Hot Deformation Behaviors and Intrinsic Hot Workability Map of Ti-12Mo-4Zr-5Sn Alloy Based on Physical Model and Polar Reciprocity Model

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Received: 17 June 2020; Accepted: 14 July 2020; Published: 15 July 2020

Abstract: The hot compression tests of Ti-12Mo-4Zr-5Sn alloy were tested on the thermo-mechanical simulator of Gleeble-3500 under isothermal and constant strain rate. We studied this alloy’s behavior during thermal deformation at the conditions of $T = 670\text{--}820 \, {^\circ}\text{C}$, $\dot{\varepsilon} = 0.001\text{--}10 \, \text{s}^{-1}$, and deformation degree 70%. The rheological stress curves of the alloy were modified, the characteristics of the rheological stress curves were analyzed, and the activation energy map of hot deformation was established. A physical constitutive model of the alloy based on strain compensation was established, which has taken the relationship between Young’s modulus and self-diffusion coefficient and temperature into account. Moreover, the intrinsic hot workability $\xi$ map of the alloy was established based on the polar reciprocity model. The results show that this alloy’s rheological stress will descend when the temperature of deformation rises and grow when the strain rate increases, and has negative sensitivity of temperature and positive sensitivity of strain rate. According to the error calculation, the physical constitutive model’s correlation coefficient is 0.9910 and the average relative error is 3.97%, which has good accuracy. Through the analysis of the microstructures of the instability zone and the stability zone, it was found that the instability mode of the instability zone was dominated by the local flow, and the deformation mechanism of the stability zone was dominated by the dynamic recrystallization. The optimum processing parameters of the alloy known from $\xi$ map and metallographic structure are the following: $T = 790\text{--}820 \, {^\circ}\text{C}$ and $\dot{\varepsilon} = 0.001\text{--}0.01 \, \text{s}^{-1}$.

Keywords: Ti-12Mo-4Zr-5Sn alloy; physical constitutive model; polar reciprocity model; intrinsic hot workability $\xi$ map

1. Introduction

Titanium alloys are widely used as biomedical metal materials for artificial joints, fracture fixation, spine implantations, soft tissue reconstruction, and other situations because of their good biocompatibility, stable physical and chemical properties, and elastic modulus similar to human bone [1–4]. Many scholars have reported the hot deformation behaviors of titanium alloys. Gao et al. [5] studied the hot deformation flow behavior and microstructure evolution of TA15 titanium alloy by using isothermal compression test and electron backscatter diffraction technique. They found that the flow stress of the inhomogeneous structure showed an obvious flow softening at low strain after reaching the peak stress, which is similar to that of the layered structure. Mandal et al. [6] carried out the thermal compression experiments on
Ti-5Al-5Mo-5V-3Cr alloy at different temperatures and strain rates. The viscoplastic self-consistent model was used to simulate the mechanical response of the material. Ti-Mo-Zr-Sn alloys have good mechanical properties and belong to the third-generation biomedical titanium alloy. Among them, Ti-12Mo-4Zr-5Sn alloy has a strength greater than 1000 MPa and an elastic modulus of 60~80 GPa. It is a metastable, low modulus, and high-strength medical titanium alloy material, which has a good practical application. For Ti-12Mo-4Zr-5Sn alloy, thermal processing is the most important processing method before obtaining the product, and also an important way of improving the internal microstructure. Therefore, it is necessary to study the thermal processing of this alloy.

The constitutive relation model of metal material is the premise of the process design and control of metal plastic forming. It can express the influence of deformation conditions (temperature, strain rate, and strain) on the rheological behavior, and is the basis of the finite element numerical simulation technology and the formulation and optimization of thermal deformation process parameters. In recent years, there have been many reports on the application of the constitutive model to exploring the thermal deformation behavior of titanium alloy [7,8], but most of the constitutive models are based on Arrhenius constitutive equation, which belongs to the macroscopic phenomenological constitutive model. Those cannot effectively reveal the physical properties of materials. The physical-based constitutive model effectively integrates the macro-variables with the physical variables to predict the rheological stress and can also reflect the physical properties of materials. However, there are many equations involved in the complex physical constitutive model, it is difficult to identify the material parameters, and the amount of computation of parameters is large [9,10]. A physical constitutive model based on creep theory has good applicability. The model takes into account the influences of temperature on Young’s modulus and self-diffusion coefficient of materials and has achieved the prediction of the hot deformation behavior of materials by adopting the hyperbolic sine function. The plastic deformation of Ti-12Mo-4Zr-5Sn alloy at higher temperatures can not only prevent the cracking caused by deformation at high strain rate, but also obtain the required product shape. However, metal plastic deformation is more important to meet the requirements of microstructure and performance, so as to meet the actual production and application. Therefore, it is very important to select appropriate technological parameters to avoid various defects in the material forming process (such as instability, torsion, adiabatic shear, etc.). Murty et al. [11] discussed the relationship of intrinsic hot workability parameter between dynamic material model (DMM) and polar reciprocity model (PRM). However, most of the current researches use DMM to draw the processing map and get some appropriate process parameters, while there are relatively few reports about PRM. Compared with DMM, PRM not only considers the effect of strain rate on flow stress but also the effect of strain history on flow stress. It is suitable for optimizing the hot working parameters of the alloy. Although the effect of strain history on flow stress is not as significant as that of strain rate, it cannot be ignored. For example, the flow stress of material with insensitive strain rate is mainly affected by the strain history [12–14].

In this paper, the rheological properties of Ti-12Mo-4Zr-5Sn alloy at high temperatures were analyzed through hot compression experiments, and the rheological stress was modified by friction and temperature. Based on the modified data, the activation energies of hot deformation were calculated under different temperatures with different strains and strain rates, and the activation energy maps were constructed. A physical constitutive model based on strain compensation was constructed by introducing the strain into the physical constitutive equation. Finally, the intrinsic hot workability $\xi$ map of the alloy was established by using the PRM theory. The microstructure of the corresponding region of the intrinsic hot workability $\xi$ map was verified, and the processing parameters of the alloy were optimized. The results can provide theoretical guidance and data support for the actual hot forming of the alloy.
2. Experimental Procedures

The nominal composition of experimental material is Ti-12Mo-4Zr-5Sn. The chemical compositions (wt.%) of this alloy are: Mo 12%, Zr 4%, Sn 5%, and the rest Ti.

