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Effects of oxygen contamination in the argon shielding gas in laser welding of commercially pure titanium thin sheet

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This work studied the effects of oxygen contamination in the argon shielding gases on weld microstructures and properties during laser welding of commercially pure titanium thin sheets. The experimental results, mainly analyzed by optical and scanning electron microscopy and mechanical testing, have indicated correlations between weld surface colour, weld microstructure and mechanical properties (strength, ductility, hardness). As the oxygen content increased, the weld surface colour changed from silver, straw to blue while the surface hardness continued to increase. On the other hand, with the increasing of oxygen content, the weld strength increased first and then decreased because the microstructure changed from mainly serrated alpha in welds made with pure argon shielding gas to mainly acicular and platelet alpha. Practical guidelines are also discussed, based on the study, to deal with shielding deficiencies in laser welding of titanium.

1. Introduction

Joining of titanium (Ti), either commercially pure or alloyed, is often required in aerospace and chemical industries, and especially in the medical field. Ti is preferred for manufacturing of surgical implants and prosthetic devices because it provides excellent combinations of corrosion resistance, mechanical properties and biocompatibility [1–3]. A number of processes, such as gas tungsten arc welding, laser welding, and brazing [4, 5], have been used successfully to join Ti. But titanium is extremely reactive, especially in the molten state, and the strength and hardness of Ti welds are increased while the ductility is reduced through picking up interstitial elements, such as oxygen, nitrogen, and hydrogen, if shielding is inadequate during welding [6–8].

A number of investigations have evaluated the effects of air contaminations (e.g., oxygen and/or nitrogen) in shielding gases on the weld microstructure and mechanical properties in gas tungsten arc welding (GTAW) of commercially pure (CP) Ti and Ti alloys [5, 9, 10]. Most studies concluded that the colour of the weld surface could be used as an indication of the degree of air contamination in the shielding gases and thus in the titanium welds. For example, Maak [10], in an investigation on GTAW of CP grade-2 (CP-2) Ti with both the face and root of the weld sufficiently shielded by a specially designed jig, concluded that weld surface discoloration correlated to the shielding gas deficiency, the oxygen and nitrogen content in welds, and weld metal mechanical properties. However, this correlation between weld surface colour and weld contamination depends also on other variables (e.g., the design of shielding devices, weld metal cooling rate). For example, Harwig *et al.* [9] found that weld colour indicated only the surface contamination that occurred during solid-state cooling at high temperature, in an investigation in which only contaminated shielding gas was used in the GTAW torch but pure argon was always used in back and trail shielding. A silver weld could be produced if a contaminated weld pool was well shielded after it solidified since the surface oxide can dissolve into the substrate [9]. On the other hand, slower cooling rates would promote darker weld colour since the weld metal is at a higher oxidation temperature as it leaves the protection of the shielding.

Laser welding is an alternative to other processes for joining Ti and has a number of advantages [4, 7], such as,

- Compared to other fusion welding processes, the highly focused laser beam energy increases welding speed and hence productivity, and also requires very low heat input, which reduces the possibility of damaging surrounding thermal-sensitive components; and

- No filler metal is required, as in brazing and soldering; therefore, biocompatibility and corrosion resistance of the base metal can be retained.

However, no systematic investigation has been published on effects of shielding gas deficiency/ impurity in laser welding of Ti, although air contamination remains an issue as in other welding processes [7]. The present work investigated the effects of oxygen contamination in the argon shielding gas on the microstructure and properties of laser welds of CP-2 Ti thin sheets. The thickness of 0.5 mm of the sheets is much thinner than the 2–3 mm material used by Harwig *et al.* [9].

2. Experimental procedure

The material used in this study was CP-2 Ti sheet of 0.5 mm thickness in 50 × 10 mm coupons. Full-penetration seam welds were made using a Lumonics JK702H pulsed Nd:YAG laser with a 600 μm fiber optic delivery system as bead on plate in the flat position. The laser system had a 200 mm focal length collimator lens and a 120 mm final focus lens. The intensity distribution of the laser beam at the focal point was found to be well approximated by a Gaussian distribution with a $1/e^2$ beam diameter or "spot size", at 413 μm [11]. To avoid back-reflection, the laser head was aligned so that the incident laser beam was at 15 degrees to the normal of the specimen surface. Ultra high purity argon (99.999%) and argon-oxygen mixtures, in which oxygen varied from 0 to 10 vol.%, were used as shielding gases in welding on both the top and bottom surfaces of the weld. Throughout the experiments, the laser was focused on the top surface of the material with following parameters: 4.5 ms pulse width, 5 Hz pulse frequency, 0.4 mm/s welding speed, 22–23 W average power and 30 cfh (cubic feet per hour) flow rate of the shielding gas. Weld bead surfaces were visually inspected and the discoloration was recorded.

