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# Effect of hydrogen content on superplastic forming/diffusion bonding of TC21 alloys

Xiaoli Wang[a,b], Yongqing Zhao[b,*], Hongliang Hou[c], Yaoqi Wang[c]

[a] School of Material Science and Engineering, Northwestern Polytechnical University, Shaanxi 710072, PR China
[b] Northwest Institute for Nonferrous Metal Research, Shaanxi 710016, PR China
[c] Beijing Aeronautical Manufacturing Technology Research Institute, Beijing 100024, PR China

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## ABSTRACT

The superplastic forming and diffusion bonding (SPF/DB) of hydrogenated TC21 alloys was carried out in the temperature range of 1073–1193 K under 1.5 MPa gas pressure. The effects of hydrogen contents on bonding ratio of SPF/DB and microstructure of interface and substrate for TC21 alloys were investigated by OM, SEM and TG–DTA. The experimental results showed that bonding ratio of SPF/DB increased with the increase of hydrogen contents. However, it decreased at hydrogenated 0.5 wt%. With the same hydrogen content, bonding ratio increased with the increase of temperature. The size and amounts of voids distributed along the interface decreased with the increase of hydrogen contents, and the recrystallization of joint grains through the interface was formed.

## 1. Introduction

Superplastic forming/diffusion bonding (SPF/DB) is a well-established process for the manufacture of components almost exclusively from titanium alloys sheet materials. It refers to a process in which a joint is made at temperatures below the melting point of the metals being joined, without any solder used, and under a pressing load sufficient to bring about the necessary plastic deformation of the pieces. Since diffusion bonding is formed from atomic migration across an interface of the solid state, there is no metallurgical discontinuity at the interface. Therefore, mechanical properties and microstructure at the bonded region are not different from those of the base metal. The superplastic forming and diffusion bonding processes have interesting applications for aerospace components. The complicated structures could be obtained during this process, such as hollow [1,2], with lower cost and weight. Therefore, the process has been applied to form sandwich construction of titanium alloys and aluminum alloys in aerospace [3–5].

As a candidate material for the structural parts of advanced aircraft, TC21 alloy is a new $\alpha+\beta$ titanium alloy with a relatively high or moderate strength, higher fracture toughness and lower crack propagation rate, which can satisfy the increasing demand of fracture mechanics and damage tolerance. In the past few years, the mechanical properties and phase transformation of TC21 alloys were investigated [6–8]. However, a combination of superplastic forming and diffusion bonding of TC21 alloys has no report. Some work has reported that hydrogen could lower the diffusion bonding temperature because the transformation temperature decreased [9,10]. This work aims to study the effect of hydrogen contents on bonding ratio and microstructure transformation of the interface.

In the present work, diffusion bonding of hydrogenated TC21 alloys has been investigated by adopting a new approach to overcome the oxidation of the surface and the release of hydrogen. It is shown that the quality of interface with hydrogen is better than that without hydrogen.

## 2. Experimental procedures

The material used in the present work was the sheet of TC21 titanium alloys with the normal composition of Ti–6Al–2Zr–2Sn–3Mo–1Cr–1Nb. The hydrogenated TC21 alloys were obtained by holding them at 1023 K in pure hydrogen atmosphere for 1 h and then air cooling to room temperature. Hydrogen contents were calculated by the weight gain after hydrogenation, which were 0.1 wt%, 0.3 wt% and 0.5 wt%. The specimens with the dimension of 30 mm × 15 mm × 2 mm were mechanically polished to remove the surface oxide layer and ultrasonically cleaned with acetone in order to maintain the specimens' surface finish. Assembled specimens were sealed with stainless sheet and maintained at vacuum. The process of SPF/DB was carried out in the temperature range of 1073–∼1193 K. 1.5 MPa gas pressure was applied for 70 min. The heating rate of the diffusion bonding was 5 °C/min.

The cross-sections of the joints were prepared by usual grinding and polishing techniques. The bonding ratio was computed by measuring the total length of SPF/DB interface and voids. The microstructure of hydrogenated TC21 alloys (before and after SPF/DB) and cross-sections of joints were observed by optical microscope (OLYMPUS-BX41M) and scanning electron microscopy (SEM: JSM-6460, VE-9800).