The hot compression of small cylindrical specimens with $\Phi 8 \text{ mm} \times 12 \text{ mm}$ obtained from raw ingot was tested on the thermo-mechanical simulator of Gleeble-3500 (Dynamic Systems Inc., Portland, OR, USA). To minimize the friction between contact surfaces, thin graphite sheets were attached as a lubricant material to both ends of the specimen. Samples were heated to 670, 700, 730, 760, 790 and 820 $^\circ\text{C}$ at a speed of $5 \degree\text{C}/s$, respectively, and held for 300 s. Thereafter, the compression tests were conducted at different strain rates (0.001, 0.01, 0.1, 1 and $10 \text{ s}^{-1}$) with a compression rate of 70%. When the reduction in the height reached 70%, to retain the deformation microstructure, the specimen should be quenched immediately. The flow diagram of hot compression deformation is shown in Figure 1, which shows four stages: heating, holding, compression, and quenching.

![Figure 1. Process scheme of thermal compression.](image)

Finally, specimens for the compression testing were sectioned parallel to the deformation axis and the cut sides were used for metallographic examination after polishing. Microstructure characterizations were observed through FL7500 optical microscope (Jinan Metallographic Instrument Equipment Co., Jinan, China) after etching by the solution of 6%HNO$_3$ + 4%HF + 90%H$_2$O (vol.%).

3. Results and Discussion

3.1. Discussion of Flow Behaviors

In thermal compression experiments, the existence of friction will cause an obvious bulging phenomenon of the compressed sample, and the accumulation of deformation heat effects will increase the experiment temperature, which is the main source of stress error. However, these effects cannot be eliminated by the existing thermomechanical simulator, it is necessary to modify the experimental data by friction and temperature. The revised data are closest to the real hot deformation behavior of materials.

The equation derived from the finite element method can be used to modify the rheological stress measured in the experiment by friction:

$$\frac{p_{\text{ave}}}{\sigma_f} = \frac{Rb}{H_i} \left[ \frac{1}{12} + \left( \frac{H_i}{Rb} \right)^{2/3} \right]^{3/2} - \left( \frac{H_i}{Rb} \right)^{3/2} - \frac{m}{24 \sqrt{3}} \left( 1 - e^{b/2} \right)$$

where $p_{\text{ave}}$ is the measured stress, MPa; $\sigma_f$ is the friction-corrected stress, MPa; $H_i$ is the height of the specimen at any time, mm; $R_i$ is the radius at any time, mm, it can be calculated by $R_i = R_0 \sqrt{H_{i0}/H_i}$.
$m$ and $b$ are the friction factor and bulging parameters, respectively, and these equations can be obtained by $\partial (p_{ave}/a_t)/\partial b = 0$:

$$m = \frac{Rb/H}{(4/\sqrt{3}) - (2b/3 \sqrt{3})}$$  \hspace{1cm} (2)$$

$$b = 4\frac{\Delta R}{R} \frac{H}{\Delta H}$$  \hspace{1cm} (3)$$

where $\Delta R = R_M - R_T$, mm; $R_T$ is the radius of the contact surface between the end face and the indenter after compression, mm, it can be calculated by $R_T = \sqrt{3}(H_0/H)R_0^2 - 2R_M^2$. $\Delta H$ is the height change of the specimen before and after compression, mm; $R$ and $H$ are the radius and height of the specimen after compression, mm.

The temperature rise caused by deformation heat effect can be expressed as Equation (4):

$$\Delta T = \frac{p}{C} \int_0^\varepsilon \sigma d\varepsilon$$  \hspace{1cm} (4)$$

where $\sigma$ is the measured stress, MPa; $\Delta T$ is the temperature rise caused by material deformation, °C; $p$ is the heat transfer coefficient related to strain rate; $C$ is the heat capacity coefficient of the material, N·mm$^{-2}$·°C$^{-1}$, and the heat capacity coefficient of titanium alloy is generally $C = 4$ [15].

Others are:

$$\begin{cases} p = 0 & \varepsilon < 0.001 \text{s}^{-1} \\ p = 1 + \varepsilon/3 & 0.001 \text{s}^{-1} \leq \varepsilon \leq 1 \text{s}^{-1} \\ p = 1 & \varepsilon > 1 \text{s}^{-1} \end{cases}$$  \hspace{1cm} (5)$$

The rheological stress change $\Delta \sigma$ caused by temperature rise $\Delta T$ can be determined by the Equation (6):

$$\Delta \sigma = \left( \frac{\partial \sigma}{\partial (1/T_n)} \right)_{\varepsilon, \dot{\varepsilon}} \left( \frac{1}{T_n + \Delta T} - \frac{1}{T_n} \right)$$  \hspace{1cm} (6)$$

where $T_n$ is the deformation temperature in the experiment, K.

According to Equations (1)–(6), the friction and temperature corrections were made for the rheological stresses of Ti-12Mo-4Zr-5Sn alloy measured at deformation temperatures of 670–820 °C and strain rates of 0.001–10 s$^{-1}$. The corrected rheological stress curves are shown in the dotted line of Figure 2. The effects of friction and temperature rise on the flow stress were interactive when the specimen was compressed. That is to say, the friction makes the experimental value higher than the true value, while the rise of temperature makes the experimental value lower than the true value.