Vickers hardness tests were carried out on both the surface and cross sections of the welds using a load of 300 g. Each data point was an average of six nominally identical hardness impressions. It has been suggested [9, 10] that transverse tensile testing does not provide an accurate assessment of the weld strength and ductility because the different strengths of the weld and base metals often lead to non-uniform straining in the gauge length of the test coupon. However, longitudinal all-weld-metal tensile testing was not practical in this work since the laser welds were very small. Instead, a specially designed transverse specimen was used (Fig. 1) to concentrate the strain in the weld metal. Tensile tests were performed using an INSTRON 4465 tensile tester with a cross-head speed of 1.0 mm/min.

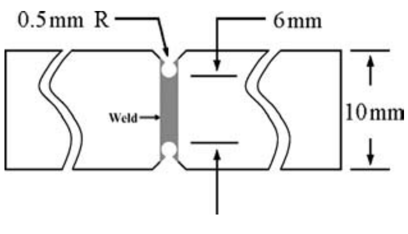

Figure 1 Design of tensile specimen.

TABLE I Surface colour and hardness of welds made with varied oxygen content in argon shielding gas

Oxygen content (%)	Surface colour	Hardness (Hv)
0	Silver	242 ± 11
0.15	Straw	246 ± 16
0.5	Dark Straw	247 ± 13
1.5	Dark Straw/Purple	254 ± 20
2.0	Dark Straw/ Purple/Blue	282 ± 18
3.0	Purple/Blue	295 ± 16
5.0	Blue	323 ± 15
10.0	Blue	373 ± 20

The welded Ti coupons were sectioned, ground, polished and etched for examination under the optical microscope. The etching solution was Kroll's reagent: 2 ml HF, 6 ml HNO_3 and 92 ml H_2O. The fractured surfaces of tensile tested samples were examined using a scanning electron microscope.

3. Results and discussion

3.1. Weld surface colour and hardness

Table I lists the changes in weld surface colour and hardness with increase of oxygen content in the argon shielding gas. The surface colour changed from silver on the welds with pure argon shielding gas, which is similar to the original colour of the base metal, to straw/dark straw as oxygen was introduced into the shielding gas, to purple and even blue as oxygen content in the shielding gas was greatly increased. This observation, which was similar to most findings for GTA welds, indicated that the degree of discoloration of the weld surface could be related to the oxygen contamination in the shielding gas, since the colour change was a direct result of the change in the thickness of oxide film [10]. Because of the very low heat input, the cooling rate in laser welding is extremely fast [12]. This prevents extreme oxidation after freezing of the weld because the temperature of the weld and heat affected zone drops very fast. However, it should be cautioned to use the discoloration as an inspection tool of the shielding deficiency because the color sequence (from straw, purple to blue) repeats itself as the oxidation thickness increases [13].

For the GTA welds made with contaminated shielding gas, the microhardness of welds increased from 165 to 225 Hv as oxygen equivalent of weld metal increased from 0 to 0.29 wt% [9]. The hardness of welds was found to depend on the content of oxygen in the weld metal as well as on other factors. The 3 mm thick welds had a microhardness that was approximately 5 Hv lower than the 2 mm thick welds as a function of oxygen equivalent, apparently due to the lower cooling rate. However, the equivalent formula was not found to be applicable to the data of laser welding. In that previous work [9], the maximum hardness was only 225 Hv for the GTA welds, even lower than the value of 242 Hv obtained under high purity Argon shielding gas in laser welding. The effect of cooling rate on the microhardness of these welds is significant, and the final hardness result depends on the interaction of cooling rate with the composition including O and N contents.

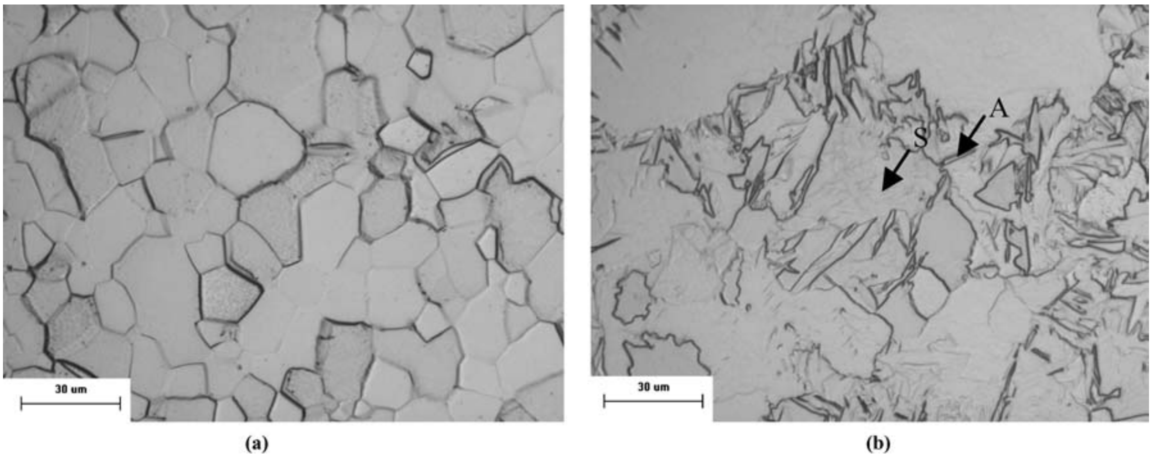

Figure 2 Cross sections of (a) base metal and (b) weld made with pure argon shielding gas.