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* Corresponding author. Tel.: +86 29 86266577; fax: +86 29 86231103.
*E-mail address:* trc@c-nin.com (Y. Zhao).

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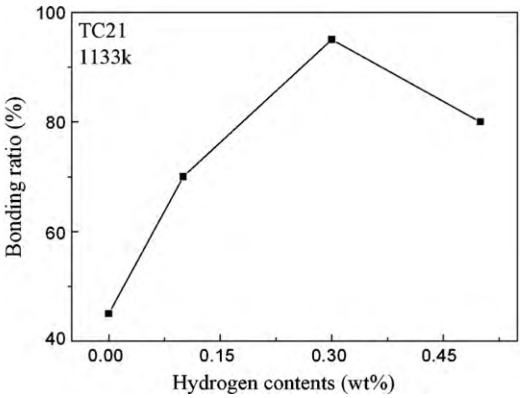

**Fig. 1.** Effect of hydrogen content on welding-on ratio of TC21 alloys (1133 K, 1.5 MPa, 70 min).

The dehydrogenation was carried out by using simultaneous thermal analysis and thermogravimetry (DTA–TG; NETZSCH-TA4), and experiments were performed against α-alumina as a reference at heating rates of 20 K/min. The apparatus was vacuumed by pump at the beginning, and then samples with the weight of 455 ± 0.5 mg were heated up to 1200 °C under flowing high purity argon atmosphere kept at a rate of 100 ml/min.

## 3. Results

### 3.1. Effect of hydrogen contents on bonding ratio of SPF/DB

The effect of hydrogen on superplastic forming and diffusion bonding of TC21 alloys has been investigated. The bonding ratio ($\eta$) can be calculated by the following equation:

$$\eta = \frac{(L_0 - L)}{L_0} \tag{1}$$

Where $L_0$ is the length of the inspected bonding seam; $L$ is the length of unclosed bonding defects in the inspected region. The inspection method is metallographic analysis. The distribution of voids was non-uniform, so optical micrograph in entire zone was used to calculate the bonding ratio.

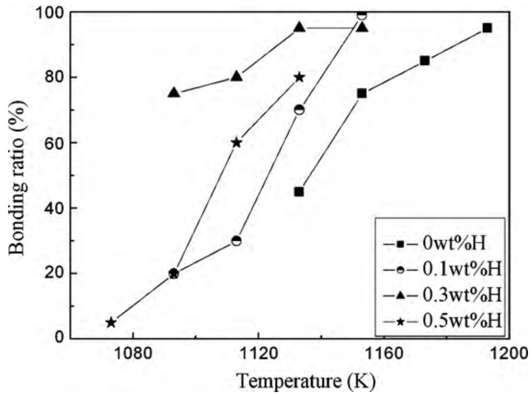

**Fig. 2.** Welding-on ratio of hydrogenated TC21 alloys at different temperature (1.5 MPa, 70 min).

Fig. 1 represents the effect of hydrogen contents on bonding ratio of TC21 alloys at 1133 K for 70 min under a pressure of 1.5 MPa. The results show that bonding ratio of TC21 alloys with hydrogen is higher than that of TC21 alloys without hydrogen, which is less than 50%. However, bonding ratio of hydrogenated TC21 alloys containing 0.5 wt% hydrogen is lower than that of those containing 0.3 wt% hydrogen. By observing the welding interface of hydrogenated TC21 alloys containing 0.5 wt% hydrogen, slight oxidation is observed, which results in the Bonding ratio decrease.

### 3.2. Effect of temperature on bonding ratio of SPF/DB

Effect of temperature on bonding ratio of SPF/DB for hydrogenated TC21 alloys has been shown in Fig. 2. The results show that bonding ratio of SPF/DB increases with the increase of temperature. It is related to plastic deformation of TC21 alloys. At low temperature, the micro-plastic deformation of interface is inadequate and atomic interaction is solely observed at individ-

