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Joining steel tubes employing Fe-Si-B metallic glasses as filling material

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Bonding of seamless, low carbon, steel tubes were performed by means of a transient liquid phase bonding (TLPB) process employing a Fe-Si-B glassy metal as filling material. The butt joints were performed by induction heating under a moderate pressure and argon flux. Thermal cycles consisting of a fast heating followed by an isothermal stage and ending with a controlled cooling down to room temperature were applied. The isothermal stages performed for a time t at temperature T ($T_W < T < T_T$ with T_W and T_T the melting temperature of the glassy metal and the tube materials respectively).

The microstructures of the joint zones were analyzed employing optical and scanning electronic microscopy, energy dispersive X-ray and electron probe microanalysis. The diffusion of the elements during the process was studied. The bonding process was modeled in order to determine the Si distribution for different length of the isothermal stage. The results from modeling are compared to the experimental data and the bonding process is discussed. *© 2005 Springer Science + Business Media, Inc.*

1. Introduction

Welding processes are of great importance in several areas of engineering. Therefore, the improvement of the existing methods and the development of new procedures in order to obtain joints with better characteristics of the microstructure and properties are required. The application of filling materials, cut to the required size from the glassy metal ribbons (washers) [1], to the transient liquid phase bonding processes (TLPB) [2, 3] offers several advantages. The toughness of the glassy metals allows the easy compliance of the metallic washer to the tube surfaces while its small thickness reduces the bonding time.

This method was used in this work to produce but joints in seamless steel tubes similar to those employed in the petroleum industry.

2. Transient liquid phase bonding process

The TLPB method, using glassy metals, can be applied to join two pieces of a metal A positioned with their matted faces in contact with a thin amorphous layer B as in Fig. 1.

This method can be explained as follows: The chemical composition of B is near to the eutectic one of the system A–C (Fig. 2). The melting temperature of B (T_B) is lower than that of A (T_A). If the temperature in the joining zone is raised to T at the higher attainable rate, being $T_B < T < T_A$, a transient liquid phase gap, with an initial composition x_B, is originated. As the temperature T remains constant the following process occurs:

(a) The A atoms are dissolved from the pieces into the liquid gap and the gap becomes wider,

(b) when the liquidus composition x_L, at temperature T, is attained, the solidification begins and the thinning of the gap proceeds. When the gap disappears in the solid joint remains a belt zone of composition x_S.

(c) The solid state diffusion occurs until the chemical composition in the joining zone becomes homogeneous. Moreover, if the diffusion coefficient of C in A is higher than the self diffusion coefficient of A, the picture must include, along the three steps, the simultaneous diffusion of C into the pieces made of A, whith a decrease of the process length.

In the case that A bars are single crystalline, a planar diffusion front can be modeled, but as the bars are polycrystalline the diffusion along grain boundaries should be considered. The glassy nature of the layer is not a precondition for the transient liquid phase bonding process but due to the toughness of the glassy metals it is possible to use a very thin layer, which contributes to

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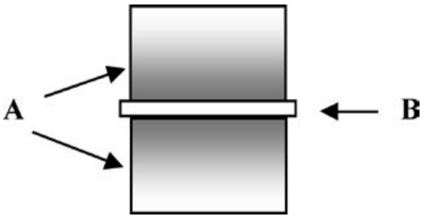

Figure 1 Outline of the accomodation of the pieces of material A to be joined by means of a TLPB procedure employing a filler of material B.

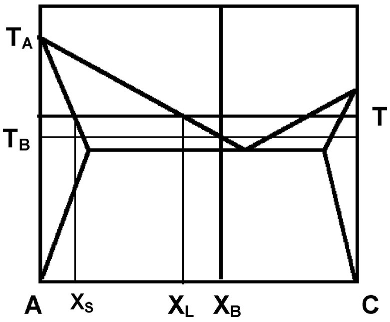

Figure 2 Equilibrium phases diagram of the *A*–C system. T is the isothermal treatment temperature, X_B the initial composition of the transient liquid gap, X_L the composition of the liquid gap as a solid phase of composition X_S precipitates.

reduce the time of the bonding process. Moreover, the toughness of the glassy metals do let to handle and cut easier any shape for the required layer. The elasticity of the glassy metals improves the contact between the layer and the pieces when pressure is applied.

The outcome of this process is a joint with similar features to those of the original material with only slight variations in composition or microstructure.