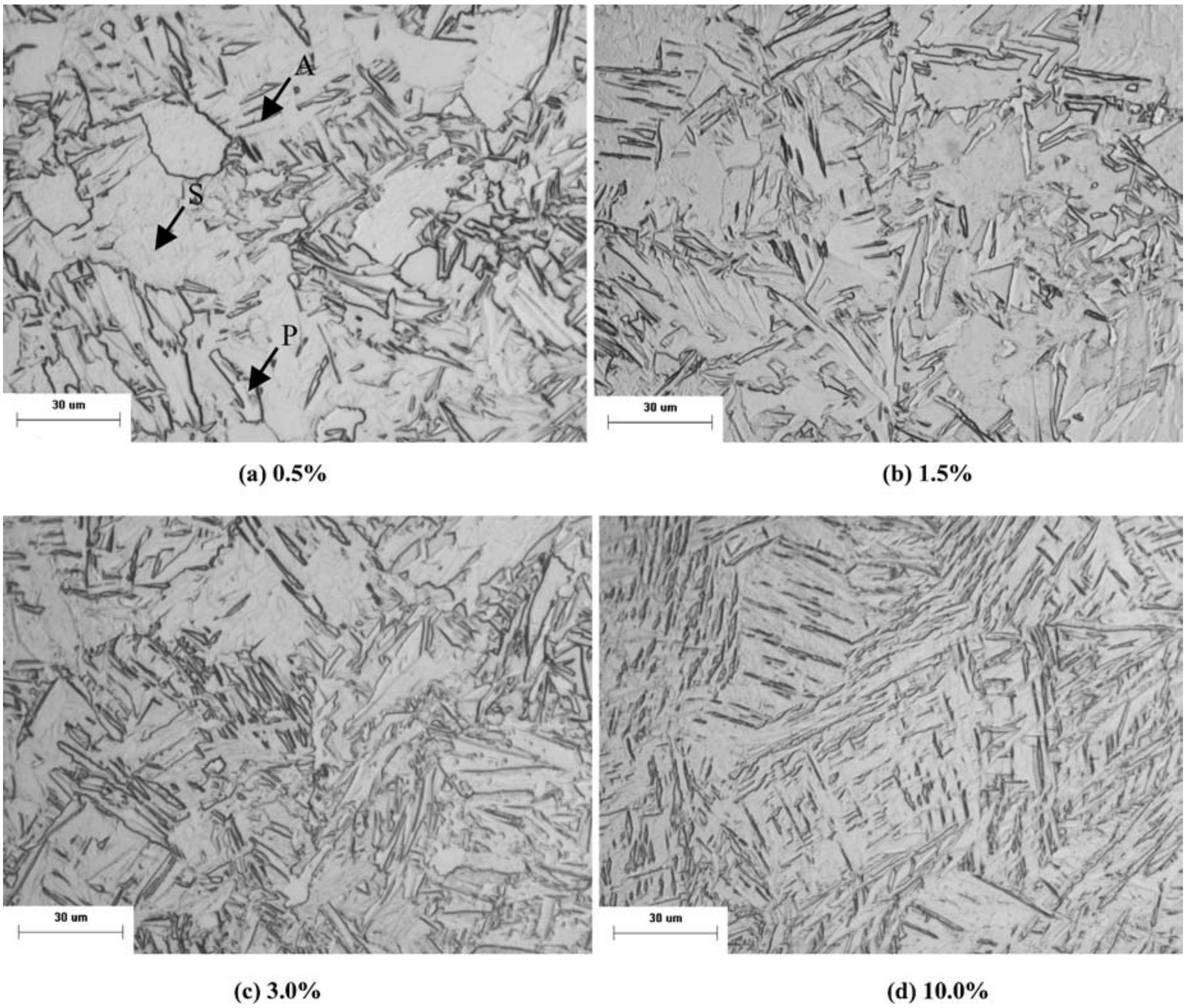

Figure 3 Cross sections of welds made with various oxygen content in argon shielding gas.

The increase in oxygen content also increased the surface hardness from 242 Hv on welds with pure argon shielding gas, to 373 Hv when the oxygen content was 10%. These data may be compared to the hardness of 191 Hv found on the surface of the base metal. This increase in hardness as the oxygen content increased was due to both increased surface oxide film thickness and increased oxygen diffused into the surface layer of the weld. Therefore, surface hardness could be a good indication of the gas shielding deficiency.