According to the comparison of the rheological stress curves before and after friction and temperature correction in Figure 2, we found that the effects of friction and temperature rise on the flow stress become more and more obvious with the decrease in deformation temperature or the increase in strain rate. With the increase in strain, the trends between the revised curve and the measured curve present two situations: one is that the revised curve is below the original curve during the whole period, as shown in some curves in Figure 2a,b. The reason is that the true stress corrected by friction is less than the measured stress, and at a low strain rate, the main factor affecting the true stress is friction, while the effect of temperature rise has little effect on the true stress. Another case is that the revised curve lies below the original curve at first; with the accumulation of deformation, the dotted line will gradually approach the solid line first, and then deviate from the solid line gradually, shown in the curve at low temperature–high strain rate. This is because the effect of temperature rise on the true stress is less than that of friction when the degree of deformation is small. When the deformation accumulates to a certain level, the temperature gathered in the material will produce a temperature rise effect; the effect of temperature rise on the true stress will be higher than that of friction at this situation, and the corresponding correction curve will gradually deviate upward.
Figure 2 depicts a series of experimental rheological stress curves of Ti-12Mo-4Zr-5Sn alloy at different conditions, which have been revised through the friction and temperature corrections. It can be seen that the effects of strain rate and temperature of deformation on the rheological stress are more significant, the machining hardening phenomenon is very obvious at a high strain rate, and the steady state flow characteristics are presented at a low strain rate. When the temperature of deformation and strain rate are constant, as the strain increases, the curve has the characteristic of a typical dynamic recrystallization, that is, the rheological stress appears as a significant rheological softening after the peak, which is because the alloy has a low stacking fault energy and is prone to dynamic recrystallization under the influence of thermal deformation.
Figure 3 shows the rheological stress of the alloy at strain 0.4, which can be seen from the figure that when the temperature or strain rate is constant, the stress increases with the increase in strain rate and decreases with the increase in deformation temperature, indicating that the alloy has negative temperature sensitivity and positive strain rate sensitivity. This is because when the temperature is constant, with the increase in strain rate, larger strain will be produced in a short period of time, and a large number of dislocations will proliferate. At the same time, the rheological stress increases with the increase in strain rate because there is not enough time to produce softening effect. When the strain rate is constant, the increase in temperature intensifies the movement of atoms, which makes the dislocation movement easier, thus promoting the nucleation and growth of dynamic recrystallization. At the same time, because the critical shear stress value of slip is reduced, dislocation density and rheological stress are reduced.

3.2. Determination of Hot Deformation Activation Energy

The metal material’s high temperature deformation may include the following processes: the slip or climb of dislocations and the deformation caused by offset of opposite sign dislocations; the deformation caused by relative sliding between grain boundaries; the deformation caused by relative movement of interfaces; and the deformation caused by directional diffusion of interstitial atoms or vacancies. Each deformation process needs to overcome a certain energy barrier which is called the deformation activation energy of the process [16–18]. Taking the modified peak stress data of flow stress curve as an example, the hot deformation activation energy of Ti-12Mo-4Zr-5Sn alloy is calculated based on the Arrhenius hyperbolic sine function equation, as shown in Equation (7):

$$
\varepsilon = A \sinh (a \sigma)^n \exp (-Q/RT)
$$

where $\varepsilon$ is strain rate, s$^{-1}$; $\sigma$ is stress, MPa; $Q$ is the activation energy of hot deformation, J/mol; $R$ is gas constant, and $R = 8.314$ J/(mol·K); $A$, $n$, and $a$ are material constants; $T$ is temperature of deformation, K; the parameter $a$ can be determined by the constants $\beta$ and $n_1$ in Arrhenius exponential equation and power function equation [19,20], as shown in Equation (8):

$$
\alpha = \frac{\beta}{n_1} = \frac{\partial \ln \varepsilon / \partial \sigma}{\partial \ln \varepsilon / \partial \ln \sigma}
$$

The natural logarithms on both sides of Equation (7) can be written into the following two forms:

$$
\ln \varepsilon = n \ln [\sinh (a \sigma)] + (\ln A - Q/RT)
$$
\[
\ln[\sinh(\alpha \sigma)] = \frac{Q}{1000} \frac{1000}{nR} \frac{T}{T + \ln(\dot{\varepsilon}/A)}
\]  
(10)  

And the equation of activation energy of hot deformation \(Q\):  
\[
Q = 1000R \left( \frac{\partial \ln \dot{\varepsilon}}{\partial \ln[\sinh(\alpha \sigma)]} \right) \left( \frac{\partial \ln[\sinh(\alpha \sigma)]}{\partial (1000/T)} \right) \dot{\varepsilon} = 1000Rn\beta
\]  
(11)  

Among the equation:  
\[
n = \left( \frac{\partial \ln \dot{\varepsilon}}{\partial \ln[\sinh(\alpha \sigma)]} \right) \dot{\varepsilon}, \quad k = \left( \frac{\partial \ln[\sinh(\alpha \sigma)]}{\partial (1000/T)} \right) \dot{\varepsilon}.
\]

Figure 4a,b show the linear fitting of \(\ln \dot{\varepsilon} - \sigma\) and \(\ln \dot{\varepsilon} - \ln \sigma\), \(\beta\) and \(n_1\) under different conditions can be obtained, and the slope of each line can be obtained. Then, the corresponding \(\alpha\) can be obtained according to Equation (8). Therefore, the average value of \(\alpha\) is 0.005695.

According to Equations (9) and (10), the curves between \(\ln \dot{\varepsilon} - \ln[\sinh(\alpha \sigma)]\) and \(\ln[\sinh(\alpha \sigma)] - 1000/T\) can be established, as shown in Figure 5a,b. From the graphs, it can be seen that the slope of \(\ln \dot{\varepsilon} - \ln[\sinh(\alpha \sigma)]\) curve is \(n\), and that of \(\ln[\sinh(\alpha \sigma)] - 1000/T\) is \(k\). Through linear fitting, we can get \(k = 6.038624\), and \(n = 5.605\), respectively. The average hot deformation activation energy obtained by introducing the calculated \(n\) and \(k\) values into Equation (11) is \(Q = 281.40 \text{ kJ/mol}\).

