium cold strips annealed in an industrial continuous annealing line. In this paper, the results concerning one of the most popular beta titanium alloys, Ti–15V–3Cr–3Sn–3Al, are reported, including the results of a test production of a 1.0 mm thick, 950 mm wide, 2.5 metric ton coil.

2. Experimental Procedure

2.1. Materials

A 5.4 mm thick hot strip of Ti–15V–3Cr–3Sn–3Al was used for this study. The ingot was forged in the beta region and then hot rolled from 120 mm thickness at an initial temperature of 1273 K by a production mill. The chemical composition of the material is given in **Table 1**. The beta transus is calculated at 983 K on the basis of the study reported by Yolton *et al.*³⁾

A plate of 50 mm^w × 300 mm^l was cut from the hot strip and solution treated at a temperature of 1073 K for 1.8 ks in air followed by water quenching. After removing the oxidized layer by shot blustering, the plate was machined to a 5.0 mm thickness. The mean beta grain size measured by the linear intercept method was 80 μm .

Table 1. Chemical composition of the titanium alloy used.

Elements (wt%)	V	Cr	Sn	Al	Fe	O	N	H	Ti
Top	15.19	3.11	3.03	3.08	0.169	0.107	0.0053	<0.0010	bal.
Middle	15.27	3.05	3.03	3.06	0.165	0.112	0.0055	<0.0010	bal.
Bottom	15.16	3.04	3.04	3.04	0.190	0.111	0.0059	<0.0010	bal.

2.2. Recrystallization and Grain Growth during Isothermal Annealing

Pieces of various dimensions cut from the plate were cold rolled at reductions of 10 to 80% in the same direction as the hot rolling. Both rolled planes of the pieces were equally machined to a constant thickness of 1.0 mm and cut into smaller pieces of 15 mm square to anneal at temperatures of 993 to 1 273 K for 3 to 1 000 sec in salt baths. In these heat treatments, dead times were taken at 4 sec for the 993 and 1 023 K, 5 sec for the 1 073 and 1 123 K, 6 sec for the 1 173 K, and 7 sec for the 1 273 K samples, according to the results obtained by a thermocouple during treatments at 1 073 and 1 273 K.

After annealing, the materials were aged at a temperature of 723 K for 1.8 ks to decorate unrecrystallized regions.⁴⁾ Microstructures of the materials were then observed by an optical microscope in the section parallel to the rolling direction and normal to the rolled plane. The recrystallized grain sizes were measured by the linear intercept method.

It is hypothesized that recrystallization would be restrained by alpha precipitation. The 80% cold reduction materials were accordingly held at temperatures near the calculated beta transus for 1.8 ks in air to determine the temperature where optically perceptible recrystallization and grain growth occur.

2.3. Recrystallization Behavior during Heating

Based on the results obtained from the experiments mentioned above, a model was proposed for estimating the grain size change where the material temperature was a function of time. In order to evaluate its feasibility, the calculated values using the model were compared with the actual grain sizes of the materials which were 80% cold rolled and heated from room temperature to 1 073 K at heating rates of 0.3 to 10 K/sec in an infrared furnace, immediately followed by quenching into water.

Additionally, in order to obtain the relationship between the temperature of the sheet material and the time during annealing in atmospheres, temperature-time curves were recorded by inserting two beta titanium sheets of 110 mm^w × 1 000 mm^t × 1.0 mm^t and × 1.2 mm^t, with a thermocouple attached on a central surface, into air furnaces of 1 073 and 1 123 K, respectively, and pulling up the titanium sheets into air.

Finally, on the basis of a furnace temperature-passing period-grain size diagram obtained numerically, a cold strip of Ti-15V-3Cr-3Sn-3Al, which was 80% cold rolled after the solution treatment mentioned above, was continuously annealed in an annealing and pickling process.

Table 2. Measured grain sizes just after full recrystallization (in upper shelf; μm) and the time they were taken (in lower shelf; sec) in the cold rolled materials. The “—” indicates that the samples are not yet fully recrystallized after 1 000 sec.