3.2. Weld structures and properties

Fig. 2 shows the microstructures of the base (2a) and weld metal (2b) with pure argon shielding gas. The welding thermal cycle when using pure argon shielding gas changed the base metal microstructure from equiaxed alpha of relatively uniform grain size (average about 10–15 μm) to a mixed coarse-grained serrated alpha and a smaller amount of fine-grained acicular alpha (Fig. 2b, pointed by arrow S and A). Addition of oxygen (up to 0.5–1.0%) in the argon shielding gas

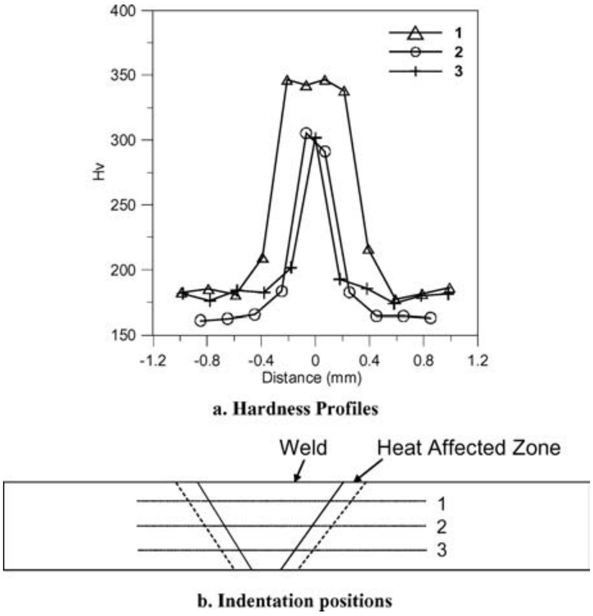

Figure 4 Hardness profiles along the cross section of weld made with 10% oxygen content.

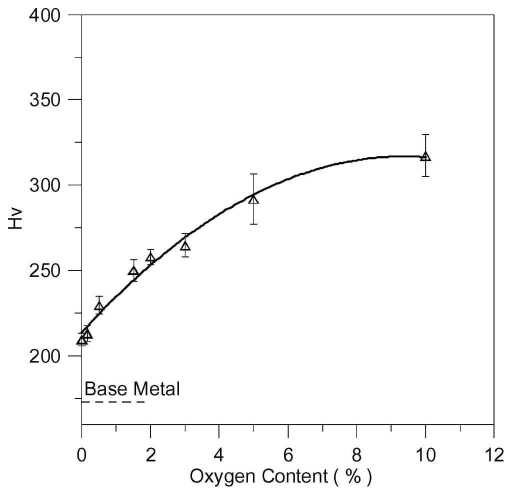

Figure 5 Weld hardness versus oxygen content in argon shielding gas.

reduced both grain size and amount of the serrated alpha, and increased the amount of acicular and platelet alpha (Fig. 3a, pointed by arrow S, A and P). As the oxygen content continued to increase (above 1.5–2.0%), weld microstructure was dominated by the acicular and platelet alpha. At very high oxygen content, the alpha platelets formed in colonies, giving a basketweave appearance (Fig. 3d).

The significant changes in weld microstructures because of increased oxygen content were not observed in GTAW of commercially pure Ti in which serrated and acicular alpha [9, 10] were the typical constituents at increased oxygen contents [9, 10]. There are two possible reasons for this discrepancy between GTAW and laser welding. First, the range of oxygen content covered in those GTAW studies was relatively narrower, with air contamination up to 2% [10], compared to the oxygen contamination up to 10% in this work. Second, the microstructural change in laser welding is much more sensitive to the oxygen content because of the much faster cooling rates compared to those in GTAW. For example, typical cooling rates in laser welding range from hundreds to thousands of degrees centigrade per second (comparable to those in water quenching) while those in GTAW are in the range of tens of degrees centigrade per second [12]. In fact, it has been suggested that alpha prime (martensite alpha) may be present in commercially pure Ti and Ti-6Al-4V laser welds because of low heat input and rapid cooling by the surrounding cold material [7]. Comparative studies of near alpha and alpha-beta Ti alloys using GTAW, and laser and electron beam welding, have indicated that the phase transformation in welds changes from diffusion-controlled nucleation and growth, to martensitic mechanisms as cooling rates increase [12, 14].