In the experiment, the material deformation degree was 70%. Similarly, the material parameters \(\alpha\), \(k\), and \(n\), with the modified strain between 0.1–1.2 (the interval is 0.1), were also calculated. The deformation activation energy \(Q\) at different temperatures and different strains was obtained, as shown in Table 1. According to the calculation results, the deformation activation energy is
higher than the self-diffusion activation energies of pure α and pure β titanium alloys, 204 and 161 kJ/mol [21], respectively. It shows that the alloy’s hot deformations under these conditions may correspond to different deformation mechanisms, which are controlled by processes other than high-temperature diffusion.

Table 1. Deformation activation energies obtained at different temperatures and different strains.

| Q(kJ/mol) | 670  | 700  | 730  | 760  | 790  | 820  |
|-----------|------|------|------|------|------|------|
| Q0.1      | 413.51 | 335.48 | 307.44 | 302.07 | 282.36 | 292.03 |
| Q0.2      | 395.96 | 324.39 | 296.35 | 286.56 | 275.13 | 281.35 |
| Q0.3      | 379.24 | 319.94 | 291.40 | 276.27 | 269.60 | 273.70 |
| Q0.4      | 375.82 | 321.10 | 287.77 | 274.13 | 266.15 | 269.55 |
| Q0.5      | 379.24 | 324.90 | 288.42 | 274.26 | 266.18 | 269.35 |
| Q0.6      | 380.45 | 326.50 | 288.38 | 273.37 | 265.75 | 270.05 |
| Q0.7      | 383.68 | 326.99 | 290.42 | 273.89 | 265.84 | 269.11 |
| Q0.8      | 385.03 | 330.09 | 289.69 | 276.20 | 266.91 | 269.00 |
| Q0.9      | 386.00 | 332.84 | 290.38 | 279.97 | 269.85 | 271.44 |
| Q1.0      | 389.53 | 340.31 | 293.75 | 284.69 | 275.86 | 274.80 |
| Q1.1      | 399.33 | 337.93 | 295.59 | 287.64 | 277.82 | 277.68 |
| Q1.2      | 411.67 | 353.35 | 303.29 | 293.24 | 290.16 | 285.39 |

To further analyze the physical mechanism of hot deformation of the alloy, the activation energy $Q$ at different strain rates and different temperatures was calculated, as shown in Table 2. The activation energy maps of deformation were constructed according to Tables 1 and 2, respectively, as shown in Figure 6a,b.

Table 2. Deformation activation energies obtained at different strain rates and different temperatures.

| $Q$ (kJ/mol) | $i$ (s$^{-1}$) | 0.001  | 0.01  | 0.1   | 1     | 10    |
|--------------|---------------|--------|-------|-------|-------|-------|
| Q670         |               | 434.57 | 488.87| 483.58| 289.71| 257.75|
| Q700         |               | 370.24 | 416.50| 412.00| 246.83| 219.60|
| Q730         |               | 327.82 | 368.78| 364.79| 218.54| 194.44|
| Q760         |               | 314.74 | 354.07| 350.24| 209.82| 186.68|
| Q790         |               | 304.57 | 342.63| 338.92| 203.05| 180.65|
| Q820         |               | 307.32 | 345.72| 341.98| 204.88| 182.28|

Figure 6. Deformation activation energy maps of different temperatures and different strains (a) and different strain rates and different temperatures (b).

It can be seen from Figure 6 that the activation energy of deformation decreases with the increase in strain. It may be due to the increase in strain, the enhancement of slip or climb of dislocations, and the enhancement of dynamic softening effect in alloys, which decreases the activation energy.
required for material deformation [22]. The activation energy of the alloy at 670~730 °C is obviously higher than the self-diffusion activation energies of pure α and pure β titanium alloys. In this situation, the deformation may be mainly caused by dislocation and pinning mechanism, requiring larger energy to initiate dislocations. In the range of strain rates from 0.01 to 10 s⁻¹, the required activation energy of deformation decreases with the increase in strain rate, which may be that the defect densities of dislocations and vacancies among grains increase accordingly with the increases in strain rate and the driving force required for dynamic softening. Thus, the energy consumed decreases for extricating the attraction from atoms during partial recrystallization of grains, resulting in the decrease in recrystallization activation energy.

3.3. Establishment and Analysis of Constitutive Model Based on Physics

The physical constitutive model studied in this paper is based on the hyperbolic sine function equation, which is similar to the Arrhenius hyperbolic sine constitutive equation. They were first proposed in creep applications [23] and later in hot working [10,24–26]. The equations are as follows:

\[ D(T) = D_0 \exp \left( \frac{-Q_{sd}}{RT} \right) \]  \hspace{1cm} (12)

\[ E(T) = E_0 \left[ 1 + \frac{T_m}{G_0} \frac{dG}{dT} \left( T - 300 \right) \right] \]  \hspace{1cm} (13)

\[ \dot{\varepsilon} / D(T) = B' \left[ \sinh \left( \alpha' \sigma / E(T) \right) \right]^{n'} \]  \hspace{1cm} (14)

where \( D(T) \) and \( E(T) \) represent the functional relationship between self-diffusion coefficient and Young’s modulus and temperature, respectively. Among them, \( D_0 \) is diffusion constant; \( Q_{sd} \) is self-diffusion activation energy, J/mol; \( G_0 \) and \( E_0 \) represent shear modulus and Young’s modulus at 300 K, GPa; \( T_m \) is melting point, K; \( \alpha' \), \( B' \), and \( n' \) are the material parameter to be determined.