3. Experimental procedure

A glassy metal washer (25 μm thick), cut to size from a wide METGLAS® SA1 ribbon of composition $Fe_{78.35}B_{13.20}Si_{8.45}$ at.% provided by Honeywell, was positioned in contact with the mated butts of the seamless low carbon steel tubes (DS 020) to be bonded. The external diameter of the tubes is 21.50 mm and the wall thickness is 3.70 mm. The steel composition is reported in Table I.

TABLE I Composition of the steel of the seamless tubes joined in this work (wt%)

C	Mn	Si	Ni	Cr	Mo	V	Cu	Sn	Al	Ti
0.19	0.47	0.26	0.038	0.05	0.03	0.001	0.102	0.008	0.03	0.002

Previously, the washer surface and the tube butts were polished with wet sandpapers of 500 and 1000 granulometry and cleaned in an ultrasound basin. The tubes were aligned under a moderate pressure and fastened employing the device depicted in Fig. 3. Then, the ensemble was located in an induction coil under Ar flux and heated up to a temperature $T = 1220^\circ$C.

Fig. 4 depicts the thermal cycles. Tests with isothermal stages of 1, 3 and 5 minutes were performed. Finally, a cooling was done with controlled rate.

Metalographic samples were prepared from longitudinal pieces cut from the wall of the tubes and mounted in bakelite. The samples were grounded and polished up to 1 μm diamond, and 1% Nital as chemical reactive was used. The microstructure was studied by means of optical microscopy (OM), scanning electron microscopy (SEM) and energy dispersive X-rays analysis (EDX). Moreover, complementary analyses using an electron probe microanalysis (EPMA) were performed to determine the distribution of Boron, Carbon, Oxygen and Silicon in the joining zone.

4. Experimental results

4.1. Metallographic observation

Micrographs obtained in an optical microscope, show the progress of the bonding process (Fig. 5). Pictures were taken at a depth of 3/4 of the thickness of the wall of the tubes relative to the external surface and at increasing distances from the joining zone.

The optical micrograph corresponding to a 1 min isothermal heating, shows a joining line that indicates a clear structural separation between the joined pieces by a remaining filling material. This separation is not so sharp in the other two samples ($3'$ and $5'$ processes). However a $\sim$100 μm fringe of ferritic phase (white) was observed in the joining zone in the three samples. Around that ferritic fringe a region with coarse grain was produced, moreover, some of them showed widmanstaten structure. This characteristic is clearly observed up to $\sim$1 mm from the joining line. The grain boundaries of the austenitic grains transformed on cooling down are pointed out by dark dashes denoting solutes segregation. Small volume

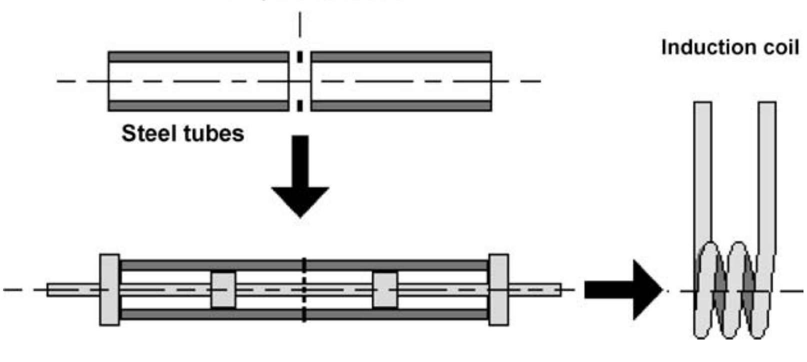

Figure 3 Assembly of the alignment and fixing device.

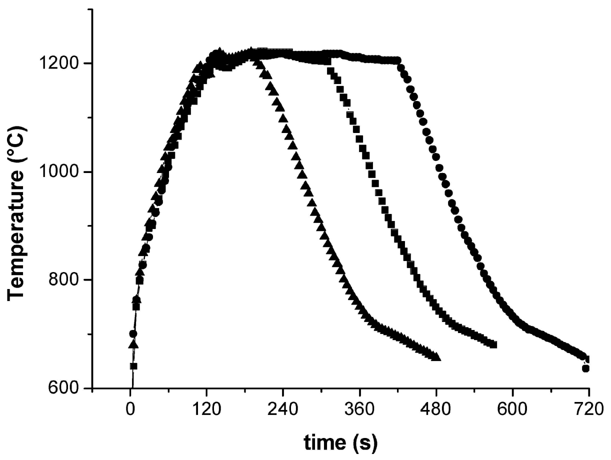

Figure 4 Thermal cycles applied to join steel tubes employing Fe-Si-B ribbons.

fraction of bainitic structure was also observed in some samples.