Red./% Temp./K	10	30	50	65	80
1273	250 1000	112 100	73 30		19 3
1173	—	95 300	46 100		20 10
1123	—	—	53 1000	32 300	26 100
1073	—	—	—	36 1000	29 300
1023	—	—	—	—	26 1000
993	—	—	—	—	—

Fig. 1. Relationship between rolling reduction and grain size immediately after full recrystallization in the cold rolled materials.

3. Results and Discussion

3.1. Recrystallization Behavior

Table 2 shows the grain sizes right after full recrystallization, together with annealing times for the combinations of cold reduction and isothermal annealing temperature. The “—” means the sample has some unrecrystallized regions still after 1 000 sec. The recrystallized grain size decreases with an increase in

Fig. 2. Optical microstructures after annealing of the 80 % cold rolled materials. The annealing temperatures and times are: (a) 1 073 K, 3 sec (b) 1 073 K, 10 sec (c) 1 073 K, 100 sec (d) 993 K, 30 sec (e) 993 K, 100 sec (f) 993 K, 1 000 sec.

Fig. 3. Logarithmic relationships between annealing time and recrystallized grain size for various annealing temperatures in the 80 % cold rolled materials.

reduction, as clearly shown in Fig. 1, which was graphed from Table 2. The recrystallized grain size is largely dependent on reduction only and is almost independent of annealing temperature if the prior grain size is constant. The recrystallized grain size gradually becomes saturated as the reduction increases. A reduction of 80 % is considered to be sufficient to refine the microstructure. However, even if the material was cold rolled at a reduction of 80 %, some unrecrystallized regions were retained when the grain sizes in recrystallized regions were less than about 20 μm .

Figure 2 shows the structural changes during annealing at temperatures of 993 and 1073 K for the 80 % cold rolled materials. A large number of nucleations tend to occur along the deformed grain boundaries, and the new grains quickly impinge on each other. If they impinge on each other, they should give rise to normal grain

growth. Moreover, even though site saturation is occurring late in the recrystallization stage, such regions where nucleations did not occur are undistinguishable after full recrystallization.

It is qualitatively implied that the grain size in the recrystallized regions is almost the same in spite of different annealing temperatures, if the fraction of recrystallization is constant. This will be quantitatively shown later in Fig. 8. It may be said in this connection that there are very small unrecrystallized regions retained in Figs. 2(c) and 2(f).

Figure 3 shows logarithmic relationships between grain size and annealing time for the 80 % cold rolled material. As for the samples which were not fully recrystallized, the grain size in the recrystallized regions is plotted, and indicated on the graph by asterisks (*). In general, for a given annealing temperature a time dependence of the

grain size during primary recrystallization differs from that during normal grain growth. However, each relationship for a series of annealing temperatures has almost the same slope including the not fully recrystallized samples. This may be due to the immediate impingement and normal grain growth of the new grains as shown in Fig. 2.

Figure 4 shows semilogarithmic relationships between grain size and the inverse of annealing temperature, which are obtained by replotting the data on Fig. 3. Similarly, each relationship for a series of annealing times has almost the same slope.

On the basis of Figs. 3 and 4, recrystallized grain size D can be approximately formulated with annealing temperature T and annealing time τ as in the following equation,⁴⁾

$$D(T, \tau) = C\tau^n \exp(-Q/RT), \quad \dots\dots\dots(1)$$

where R is the gas constant, n is an exponential constant, and Q is activation energy. n and Q can be determined from the slopes in Figs. 3 and 4. Given n and Q , then, C can be calculated from any set of T , τ , and D .

In this study, however, n , Q (cal/mol) and C were determined by the method of least squares, leading to the equation,

$$D(T, \tau) = 0.80 \times 10^4 \tau^{0.24} \exp(-1.50 \times 10^4/RT)/\mu\text{m}. \quad \dots\dots\dots(2)$$

The theoretically expected value for n in a single phase is 0.5,⁵⁾ whereas the obtained value is 0.24. The reason for this disagreement is not clear. However, as for Beta III,⁶⁾ which is one of other beta titanium alloys, it was reported that n was between 0.34 and 0.44, depending on oxygen contents and annealing temperatures, possibly due to a drag effect caused by oxygen on the grain boundaries. Moreover, for Ti-15V-3Cr-3Sn-3Al,⁷⁾ n in the range of 0.05 to 0.14 was reported even though the data appears to be somewhat scattered.