The primary problem of constructing the above constitutive model with a physical basis is used to solve the material parameters in the equations of \( D(T) \) and \( E(T) \). Based on the literature [23], the material parameters of Ti-12Mo-4Zr-5Sn alloy are shown in Table 3.

| Parameter | Value |
|-----------|-------|
| \( D_0 \) (m²/s) | 1.9 × 10⁻⁷ |
| \( Q_{sd} \) (J/mol) | 153,000 |
| \( G_0 \) (MPa) | 2.05 × 10⁴ |
| \( E_0 \) (MPa) | 1.1 × 10⁵ |
| \( T_m \) (K) | 1933 |
| \( T_m \) (K) | -0.5 |

By substituting the related material parameters in Table 3 into the Equations (12) and (13), the functions between the self-diffusion coefficient and Young’s modulus and temperature of the alloy can be obtained:

\[ D(T) = 1.9 \times 10^{-7} \exp \left( -\frac{153000}{RT} \right) \]  \hspace{1cm} (15)

\[ E(T) = 1.1 \times 10^{5} \left[ 1 - 0.5 \left( \frac{T - 300}{1933} \right) \right] \]  \hspace{1cm} (16)

Peak stress reflects the maximum load-bearing capacity of materials under experimental conditions. Therefore, taking the peak stress of the modified rheological stress curve as an example, the process of solving material constants \( \alpha' \), \( n' \), and \( B' \) in the physical constitutive equation is described. Because the physical constitutive equation is similar to the Arrhenius constitutive equation, the value of material constant \( \alpha' \) can be solved by power function and exponential equation with \( D(T) \) and \( E(T) \). That is to say, calculating \( n' \) in the power function equation and \( B' \) in the exponential equation are needed, and the value of \( \alpha' \) in \( \alpha' = \beta' / n_1' \) can be obtained. The corresponding equations are as follows:

\[ \dot{\varepsilon} / D(T) = B_1' \left( \sigma / E(T) \right)^{n_1'} \]  \hspace{1cm} (17)
\[
\dot{\varepsilon} / D(T) = B_2' \exp \left( \beta' \sigma / E(T) \right)
\] (18)

Taking natural logarithms on both sides of Equations (17) and (18), and taking derivatives after that, it can be concluded that:

\[
n'_1 = \partial \ln [\dot{\varepsilon} / D(T)] / \partial \ln [\sigma / E(T)]
\] (19)

\[
\beta' = \partial \ln [\dot{\varepsilon} / D(T)] / \partial [\sigma / E(T)]
\] (20)

Therefore, \(n'_1\) can be determined by the slope of the fitted linear curve of the curve between \(\ln [\dot{\varepsilon} / D(T)]\) and \(\ln [\sigma / E(T)]\), and \(\beta'\) can be determined by the slope of the fitted linear curve of the curve between \(\ln [\dot{\varepsilon} / D(T)]\) and \(\sigma / E(T)\). The values of \(n'_1\) and \(\beta'\) can be obtained as 6.874 and 3425.698, as shown in Figure 7a,b. From this, \(\alpha' = 498.356\).

\[
\ln [\dot{\varepsilon} / D(T)] = n' \ln [\sinh (\alpha' \sigma / E(T))] + \ln B'
\] (21)

Taking the natural logarithm on both sides of Equation (14), the Equation (21) is obtained:

\[
\ln [\dot{\varepsilon} / D(T)] = n' \ln [\sinh (\alpha' \sigma / E(T))] + \ln B'
\] (21)

According to Equation (21), The values of \(n'\) and \(B'\) can be determined by the slope and intercept obtained by linear fitting of the curve between \(\ln [\dot{\varepsilon} / D(T)]\) and \(\ln [\sinh (\alpha' \sigma / E(T))]\). As shown in Figure 8, the results are \(n' = 5.740\) and \(\ln B' = 29.493\).

The physical constitutive equation of the alloy with peak stress data as substitution parameters can be obtained by combining the above-mentioned material constants \(n', \alpha', \) and \(\ln B'\) and Equations (14)–(16). The equation is shown in Equation (22):

\[
\dot{\varepsilon} / D(T) = 6.436 \times 10^{12} \sinh[498.356 \sigma / E(T)]^{5.740}
\] (22)
To sum up, the material constants in the physical constitutive model can be calculated by the steps in the flow chart, as shown in Figure 9.

![Flow chart for solving material constants in constitutive model with physical basis.](image)

Figure 9. Flow chart for solving material constants in constitutive model with physical basis.

However, the physical constitutive model mentioned above only considers the peak stress but does not consider the effect of strain on the flow stress. Therefore, the strain factor was introduced into the physical-based constitutive model constructed in this paper for optimization. The rheological stress values under the strain conditions of 0.1~1.2 (the interval is 0.1, 12 strain variables in total) were selected to compute the material constants $n'$, $\alpha'$, and $\ln B'$ under different strains according to the above method. The results are shown in Table 4. The results were fitted by linear regression with a six-degree polynomial, and the functions between material constants and strain can be obtained, which can be used to construct the physical constitutive model with strain compensation.

Table 4. Material constants of constitutive models with physical basis under different strains.

| $\varepsilon$ | $\alpha'$ | $n'$ | $\ln B'$ |
|---------------|-----------|------|----------|
| 0.1           | 556.460   | 4.441| 29.636   |
| 0.2           | 549.260   | 4.434| 29.587   |
| 0.3           | 547.525   | 4.453| 29.564   |
| 0.4           | 549.490   | 4.476| 29.533   |
| 0.5           | 552.220   | 4.518| 29.520   |
| 0.6           | 554.813   | 4.568| 29.501   |
| 0.7           | 553.386   | 4.646| 29.500   |
| 0.8           | 550.291   | 4.726| 29.502   |
| 0.9           | 545.913   | 4.835| 29.512   |
| 1             | 538.003   | 4.987| 29.533   |
| 1.1           | 528.799   | 5.193| 29.550   |
| 1.2           | 521.824   | 5.414| 29.568   |