4.2. Energy dispersive X-ray analysis

The elements searched by means of EDX were Si, Fe and Mn. Mn atoms were observed to be homogeneously distributed in the sample area analysed. Si and Fe concentration profiles present complementary shapes. Fig. 6 shows the concentration of Si (in at.%) as a function of the distance from the joint of the bonded tubes. It is observed that: (a) the Si concentration profiles present a maximum in the joining zone due to the higher initial Si concentration in the glassy metal. The initial Si profile has a barrier form with sharp borders in coincidence with the glass surfaces; (b) the maximum of the Si profile decrease as the time increase, meanwhile the diffusion process progress; (c) far from the joint, the Si concentration tends towards the Si nominal concentration in steel.

4.3. Electron probe microanalysis

Fig. 7a shows the secondary electron image from the joint line of a sample isothermally treated for 3 min at 1220°C.

Mappings of boron and silicon are also depicted in Figs 7b and c. It is noteworthy that boron is mainly localized in the remain of the glassy metal and also in grain boundaries. Nevertheless, a rather uniform distribution of boron is spread all over the rest of sample area observed pointing out that boron diffusion intra grains during the isothermal stage is significative.

Silicon is observed to be localized in a frame at the joining zone.

Boron and silicon profiles (Fig. 7d) were also obtained on a line parallel to the axe across the tubes joint. Boron concentration profile presents relative maximums in coincidence with the grain boundaries. That could means that (a) boron grain boundary diffusion is relatively more significant than boron intragrain diffusion in austenite [4] and (b) boron segregation towards grain boundaries proceeds on cooling down [4, 5]. On the other side, the diffusion front of Silicon is plane but its diffusion coefficient is relatively very small in comparison with that of boron.

As a consequence, the time needed for to homogenize the chemical concentration of the alloying elements along the joined samples is determined by the diffusion of silicon.

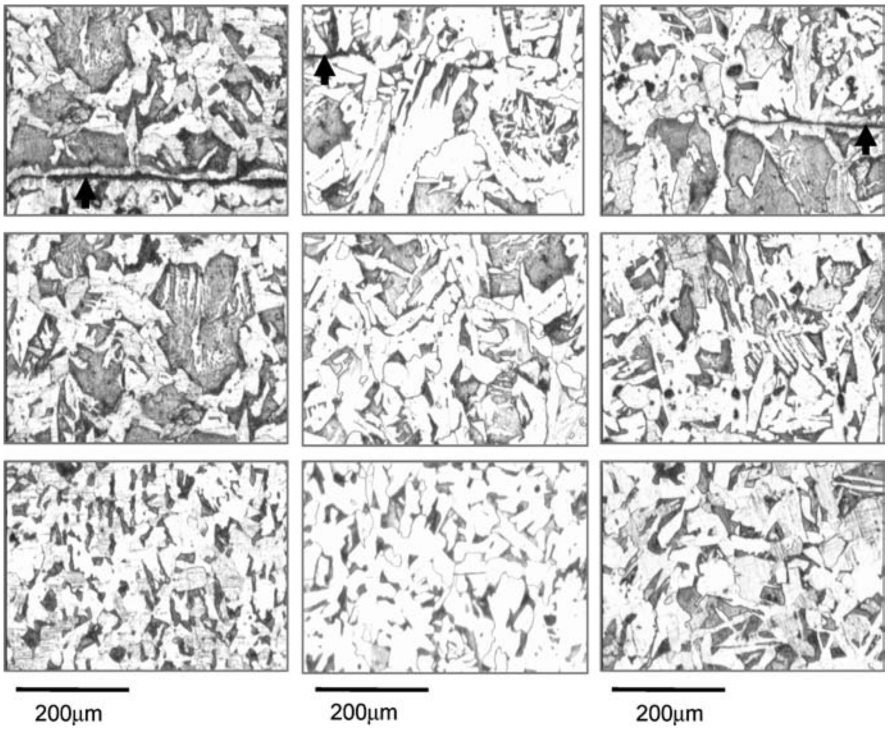

Figure 5 Optical micrographs of samples isothermally heated at 1220°C for different times: 1 min (left hand column), 3 min (central column) and 5 min (right hand column). The joining zones are depicted in the top file. Joining lines are indicated by arrows. Heat affected zones are shown in the central file. The microstructure of zones located 10 mm far from the joining line are shown in the bottom file.