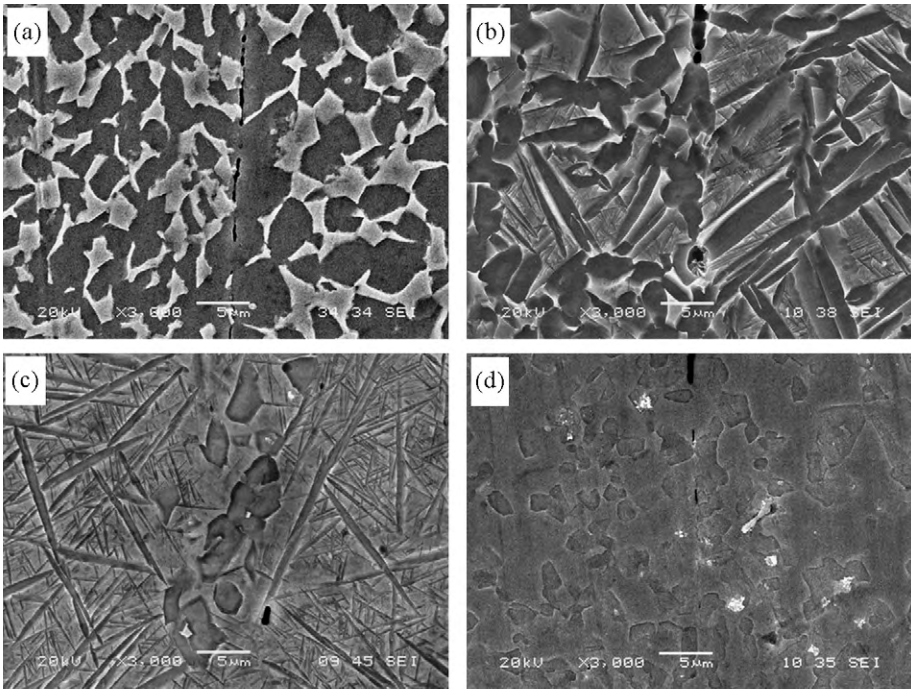

**Fig. 3.** The SPF/DB interface microstructure of TC21 alloys containing different hydrogen contents: (a) 0 wt%; (b) 0.1 wt%; (c) 0.3 wt%; (d) 0.5 wt%.

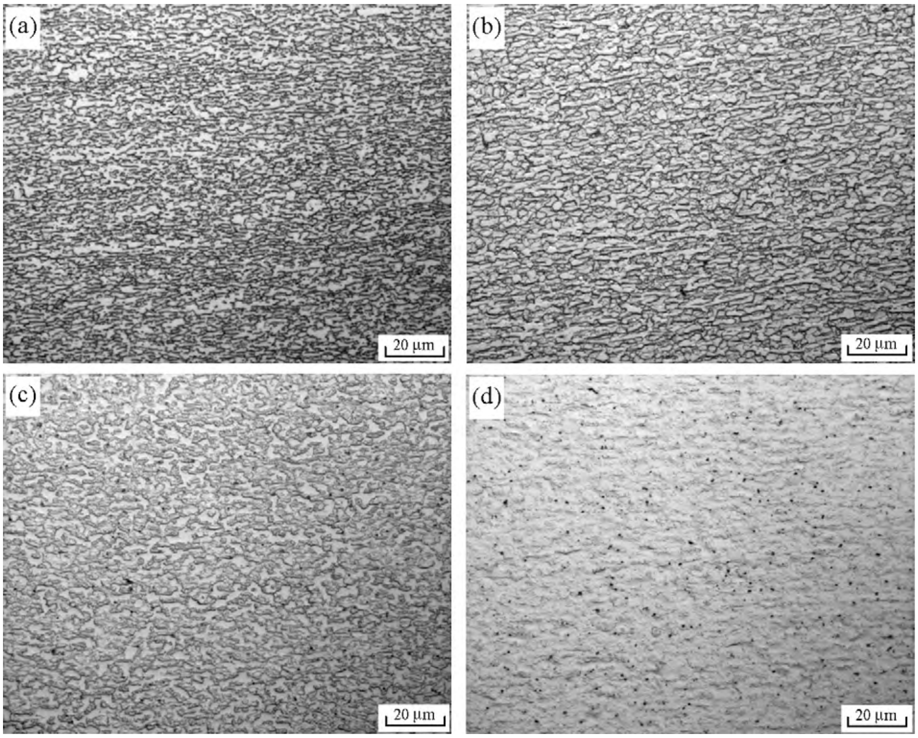

**Fig. 4.** The microstructure of TC21 alloys containing different hydrogen contents: (a) 0 wt%H; (b) 0.1 wt%H; (c) 0.3 wt%H; (d) 0.5 wt%H.