Figure 10 shows the fitting results of six-degree polynomial between material constants $n'$, $\alpha'$, and $\ln B'$ and strain. It was found that the graphs that the linear correlation coefficient $R$ of the fitting results are all above 0.99, which indicates that the data points in the graphs can be described by the functions obtained by the fitting of the six-degree polynomial. Medina et al. [27] discussed the
general expression of the Zener–Hollomon parameter as a function of the chemical composition of low alloy and microalloyed steels through torsion tests. An expression between activation energy $Q$ and the content of each alloying element is given and the equation is completed by determining the optimum values of $\alpha$ and $n$ for all the steels. They demonstrated that $A$ is a function of the activation energy. Hernandez et al. [28] developed a model to predict the austenite flow curves of low alloy and microalloyed steels. The parameters in model are a function of the dimensionless parameter $Z/A$ of the equation of Sellars and Tegart, which has been modeled not only as a function of temperature and strain rate but also as a function of the chemical composition. The previous two methods are mainly based on the traditional Arrhenius-type hyperbolic sine function $\varepsilon \exp (Q/RT) = A\sinh (\alpha \sigma)^n$. Compared with them, the method used in this paper has the biggest difference that the influence of strain variables on material parameters in the physical constitutive model is considered. The results showed that these parameters ($\alpha$, $n$, activation energy $Q$, and $A$) are material constants and are not dependent on deformation. The functions determined by Figure 10 are shown in Equation (23):

$$\begin{align*}
\alpha'(\varepsilon) &= 574.753 - 253.466\varepsilon + 815.467\varepsilon^2 - 1099.942\varepsilon^3 + 756.636\varepsilon^4 - 345.853\varepsilon^5 \\
n'(\varepsilon) &= 4.422 + 0.470\varepsilon - 4.247\varepsilon^2 + 15.744\varepsilon^3 - 24.460\varepsilon^4 + 17.971\varepsilon^5 - 4.909\varepsilon^6 \\
\ln B'(\varepsilon) &= 29.727 - 1.277\varepsilon + 4.572\varepsilon^2 - 10.459\varepsilon^3 + 12.896\varepsilon^4 - 7.677\varepsilon^5 + 1.750\varepsilon^6
\end{align*}$$

(23)

Figure 10. The fitting result of six-degree polynomial between material constants $\alpha'$, $n'$, and $\ln B'$ and strain. (a) $\alpha'$-$\varepsilon$; (b) $n'$-$\varepsilon$; (c) $\ln B'$-$\varepsilon$.

The relationship between material constants $n'$, $\alpha'$, and $\ln B'$ and strain in Equation (23) is embedded into Equation (14), and the constitutive model with physical basis of the alloy optimized.
under compression deformation conditions can be obtained by combining Equations (15) and (16). This equation is shown in Equation (24):

\[ \sigma = \frac{E(T)}{\alpha'(\dot{\varepsilon})} \ln \left\{ \left( \frac{\dot{\varepsilon}}{D(T)B'(\dot{\varepsilon})} \right)^{1/n'(\dot{\varepsilon})} + \left( \frac{\dot{\varepsilon}}{D(T)B'(\dot{\varepsilon})} \right)^{2/n'(\dot{\varepsilon})} + 1 \right\}^{1/2} \]  

To verify the accuracy of the physical-based constitutive model for predicting flow stress, the predicted and experimental values are compared, and the results are shown in Figure 11a,b. It can be seen that the rheological stress predicted by the constitutive model with physical basis basically coincides with the experimental value and has certain reliability. Besides, this model’s prediction accuracy was further evaluated by correlation coefficient (R) and average absolute relative error (AARE). The equations are as follows:

\[ R = \frac{\sum_{i=1}^{n} (C_i - \bar{C})(F_i - \bar{F})}{\sqrt{\sum_{i=1}^{n} (C_i - \bar{C})^2} \sqrt{\sum_{i=1}^{n} (F_i - \bar{F})^2}} \]  

\[ AARE = \frac{1}{n} \sum_{i=1}^{n} \left| \frac{C_i - F_i}{C_i} \right| \times 100\% \]  

where \( C_i \) is the rheological stress’s experimental data; \( F_i \) is the rheological stress’s prediction data; \( \bar{C} \) is the experimental data’s average values; \( \bar{F} \) is the prediction data’s average values; \( n \) is the sample’s total number.

Figure 11. Comparison of predicted and experimental values of constitutive models with physical basis. (a) \( T = 760 °C \); (b) \( \dot{\varepsilon} = 1 \) s\(^{-1}\).

All the prediction data points and the corresponding experimental values were contrasted, as shown in Figure 12. We found that AARE between prediction stress and the experimental stress is 3.97%, \( R \) is 0.9910, and the data points of the constitutive model of which predicted value deviation is less than 10% account for 97.73%. Moreover, it can be obtained from Figure 12b that values of absolute error between 0 and 15 MPa account for approximately 92.42% of the total data. This indicates that the physical-based constitutive equation established by strain compensation is relatively good in accuracy and can accurately predict the high-temperature rheological stress of this alloy.

The solving process of material parameters based on the physical constitutive model is not only simple and effective but also has a certain physical basis. It can be used as a new prediction model for the hot deformation rheological stress.
3.4. Intrinsic Hot Workability Map Based on Polar Reciprocity Model

3.4.1. Construction and Analysis of Intrinsic Hot Workability $\xi$ Map

The polar reciprocity model (PRM) is based on Hill correlation plastic flow rule. Based on the PRM, the intrinsic hot workability map can accurately reflect the evolution mechanism of the microstructure under different deformation conditions after the observation and verification of the microstructure. Thus, the safety zone and instability zone of the material in the thermal processing process are obtained, and the technological parameters (temperature, strain rate, strain variable, etc.) in the thermal processing process are optimized. Finally, the goal of controlling the evolution of microstructure and avoiding defects is achieved, and the microstructure and performance are obtained. In the polar reciprocity model, the total instantaneous power is divided into two parts, and then the intrinsic hot working parameters are defined. Figure 13 shows a convex potential surface $\Phi = K$ in stress space with origin $O_1$. At point $P$ on this surface, the radius vector $O_1P$ with arbitrary diameter and normal $PN$ of $\Phi$ are made. The magnitude of normal $PN$ is equal to $KK'/\Phi$ and $KK'$ is the dissipation power. A vector $O_1P'$ that is parallel to the normal $PN$ and equals to its size is made through $O_1$, a point $P$ on $\Phi$ can have a corresponding point $P'$ on $\Phi'$. The trajectory of point $P'$ defines a potential surface $\Phi'$ in strain rate space. $K$ and $K'$ are the scalar functions of temperature and deformation history, which are affected by strain and deformation history.