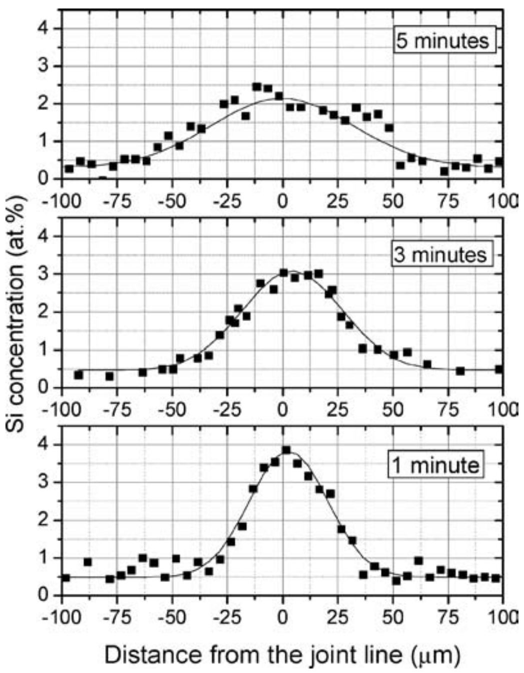

Figure 6 EDX determination of Si concentration (Dots: experimental points, solid lines: fittings).

5. Discussion

As stages (a) and (b) of a TLPB process are relatively very short, [6] a quantitative analysis of the concentration profiles can be performed assuming that the process consist only on stage (c). That is, the concentration profile of Silicon can be analyzed employing the expression that represents the dependence of the concentration of the diffusing component $c(x, t)$ on time and position relative to the center of a source of finite thickness $2a$ and initial concentration c_0 [7].

$$c(x,t) = c_o/2\{\mathrm{erf}[(a-x)/(2(D(T)t)^{1/2})] + \mathrm{erf}[(a+x)/(2(D(T)t)^{1/2})]\} \qquad (1)$$

D (T) is the diffusion coefficient and depends only on temperature T.

Equation 1 was employed to fit the Si concentration profiles for each t. The fittings are depicted close to the experimental data in Fig. 6. The most important parameters obtained from the fitting are $c_o = 7.42$ at.%, $a = 10$ μm and D $(T) = \times\ 1.86 \times 10^{-8}$ cm^2/s.

Alternatively, the diffusion coefficient can be obtained from the following expression [7]

$$D(T) = D_o \exp(-E_a/RT) \qquad (2)$$

where D_o is the pre-exponential factor, E_a the activation energy for diffusion and R the gases constant. In the case of Si diffusing in crystalline Fe [8]:

$$D_o = 0.735\mathrm{cm}^2/\mathrm{s} \quad \text{and} \quad E_a = 52.51\mathrm{Kcal/mol}. \qquad (3)$$

Employing these data, the diffusion coefficient at T = 1220°C results D $(T) = 1.55 \times 10^{-8}$ cm^2/s. This value is in agreement with the diffusion coefficient obtained from the fitting of Equation 1.