ual asperities. As the temperature of the material is raised, yield strength of titanium alloys decreases sharply and strong heat would enhance the activity and mobility of the species in the crystal lattice. When the diffusion pressure and heat are applied, they produce in the weld zone a concentration of energy which is sufficient to bring about a rearrangement of the surface layers in the mating specimens and also to give rise to the slower secondary processes, including interdiffusion, recrystallization and so on.

### *3.3. Effect of hydrogen contents on the interface and substrate microstructure of SPF/DB*

The SPF/DB interface microstructure of TC21 alloys containing different hydrogen contents has been shown in Fig. 3. Without hydrogen, narrow and long voids are distributed along contacting plane. The voids decrease gradually with the increase of hydrogen content and nearly disappear at the hydrogen content up to 0.3 wt%. Therefore, the existence

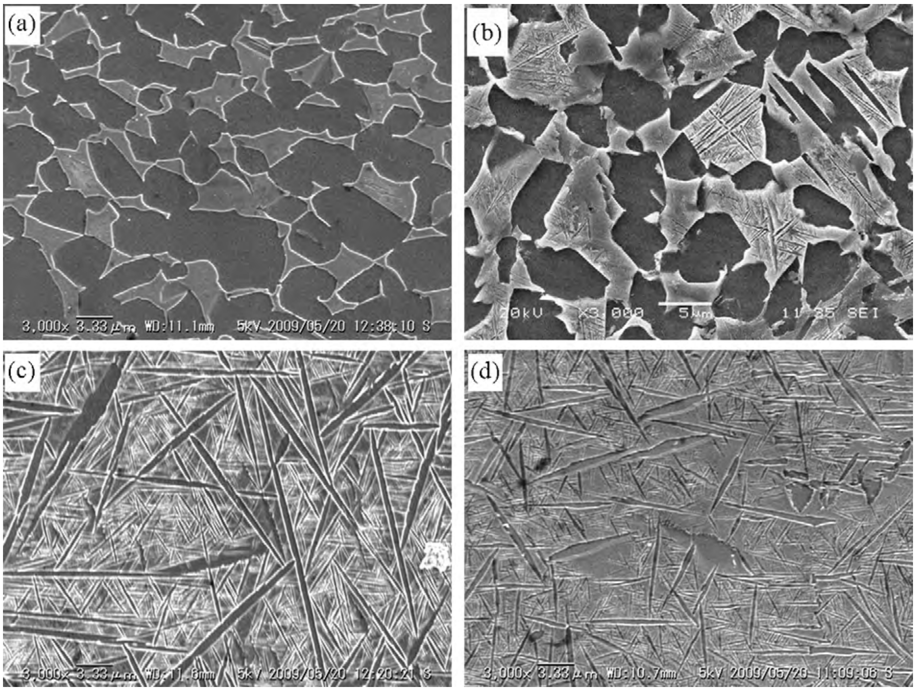

**Fig. 5.** The substrate of SPF/DB for TC21 alloys containing different hydrogen contents: (a) 0 wt%H; (b) 0.1 wt%H; (c) 0.3 wt%H; (d) 0.5 wt%H.

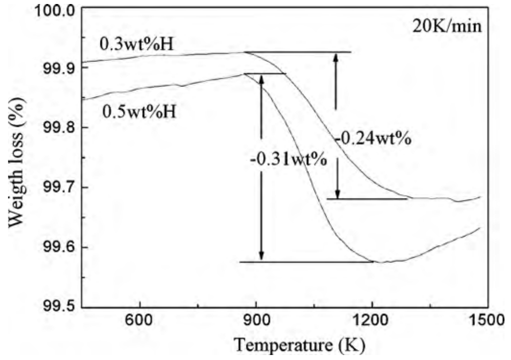

**Fig. 6.** TG curves of hydrogenated TC21 alloys.

of hydrogen accelerates the closure of voids at the interface.