For the purpose of this study, it is most important to obtain a simple and suitable relationship among D , τ , and T to predict grain sizes under various annealing conditions. From this point of view, a good agreement is obtained by Eq. (2), as graphically demonstrated in Fig. 5. The correlation coefficient is 0.974. Grain sizes during the period from primary recrystallization to normal grain growth may be well-interpolated by Eq. (2) in the range of temperatures below 1273 K.

Equation (2) was derived from the data for the material which had the initial grain size of 80 μm and was 80% cold rolled before annealing. On the other hand, this equation is also based on the normal grain growth including the grain growth which may be considered as

Fig. 4. Semilogarithmic relationships between the inverse of annealing temperature (T) and recrystallized grain size for various annealing times in the 80% cold rolled materials.

Fig. 5. Graphic correlation between the formulated annealing temperature-annealing time-grain size relationship and measured grain size.

Fig. 6. Optical microstructures after annealing for 1.8 ks at the temperatures of 983 K (a) and 988 K (b) in the 80% cold rolled materials.

Fig. 7. Optical microstructures after being heated from room temperature to 1073 K at the following rates until immediately quenched into water.

(a) 0.3 K/sec (b) 1 K/sec (c) 3 K/sec (d) 10 K/sec
 Recrystallized grain sizes measured are:
 (a) 24 μm (b) 18 μm (c) 14 μm (d) 9 μm

normal grain growth in the partial recrystallized regions. Equation (2) may therefore be independent of initial grain size and cold reduction to some degree, as long as the material is cold rolled sufficiently for many new grains to impinge immediately on each other and give rise to normal grain growth during recrystallization.

Figure 6 shows the microstructures of the materials which were 80% cold rolled and heat treated at temperatures of 983 and 988 K for 1.8 ks. In the case of 983 K (a), there are few optically perceptible new grains. On the other hand, with the 988 K sample (b), some new grains are unmistakably observable. The difference between Figs. 6(a) and 6(b) is likely to correspond to whether or not the annealing temperature is higher than the beta transus. If it is below the beta transus, it seems that recrystallization would compete with alpha phase precipitation, and so, would be restrained.

Compared with Figs. 2(d) to 2(f), Fig. 6(b) looks more irregular in terms of the localization of recrystallization. This is probably because of the faint fluctuation in compositions. However, the substantial beta transus seems to be between 983 and 988 K. As for grain growth, it may be acceptable to take into account only the behavior above the beta transus.

Figure 7 shows the microstructures of the 80% cold rolled materials after being heated from room temperature to 1073 K at the indicated heating rates and being immediately quenched into water. The grain sizes in the recrystallized regions are also indicated. From the viewpoint of the balance between the fraction of recrystallized region and grain size, the microstructures shown in Fig. 7 are quite similar to the isothermally annealed microstructures shown in Fig. 2.

Figure 8 shows a relationship between the fraction of recrystallization (the recrystallization ratio) and the grain size in the recrystallized regions. This graph was obtained

Fig. 8. Relationship between recrystallization ratio and grain size after isothermal and unisothermal annealings in the 80% cold rolled materials.

using most of the samples which had been 80% cold rolled and had not fully recrystallized. The marks with the annealing temperatures represent the isothermally annealed samples, and the marks with the heating rates, the samples shown in Fig. 7. It is clear that there is a specific relationship between the recrystallization ratio and the grain size in spite of various annealing conditions. This proves conclusively that some unrecrystallized regions are retained with the recrystallized grain sizes of less than about 20 μm .

3.2. Modeling for Estimating Grain Sizes after Annealing under Continuously Changing Temperatures

As mentioned in the Introduction, the relationship of

D , T , and τ indicated by Eq. (2) is not necessarily directly applicable to practical annealing conditions. This is because, for example, the exact annealing time cannot be defined for heat treatments conducted in atmosphere furnaces where the material temperature is gradually rising up to a specific temperature. In order to estimate appropriately the grain size in such cases, some kind of model is necessary.