Figure 13. Pole–pole relationships between two first-order non-linear potential functions.
Under the given temperature and strain history conditions, Equation (27) can be obtained by the one-to-one correspondence between the strain rate space and the stress space:

\[
\int \sigma \, d\dot{\epsilon}^P + \int \dot{\epsilon}^P \, d\sigma = \sigma \dot{\epsilon}^P = KK'
\]  

(27)

Two integral expressions on the left side of the equation represent the hardening power $W_H$ and the dissipative power $W_D$, respectively. In the earlier dynamic material model (DMM), the total dissipative power is also divided into two parts, but the theories used in the two models to divide the total dissipative power are different. Thus, the intrinsic hot workability parameter is proposed. The equation is shown in Equation (28):

\[
\xi = \frac{W_H}{W_{H_{\text{min}}}} - 1
\]  

(28)

In the PRM, the change process of $W_H$ and $W_D$ caused by the influence of strain history can be normalized by $W_{H_{\text{min}}}$, which can reflect the polar reciprocity. Based on this, Raj et al. [12] proposed a constitutive equation shown in Equation (29):

\[
\sigma = H(\epsilon^P) + C F(\dot{\epsilon}^P)
\]  

(29)

where $\sigma$ denotes stress, MPa; $\epsilon$ is strain; $\dot{\epsilon}$ is strain rate, s$^{-1}$; $H(\epsilon^P)$ and $C$ are two functions related to strain history. The equation of $H(\epsilon^P)$ is shown in Equation (30), which records the strain history from the beginning of plastic flow to strain $\epsilon_1$. $\sigma_{\text{min}}$ is the minimum flow stress from the beginning of plastic flow to strain $\epsilon = \epsilon_1$. When the material presents ideal plastic flow, $H(\epsilon^P) = 0$.

\[
H(\epsilon^P) = \frac{\int_{\epsilon_0}^{\epsilon_1} \sigma \, d\epsilon - \int_{\epsilon_0}^{\epsilon_1} \sigma_{\text{min}} \, d\epsilon}{\int_{\epsilon_0}^{\epsilon_1} \sigma \, d\epsilon}
\]  

(30)

When there is a polar reciprocity between potential functions $\Phi$ and $\Phi'$, the Equation (31) can be obtained:

\[
F(\dot{\epsilon}^P) = \left(\dot{\epsilon}^P\right)^{m'}
\]  

(31)

where $m'$ is a modified strain rate sensitivity coefficient. Equation (32) can be obtained from Equations (28)–(31):

\[
\xi = 1 - \left[\frac{2m'}{m' + 1} \left|\frac{\sigma - H(\epsilon^P)}{\sigma}\right|\right]
\]  

(32)

Raj and Kutumbarao [12] concluded the criterion of the instability of the polar reciprocity model by studying the hot working behavior of metals and basing on Hill correlation plastic flow rule: the material will lose its stability when $\xi$ approaches 1.

To reduce the experimental error, Matlab was used to interpolate the flow stress data by the cubic spline function. The data can be substituted into Equation (30) to calculate the strain function value $H(\epsilon^P)$, Equation (31) to obtain the modified strain rate sensitivity coefficient $m'$ under different deformation conditions, and Equation (32) to calculate the value of $\xi$. The intrinsic hot workability $\xi$ maps with true strains of 0.3, 0.6, 0.9 and 1.2 are drawn on the two-dimensional plane, as shown in Figure 14.
Figure 14. Intrinsic hot workability $\xi$ map under different true strains during thermal compression deformation. (a) $\epsilon = 0.3$; (b) $\epsilon = 0.6$; (c) $\epsilon = 0.9$; (d) $\epsilon = 1.2$.

According to the PRM criterion mentioned above, the region where $\xi$ approaches 1 is the unstable region. To avoid structural instability, it is necessary to consider the instability of the material under different strains to ensure that the material has qualified properties after processing and forming. Figure 14 shows that the instability region of $\xi$ map varies under different strains. The area of the rheological instability region at $T = 670$~$820 \degree C$ and $\dot{\varepsilon} = 0.316$~$10 \text{ s}^{-1}$ increases with the increase in strain and gradually moves toward the direction of low strain rate. In the deformation process, the processing window of the rheological instability zone of Ti-12Mo-4Zr-5Sn alloy can be characterized by the following parameters: $T = 670$~$820 \degree C$ and $\dot{\varepsilon} = 0.316$~$10 \text{ s}^{-1}$. In the region, the corresponding instability parameters of the material are relatively high, and instability phenomena such as local plastic flow or adiabatic shear band are prone to occur.

Among the four types of true strain $\xi$ maps, there is mainly one $\xi$ minimum region. The region is characterized by the following parameters: $T = 790$~$820 \degree C$ and $\dot{\varepsilon} = 0.001$~$0.01 \text{ s}^{-1}$. When the true strains are 0.3, 0.6, 0.9, and 1.2, the minimum values are 0.42, 0.44, 0.44, and 0.47, respectively, which is less affected by the strain. The basin area in $\xi$ map corresponds to favorable deformation mechanisms such as dynamic recovery, dynamic recrystallization, and superplasticity; while the peak area corresponds to disadvantageous deformation mechanisms such as fracture, local flow, and adiabatic shear; and the grain growth leads to the increase in $\xi$ value [13]. When determining the parameters of the hot working process, the range of parameters of favorable deformation should be given priority, and whether the range of parameters of the hot working process in a stable region should be considered. The ranges of favorable deformation mechanism in Figure 14 are the following: $T = 790$~$820 \degree C$ and $\dot{\varepsilon} = 0.001$~$0.01 \text{ s}^{-1}$, which are the suitable processing parameters.
3.4.2. Verification of Microstructure