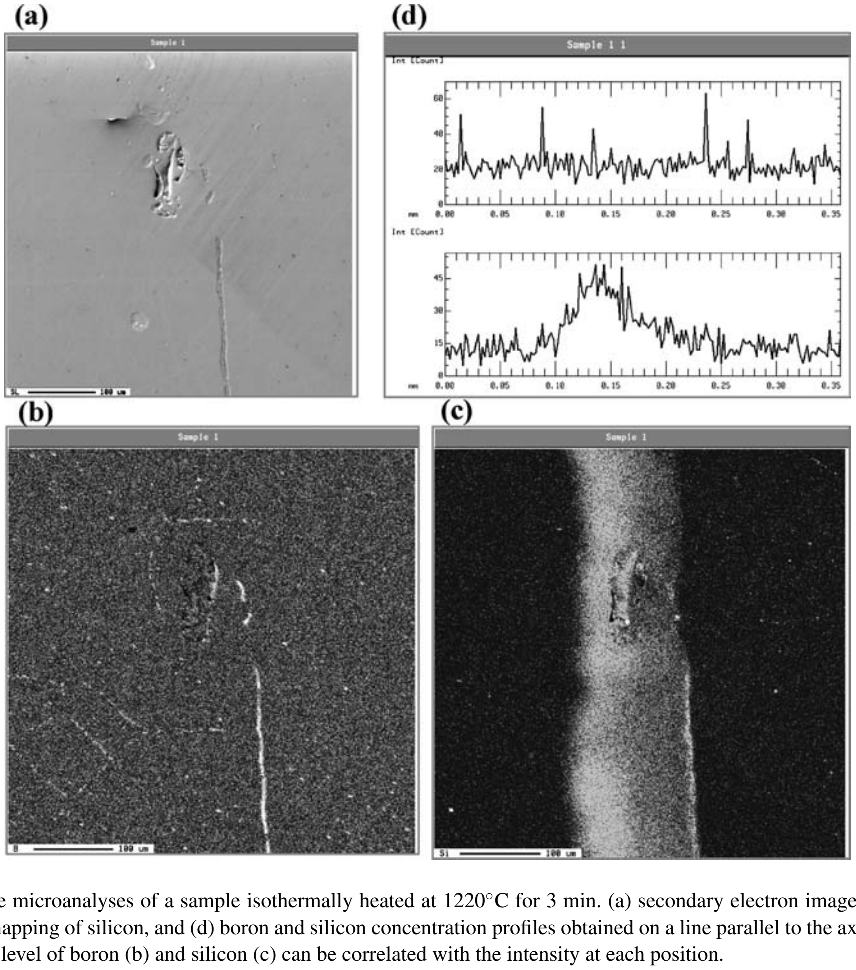

Figure 7 Electron probe microanalyses of a sample isothermally heated at 1220°C for 3 min. (a) secondary electron image of the joining line, (b) mapping of boron, (c) mapping of silicon, and (d) boron and silicon concentration profiles obtained on a line parallel to the axe across the tubes joint. The local concentration level of boron (b) and silicon (c) can be correlated with the intensity at each position.

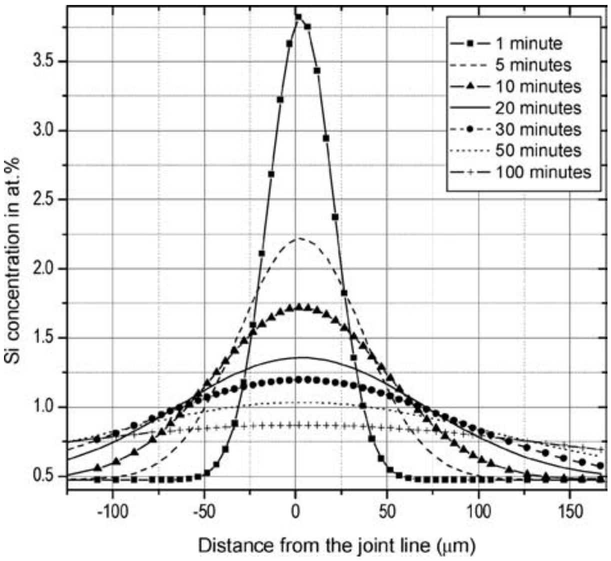

Figure 8 Si concentration profiles evaluated as a function of the isothermal stage time *t* at 1220°C.

The fact that $2a$ is smaller than the thickness of the amorphous ribbon and consequently much smaller than the liquid gap thickness at the end of stage b) of TLPB process suggests that some liquid is lost as a consequence of the moderate pressure applied. The composition $c_0 = 7.42$ at.% fitted from Equation 1 is reasonable on the basis that the Si concentration of METGLASS® SA1 is 8.45 at.%. When the liquid gap widens up to its maximum width Si concentration lowers approaching the c_0 value estimated.

The knowledge of the relevant parameters of Equation 1 let us to predict the temporal evolution of Si concentration profile at the temperature of the isothermal stage. If, in addition, Equation 2 and the experimental data (3) are assumed as true, the concentration profile can be obtained for any T and t values.

This fact let us to emphasize the significance of the analyses performed. Fig. 8 depicts the concentration profiles of Si at different t, evaluated as pointed above. At $T = 1220°\mathrm{C}$ the time needed to obtain a homogeneous distribution of Si atoms is higher than 20 min.

6. Conclusions

Seamless steel tubes were bonded employing METGLAS® SA1 glassy ribbons as filler material. The joints were characterized by means of optical and electron microscopy with EDX and EPMA. It is observed that the diffusion coefficient of boron is significantly larger than that of silicon. Moreover, while diffusion of silicon can be represented by means of a plane diffusion front, boron atoms seem to diffuse distinctly in grains and in grain boundaries. The segregation of Boron atoms in the grain boundaries is observed.

The influence of the isothermal process length was analyzed. It is concluded that the presence of Si in the composition of the metallic glass compels to enlarge the duration of the isothermal stage above 20 min for $T = 1220°\mathrm{C}$.

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