Figure 2 showed that recrystallized grains immediately impinged on each other. Normal grain growth should therefore be occurring in the recrystallized regions even before full recrystallization. Moreover, site saturation does not seem to be so significant as to largely influence mean grain sizes after full recrystallization because the regions without nucleations become undistinguishable after that. In addition, Fig. 6 indicated that it may be acceptable to take into account only the behavior above the beta transus.

Therefore, recrystallized grain sizes on heating should be estimated simply by considering that recrystallized grains are formed just above the beta transus to immediately impinge and give rise to normal grain growth as indicated by Eq. (2). The following model was accordingly proposed in this study. The concept of the model is schematically shown in Fig. 9.

On the premise that the material temperature T is given as a function of time t , $T = T(t)$ can be treated stepwise by dividing the period from t_0 to t_f by some positive integer m . Here, t_0 is the time when the material temperature arrives at the beta transus, and t_f is any time when the grain size needs to be estimated. This leads to the equations:

$$T_i = T(t_i), \quad \dots\dots\dots(3)$$

$$t_i = \Delta t \cdot i + t_0, \quad \dots\dots\dots(4)$$

$$\Delta t = (t_f - t_0)/m, \quad \dots\dots\dots(5)$$

where t_i is the i^{th} time, and T_i is the material temperature

Fig. 9. Schematic display of the model for grain growth estimation when the material temperature is continuously changing.

corresponding to t_i . Considering the material temperature during the period, t_{i-1} to t_i , to be a constant temperature of T_i , the locus of grain size increment during t_{i-1} to t_i has to correspond locally with the curve indicated by the following equation,

$$D(T_i, \tau) = C\tau^n \exp(-Q/RT_i) \quad \dots\dots\dots(6)$$

Since the rate of normal grain growth is at least a function of grain size itself, and the increment of grain size is derived from integrating the growth rate with time, the starting point of that locus must be the point where the grain size is $D(t_{i-1})$. Here, $D(t_{i-1})$ is the grain size at $t = t_{i-1}$. Therefore, the effective time τ_i for τ in Eq. (6) is indicated by

$$\tau_i = \tau'_{i-1} + \Delta t, \quad \dots\dots\dots(7)$$

where τ'_{i-1} is satisfying the following equation,

$$D(T_i, \tau'_{i-1}) = D(t_{i-1}) \quad \dots\dots\dots(8)$$

In this context, the grain size at $t = t_i$, $D(t_i)$, is indicated by

$$D(t_i) = D(T_i, \tau_i) \quad \dots\dots\dots(9)$$

From Eqs. (6), (8), and (9) the following equation is derived,

$$C\tau'^n_{i-1} \exp(-Q/RT_i) = C\tau'^n_{i-1} \exp(-Q/RT_{i-1}) \quad \dots\dots\dots(10)$$

Then,

$$\tau'_{i-1} = \exp(Q/nR \cdot (1/T_i - 1/T_{i-1}))\tau_{i-1} \quad \dots\dots\dots(11)$$

Therefore, from Eqs. (7) and (11), the following relationship is obtained between τ_{i-1} and τ_i ,

$$\tau_i = \exp(Q/nR \cdot (1/T_i - 1/T_{i-1}))\tau_{i-1} + \Delta t \quad \dots\dots\dots(12)$$

τ_m is the effective time to give the grain size at $t = t_m$, that is, $t = t_f$. τ_m is obtained by repeating this operation from t_1 to t_f step by step. The boundary conditions are $\tau_0 = 0$ and $D(t_0) = 0$. With m being large enough to regard the temperature change as continuous, $D(t_f)$ is given by

Fig. 10. Graphic correlation between grain size estimated by the model and measured grain size when the 80% cold rolled materials are heated from room temperature to 1073 K at various rates.

Fig. 11. Temperature-time curves of the material during annealing in air furnaces. Furnace temperatures were 1 073 K (a) and 1 123 K (b). Some points calculated from Eq. (15) are also plotted.

$D(t_m)$, that is,

$$D(t_f) = D(T_f, \tau_m), \dots\dots\dots(13)$$

where T_f is the material temperature at $t = t_f$.