The validity of the PRM $\xi$ map is verified by observing the microstructure. Figure 15 shows the microstructures of the deformation instability zone of the alloy. Figure 15a corresponds to deformation parameters: $T = 670 \, ^\circ C$ and $\dot{\varepsilon} = 1 \, s^{-1}$; Figure 15b corresponds to deformation parameters: $T = 760 \, ^\circ C$ and $\dot{\varepsilon} = 10 \, s^{-1}$; Figure 15c corresponds to deformation parameters: $T = 820 \, ^\circ C$ and $\dot{\varepsilon} = 10 \, s^{-1}$. Figure 15a–c all have local plastic flow phenomena, which is due to the low thermal conductivity of the alloy and the uneven deformation of different parts of the sample, resulting in obvious local temperature rise. That means that, during plastic deformation, the alloy produces a lot of heat, which cannot diffuse to the surrounding area, resulting in local rheological instability. Local plastic flow generally occurs at a high strain rate, forming a zigzag-shaped micro-band with an angle of 35~40° from the direction of the principal stress. Local plastic flow occurs in the deformed structure, and the uniformity of deformation and structure will be poor, which will affect the properties of the material after forming.

![Microstructure of deformation instability zone](image1.png)

Figure 15. Microstructure of deformation instability zone. (a) $T = 670 \, ^\circ C$, $\dot{\varepsilon} = 1 \, s^{-1}$; (b) $T = 760 \, ^\circ C$, $\dot{\varepsilon} = 10 \, s^{-1}$; (c) $T = 820 \, ^\circ C$, $\dot{\varepsilon} = 10 \, s^{-1}$.

Figure 16 shows the microstructures of the deformation stable zone of the alloy. Figure 16a corresponds to deformation parameters: $T = 790 \, ^\circ C$ and $\dot{\varepsilon} = 0.001 \, s^{-1}$; Figure 16b corresponds to deformation parameters: $T = 820 \, ^\circ C$ and $\dot{\varepsilon} = 0.001 \, s^{-1}$. Figure 16a’s corresponding $\xi$ value is 0.65, and its corresponding intrinsic hot workability $\xi$ map is located near the basin of the PRM $\xi$ map. It can be seen from Figure 16a that the dynamic recrystallization is more sufficient, the structure is uniform, the $\beta$ grain is smaller, the size difference between grains is small, and the deformation is stable. Figure 16b’s corresponding $\xi$ value is 0.62, which is close to the minimum of $\xi$. The dynamic recrystallization under this deformation parameter is more sufficient and the structure is finer. The volume fraction of
recrystallization increases. The average grain size is about 20 µm, and the structure is in a rheological stable state.

Figure 16. Microstructure of deformation stability zone. (a) \( T = 790 \, ^\circ\text{C}, \dot{\varepsilon} = 0.001 \, \text{s}^{-1} \); (b) \( T = 820 \, ^\circ\text{C}, \dot{\varepsilon} = 0.001 \, \text{s}^{-1} \).

It can also be seen from Figure 16 that the temperature has an important effect on the dynamic recrystallization of the alloy at a strain rate of 0.001 s\(^{-1}\). Dynamic recrystallization is generally a continuous process in which the phase difference increases gradually through the accumulation of dislocations, and the small-angle grain boundary transforms gradually to the large-angle grain boundary, thus producing new grains [29]. In the process of deformation, the grain boundaries are serrated due to the local intergranular migration. At a low deformation rate, the small-angle grain boundaries have enough time to transform to the large-angle grain boundaries, and dynamic recrystallization easily occurs. Deformation temperature also has an effect on grain boundary migration, so when the temperature increases, atomic diffusion speed increases, grain boundary mobility increases, and dynamic recrystallization is more likely to occur.

Through the prediction of the intrinsic hot workability \( \xi \) map and the observation of the microstructure, the validation was carried out. The optimum processing parameters of the alloy are the following: \( T = 790 - 820 \, ^\circ\text{C} \) and \( \dot{\varepsilon} = 0.001 - 0.01 \, \text{s}^{-1} \). In addition, dynamic recrystallization is the main deformation mechanism in the deformation stabilization region. Fine recrystallization is conducive to the subsequent processing.

4. Conclusions

When the temperature or strain rate is constant, the peak stress will rise when the strain rate grows and descend when the deformation temperature rises; Ti-12Mo-4Zr-5Sn alloy has negative sensitivity of temperature and positive sensitivity of strain rate.

(1) The hot deformation activation energy of Ti-12Mo-4Zr-5Sn alloy is higher than that of pure \( \alpha \) and pure \( \beta \) titanium alloys. The results show that the hot deformation of the alloy is controlled by a process other than high-temperature diffusion and may correspond to different deformation mechanisms.

(2) A physical-based constitutive model considered the effect of deformation temperature on the self-diffusion coefficient and Young’s modulus of the alloy was established, and the model was optimized. Strain compensation was also considered in the optimized model. After error calculation, this model’s correlation coefficient is 0.9910, the average relative error is 3.97%, and the data points within the deviation of 10% accounting for 97.73%, respectively, which have good accuracy. The model is not only simple and effective but also has a certain physical basis. It can be used as a new prediction model for the hot deformation rheological stress.
Based on the polar reciprocity model, the intrinsic hot workability $\xi$ maps of Ti-12Mo-4Zr-5Sn alloy were drawn. The range of rheological instability zone of the alloy was obtained by observing the microstructures: $T = 670$–$820 \degree C$ and $\dot{\varepsilon} = 0.316$–$10 s^{-1}$. The optimum processing parameters of the alloy are as follows: $T = 790$–$820 \degree C$ and $\dot{\varepsilon} = 0.001$–$0.01 s^{-1}$. Local flow is the main instability form in the instability region, while dynamic recrystallization is the main deformation mechanism in the stability region.

Author Contributions: Conceptualization, K.W. and Z.Z.; Data curation, Z.Z., S.L. and H.N.; Formal analysis, H.Z.; Software, P.W.; Writing—original draft, P.W.; Writing—review and editing, K.W. All authors have read and agreed to the published version of the manuscript.

Funding: The National Natural Science Foundation of China (Grant No. 51464035) sponsored this study.

Conflicts of Interest: The authors declare no conflict of interest.

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