The hardness varied across the weld region because oxygen diffused into weld metal through the surfaces and the oxygen content was higher near surfaces compared to the centre (Fig. 4). Therefore, the average value (from three, two and one indentations in weld region respectively at locations 1, 2 and 3 as shown in Fig. 4) was plotted in Fig. 5 for the effect of oxygen content. The hardness of the weld metal with pure argon shielding gas was 209 Hv, which compared to that of the base metal at 173 Hv. The oxygen content increased the weld hardness all the way to 317 Hv for the weld with 10% oxygen, which came from both microstructural change and absorbed and diffused oxygen in the weld metal, as indicated by Wang and Welsch [5]. It is interesting to note the large increase of hardness of the welds with pure argon shielding gas over the base metal, which is opposite to the observation that GTAW bead-on-plate welds with low oxygen content were softer than the CP-2 Ti base metal [9]. The difference in hardness change is believed to be caused by the different cooling rates between GTAW and laser welding, similar to the difference in weld microstructure of GTAW and laser welds. In fact, Harwig [9] also noted that the extent of weld softening became smaller as the cooling rate increased.

Fig. 6 shows the effect of oxygen content on the tensile strength of the welds. The oxygen content increased the strength from 594 MPa for the welds with pure argon shielding (compared to 592 MPa for the base metal) to about 620–630 MPa when the oxygen content was

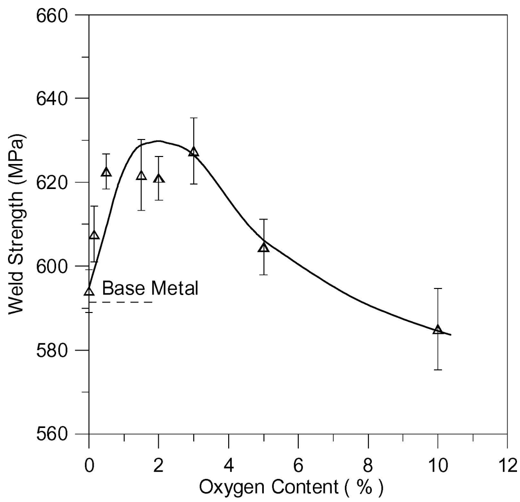

Figure 6 Weld tensile strength versus oxygen content in argon shielding gas.

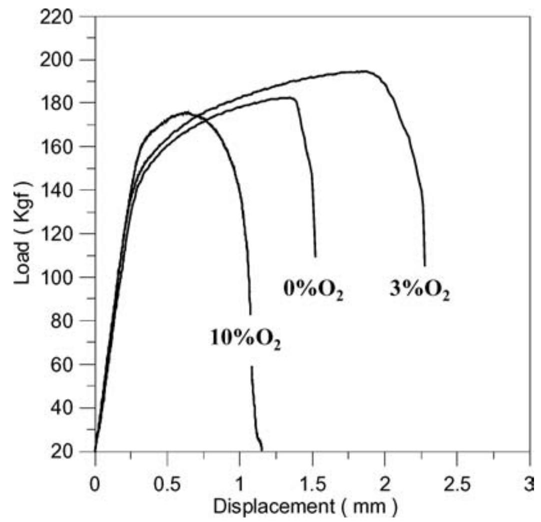

Figure 7 Load-displacement curves for welds made with various oxygen contents.

about 2–3%. However, as the oxygen content continued to increase, the weld strengths started to decrease and eventually became even lower than that of the base metal (Fig. 6). This was not surprising, since the tensile specimen geometry shown in Fig. 1 causes a substantial strain concentration at the edge notches, which should cause early crack initiation in the lower ductility welds made with high oxygen content. The resultant reduced maximum force associated with low overall strain is indicated in the typical load-displacement curves in Fig. 7.

Generally, the tensile fractures initiated at the notch and crossed the three areas including weld, fusion line and base metal (Fig. 8). Weld metal showed less ductile behaviour, especially when the oxygen content was high. Fig. 9 shows the typical fracture surfaces of the welds (in the weld zone in Fig. 8) made with various oxygen contents in the shielding gas. While the base metal fractured in a ductile manner with classical dimple morphology, the fracture of the welds made with pure argon shielding gas was similar to that of the base metal but included some locations with less ductile features (Fig. 10). The introduction of oxygen into the shielding gas greatly reduced the ductility by replacing the ductile dimple structure with relatively brittle quasi-cleavage morphology. When the oxygen content was high (e.g., at 10% in Fig. 9d), there were little signs of plastic deformation, with the fracture path appearing to be intergranular along colony boundaries or cleavage across colonies, which indicates that the fracture mode changed completely from ductile to brittle. This was very similar to an investigation on the effects of oxygen on mechanical properties of an alpha titanium alloy Ti-6Al-2V, in which oxygen caused a change from good ductility at low oxygen concentration (0.07 wt%) to total brittleness at high 0.65 wt% [15].