3.3. Verification of the Model

In the model mentioned above, the period from t_0 to t_f was divided by m in order to simplify the explanation of the concept of the model. However, the same values can also be calculated by integrating each grain size increment stepwise by a certain sufficiently short time division from t_0 up to t_f .

Figure 10 shows a graphic comparison between the measured grain sizes and the calculated ones for the samples whose microstructures are shown in Fig. 7. The time division is 0.01 sec. The demonstrated correlation is good enough that the model can be considered to be fairly applicable.

3.4. Application to a Test Production

In order to apply the model to practical production, it is advantageous to determine the material temperature-time relationship in the form of $T = T(t)$. Figure 11 shows the sheet material temperature-time curves when 1.0 and 1.2 mm thick sheets are put into air furnaces of 1 073 and 1 123 K, and, some minutes later, pulled up. Since, in its internal dimensions, the furnace was about 1 000 mm in diameter and 3 000 mm in length, changes in the furnace temperature with handling can probably be ignored. The sheets are so thin that the material

Fig. 12. Estimated furnace temperature-passing period-recrystallized grain size diagram.

temperature can be considered uniform in the direction of thickness.

According to the theory on heat conduction,⁸⁾ when an infinite plate, the initial temperature of which is T_a , is put into an atmosphere of T_b , at a given position the ideal relationship between the material temperature and time can be expressed by the following equation,

$$T(t) = (T_a - T_b) \exp(-kt) + T_b, \dots\dots\dots(14)$$

where k is a certain constant independent of the

Fig. 13. Optical microstructure of the Ti-15V-3Cr-3Sn-3Al coil annealed continuously in an annealing and pickling process.

temperatures. Thus, in this study, $T(t)$ was approximated by using k which was the most suitable value for representing the two curves in Fig. 11,

$$T(t) = (293 - T_b) \exp(-0.058t) + T_b, \dots\dots\dots (15)$$

where the initial temperature (T_a) was fixed at 293 K. In Fig. 11 some points calculated from Eq. (15) are also plotted. Good correlations were obtained for both curves.

On the other hand, the cooling rate is so rapid that grain growth on cooling is negligible.

Given a furnace temperature T_b and a period t during which the material is in the furnace (referred to as the passing period), its recrystallized grain size after passing can be calculated on the basis of the model. Figure 12 shows a furnace temperature-passing period-recrystallized grain size diagram which was numerically obtained with a time division of 1 sec. It was confirmed that under some conditions a time division of 0.01 sec led to almost the same results.

Finally, after considering other restrictions such as the capacity of the apparatus and the descalability of the sheet after annealing, the cold strip was continuously annealed under conditions almost corresponding to the combination of a furnace temperature of 1093 K and a passing period of 120 sec. This condition was chosen in order to obtain a fine-grained and fully recrystallized microstructure. As a result, a coil with a uniform fine-grained microstructure was successfully obtained.

That microstructure is shown in Fig. 13. The grain size is almost the same as estimated, that is, 21 μm .

4. Conclusion

In order to produce beta titanium coils with fully recrystallized but also fine-grained microstructure, a model for estimating the recrystallized grain size after continuous annealing was investigated using Ti-15V-3Cr-3Sn-3Al. The conclusions were as follows:

(1) Pronounced recrystallization and grain growth does not occur below the beta transus.

(2) In the case of annealing 80% cold rolled materials, a large number of nucleations occur, specifically along the deformed grain boundaries, and subsequently quickly impinge on each other.

(3) As for normal grain growth, the relationships among annealing temperature T , annealing time τ , and recrystallized grain size $D(T, \tau)$ can be described in the form of $D(T, \tau) = 0.80 \times 10^4 \tau^{0.24} \exp(-1.5 \times 10^4/RT)$.

(4) Estimation of grain size was successfully accomplished by integrating, stepwise, each increment as indicated by the above mentioned equations with each temperature and each effective time related to the last step above the beta transus.

(5) On the basis of the estimation of recrystallized grain size, a fine-grained Ti-15V-3Cr-3Sn-3Al coil was successfully obtained in an annealing and pickling process.

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