The microstructure of TC21 alloys containing different hydrogen contents has been shown in Fig. 4. It can be seen that TC21 alloy before hydrogenation consists of α (white phase) and β phase (black phase), as shown in Fig. 4(a). After hydrogenation, the BCC $\beta_H$ hydrogen-rich phases gradually increase with the increase of hydrogen contents because hydrogen is the element of stabilizing β phase (Fig. 4(b)–(d)). After hydrogenation, the color of α and β phase has some change. The reason is that the electrochemical bit of α and β phase changes with the role of hydrogen.

After superplastic forming and diffusion bonding, microstructure of TC21 alloys with different hydrogen contents has been shown in Fig. 5. Substrate of TC21 alloys without hydrogen is also equiaxed (α + β) dial phase structure (Fig. 5(a)), but that of TC21 alloys with hydrogen has changed. Fig. 5(b) shows that β phase becomes coarsely, and some fine needle-like secondary α phase and thin-lamellar secondary α phase are distributed on lamellar transformed β phase. The substrate of TC21 alloys with 0.3 wt%H is fine needle α′ matersite (Fig. 5(c)), which is also found in the substrate of TC21 alloys with 0.5 wt%H (Fig. 5(d)). However, the size of needle α′ matersite of TC21 alloys with 0.5 wt%H is smaller than that of those with 0.3 wt%H.

### 3.4. The TG curves of dehydrogenation

The TG curves of TC21 alloys containing different hydrogen contents are shown in Fig. 6. As seen from Fig. 6, the TG curves of hydrogenated TC21 alloys after SPF/DB rapidly descend above 873 K because of the release of hydrogen, and then they ascend slightly due to the oxidation of samples. Hydrogen contents of TC21 alloys with 0.3 wt% and 0.5 wt% reduce to 0.06 wt% and 0.19 wt%, respectively.

TG results show that this method (using stainless sheets to seal the samples) inhibits the release of hydrogen and causes more phase transformation, which improve the quality of diffusion bonding joints.

## 4. Discussions

Effects of hydrogen contents and temperature on the quality of joints have been investigated. The results are similar to that obtained by Liu et al. [10] and Feng et al. [9]. The existence of hydrogen can improve the quality of diffusion bonding joints. Based on the definition of superplastic forming and diffusion bonding, superplastic deformation, creep and diffusion of alloying elements can explain the effect of hydrogen. Firstly, Hydrogen alloying destabilizes low-temperature hcp α phase and stabilizes more ductile high-temperature bcc β phase in α + β titanium alloy, which is beneficial of the high-temperature deformation. Secondly, the temperature of α to β phase transformation is decreased and phase transformation proceeds at relatively low temperature. Thirdly, the addition of hydrogen increases the ductility and reduces the flow stress of α + β titanium alloy [11–16], which can decrease the temperature of superplastic deformation. Finally, hydrogen affects the diffusivity of alloying elements as well as the dislocation slip systems [17]. By activating the material surface, it promotes better diffusion bonding.

In addition, the heating rate of the diffusion bonding was 5 °C/min. According to the dehydrogenation experiment, the initial dehydrogenation temperature of TC21 alloy with heating rate 5 °C/min was 590 °C [18]. Therefore, the dehydrogenation process which is dynamic equilibrium occurred during the heating process. The release absorption of hydrogen repeatedly can accelerate the decomposition of oxide film in contact zone.

However, it has been found that flow stress decreases firstly, reaches a minimum, and then increases with the increase of hydrogen concentration at a particular processing temperature. It can explain that the bonding ratio of TC21 alloys with 0.5 wt%H is lower than that of those with 0.3 wt%H.

## 5. Conclusions

(1) The bonding ratio increases with the increase of hydrogen contents. However, when hydrogen content reaches 0.5 wt%, it decreases.
(2) The voids of interface decrease with the increase of hydrogen contents and common grains are formed on interface.
(3) After superplastic forming and diffusion bonding, substrate of TC21 alloys without hydrogen is also equiaxed (α + β) dial phase structure, but that of TC21 alloys with hydrogen has changed. Some fine needle-like secondary α phase and thin-lamellar secondary α phase are distributed on lamellar transformed β phase with 0.1 wt%H. Fine needle α′ matersite is formed on TC21 alloys with 0.3 wt%H and 0.5 wt%H.

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