3.3. Shielding gas deficiency and mechanical property requirements

The above experimental results have indicated correlations between weld surface color, and weld

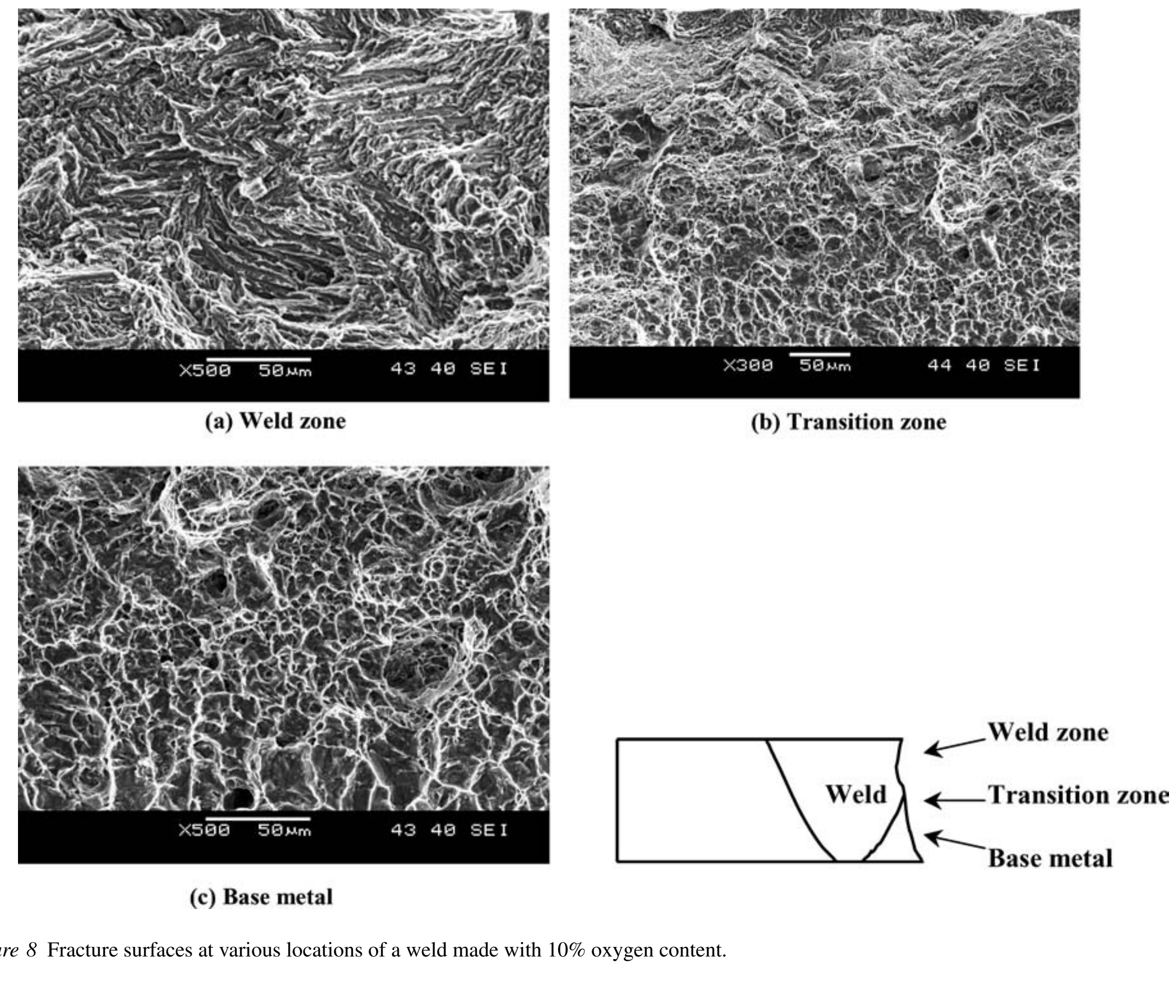

Figure 8 Fracture surfaces at various locations of a weld made with 10% oxygen content.

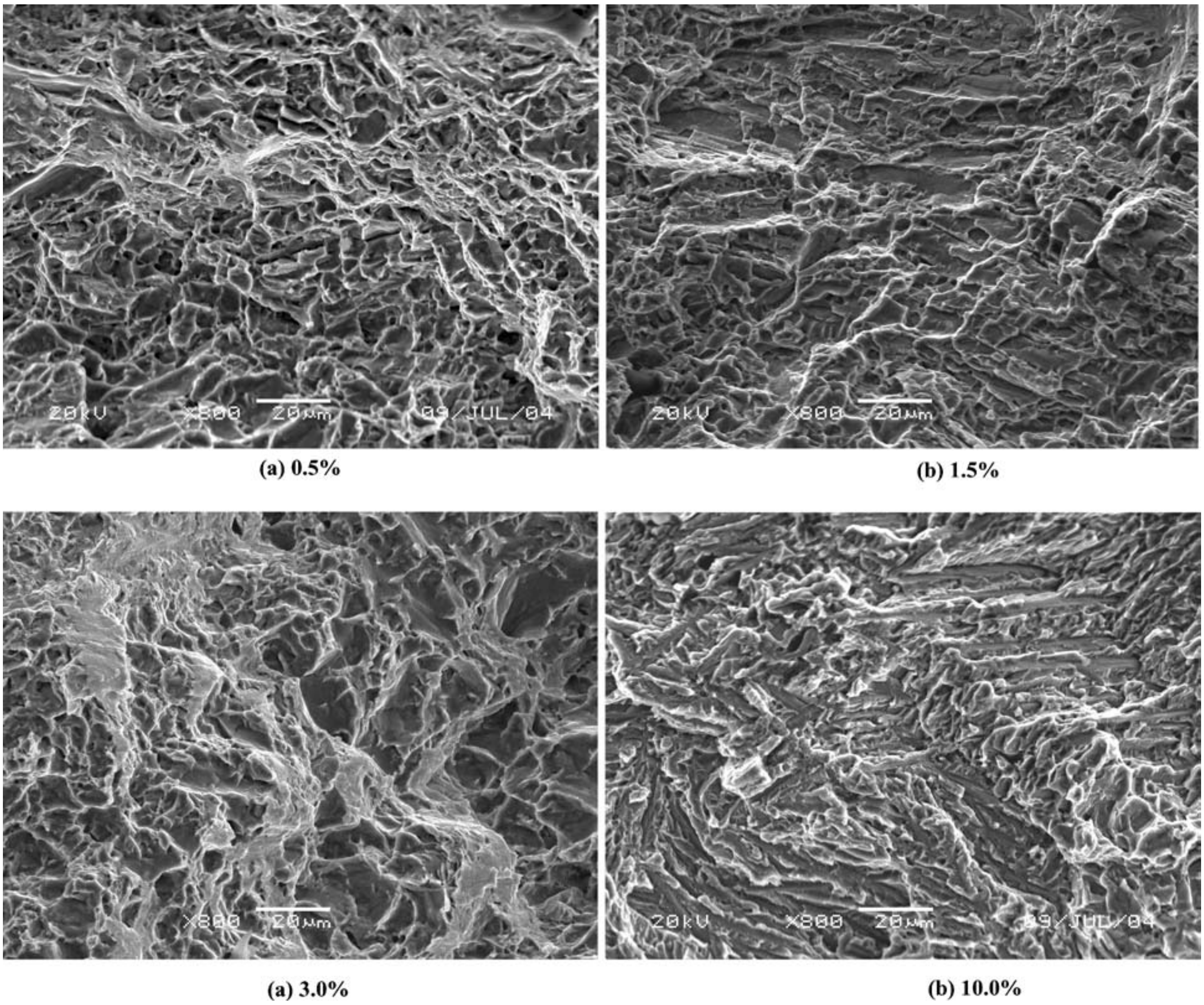

Figure 9 Fracture surfaces of welds made with various oxygen content in argon shielding gas.

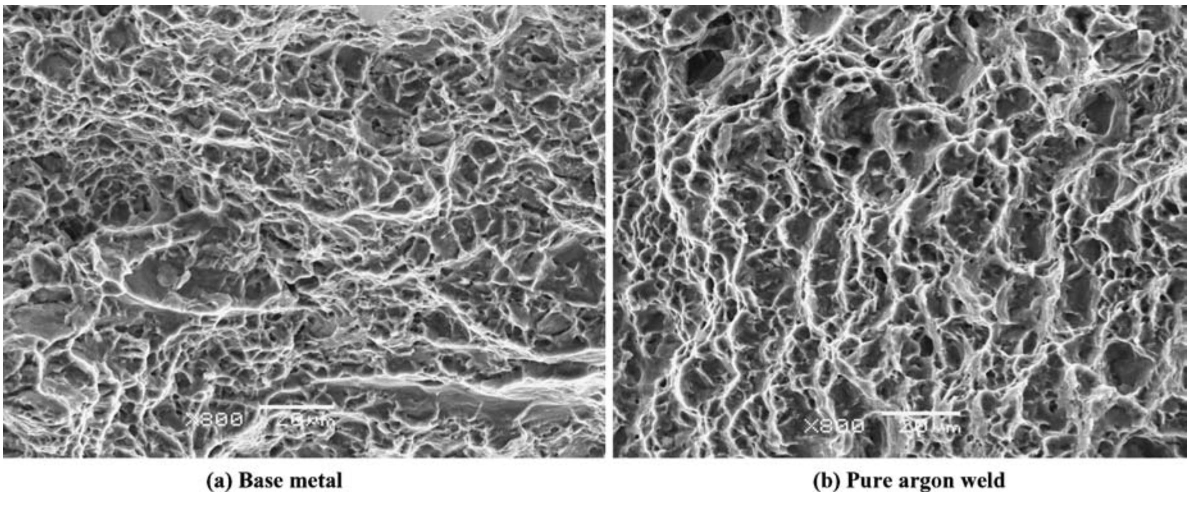

Figure 10 Fracture surfaces of (a) base metal and (b) weld made with pure argon shielding gas.

microstructure and mechanical properties (strength, ductility, hardness). But the question remains as to what level of oxygen contamination would be acceptable during laser welding of Ti.

For structural welding, AWS D10.6-91 [9] suggests that a satisfactory weld should have a hardness that does not exceed the base metal hardness by 5 R_B (which is equivalent to about 30 Hv). The ASME Boiler and Pressure Vessel Code Division 1, Section VIII [10] also suggests that a weld hardness increase over the base metal of more than 40 Brinnel hardness number (which is equivalent to about 40 Hv in the range of hardness values considered for Ti welds) indicates excessive weld contamination. But, in this work, the welds with pure argon shielding gas had already a hardness increase over base metal of about 36 Hv.

Another commonly used hardness standard for Ti welds suggests a maximum weld hardness of 250 Hv [10]. This would translate into an acceptable oxygen content up to 2% in Fig. 5, which appears to be consistent with the shielding oxygen content at 1.5–2% that caused significant changes in weld microstructure and fracture surface morphologies (Fig. 9). However, any acceptable levels of oxygen contamination in the shielding gas would also depend on how much oxygen could be absorbed into weld metal and how fast the weld would cool down from high temperature. Further work is needed to study the effects of oxygen contamination under different welding conditions, such as heat input, sheet thickness, and joint and fixture design.

4. Summary

This work investigated the effects of oxygen contamination in the argon shielding gas on weld microstructures and properties during laser welding of commercially pure titanium thin sheets. The experimental results, mainly analyzed by optical and scanning electron microscopy and mechanical testing, have indicated correlations between weld surface color, weld microstructure and mechanical properties (strength, ductility, hardness). The main conclusions include:

- Weld surface colour changed from silver to dark blue as the oxygen content in argon shielding gas increased from 0 to 10% but may not be a good indication of the shielding deficiency because the color sequence would repeat itself as the oxidation thickness increases. On the other hand, weld surface hardness, while increased as the oxygen content increased and also correlated to weld metal hardness, could be a better inspection tool of the shielding deficiency.
- Addition of oxygen in the shielding gas, while reducing coarse-grained serrated alpha, increased finer grained acicular and platelet alpha in weld metal. When the oxygen content was above 1.5%, weld metal was dominated by acicular and platelet alpha. As the oxygen content continued to increase, the alpha platelets occurred in colonies, giving a basketweave appearance.
- As the oxygen content increased, weld metal strength increased first and then started to drop when the oxygen content was above 2%. This was apparently caused by the occurrence of brittle fracture as a result of the formation of acicular and platelet alpha.

References

1. ASM Handbook, *Propert. Select.: Nonferr. Alloys Spec.-Purp. Mater.* **2** (1990) 586.
2. J. GEDOPT and E. DELARBRE, *Proc. SPIE* **4088** (2000) 264.
3. S. SHRIVASTAVA, Medical Device Materials, 8–10 September 2003, Anaheim CA, in Proceedings of the Materials and Processes for Medical Devices Conference, p. 417.
4. J. LIU, I. WATANABE, K. YOSHIDA and M. ATSUTA, *Dental Mater.* **18** (2000) 143.
5. R. R. WANG and G. E. WELSCH *J. Prosth. Dent.* **74**(5) (1995) 521.
6. J. C. BORLAND, *Brit. Weld. J.* **February** (1961) 61.
7. T. YAMAGISHI, M. ITO and Y. FUJIMURA, *J. Prosth. Dent.* **70**(3) (1993) 264.
8. H. W. A. WISKOTT, T. DOUMAS, S.S. SCHERRER and U. C. BELSER, *J. Mater. Sci.: Mater. Med.* **12** (2001) 719.
9. D. D. HARWIG, C. FOUNTAIN, W. ITTIWATTANA and H. CASTNER, *Weld. J. Weld. Res.* **Nov. Suppl.** (2000) 305s.
10. P. Y. Y. MAAK, The Effect of Air Contamination in the Argon Shielding Gas on the Mechanical Properties of Titanium Gas-Tungsten-Arc Welds, TR-282, Scientific Document Distribution Office (SDDO), Atomic Energy of Canada Limited, Chalk River, Ontario, K0J 1J0, July 1984.
11. E. BIRO, D. C. WECKMAN and Y. ZHOU, *Metall. Mater. Trans. A* **33A** (2002) 2019.
12. W. A. BAESLACK III and F. D. MULLINS, *J. Mater. Sci. Lett.* **1** (1982) 371.
13. J. TALKINGTON, D. HARWIG, H. CASTNER and G. MITCHELL, *Weld. J.* **March** (2000), 35.
14. K. K. MURTHY and S. SUNDARESAN, *J. Mater. Sci.* **33** (1998) 817.
15. Z. LIU and G. WELSCH, *Metallu. Trans. A* **19A** (1988) 527.

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