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# Numerical Simulation of Brazing Aluminium Alloys with Al–Si Alloys

J. Lacaze<sup>1</sup>  · B. Jacques<sup>1,2</sup> · T. Mazet<sup>2</sup> · M. Vynnycky<sup>3</sup>

**Abstract** Joining parts using low-melting temperature alloys has long been used for manufacturing complex components such as heat exchangers made of aluminium alloys. Investigations of the process have shown that core/clad interaction during heating and brazing can lead to a significant decrease in the amount of liquid available for joint formation. This study presents a transient one-dimensional model for the process that takes into account the diffusion of silicon and the movement of the core/clad interface, with the model equations being implemented in the finite element software COMSOL Multiphysics; the results are compared to literature experimental data. Silicon profiles in the core are well described, while there appears a significant difference between predicted and experimental values of remaining clads which suggest a strong effect of silicon diffusion and liquid penetration at core grain boundaries.

**Keywords** Brazing · Aluminium alloys · Solid-state diffusion · Melting

## 1 Introduction

Controlled atmosphere brazing (CAB) of aluminium 3xxx alloys with Al–Si clad alloy is used to manufacture complex components such as heat exchangers. Figure 1a shows schematically the original brazing sheet with a clad layer on top of the core plate. The clad layer is a silicon-rich alloy of the 4xxx series and consists of rounded silicon precipitates within an (Al) matrix, while the core contains very little silicon but some manganese and is made of small-sized Mn-rich precipitates within an (Al) matrix. Brazing is more often carried out at a temperature within the melting interval of the clad or at a higher temperature, in which case the clad is fully liquid. Accordingly, the brazing process may be described by three or four successive steps [1, 2]:

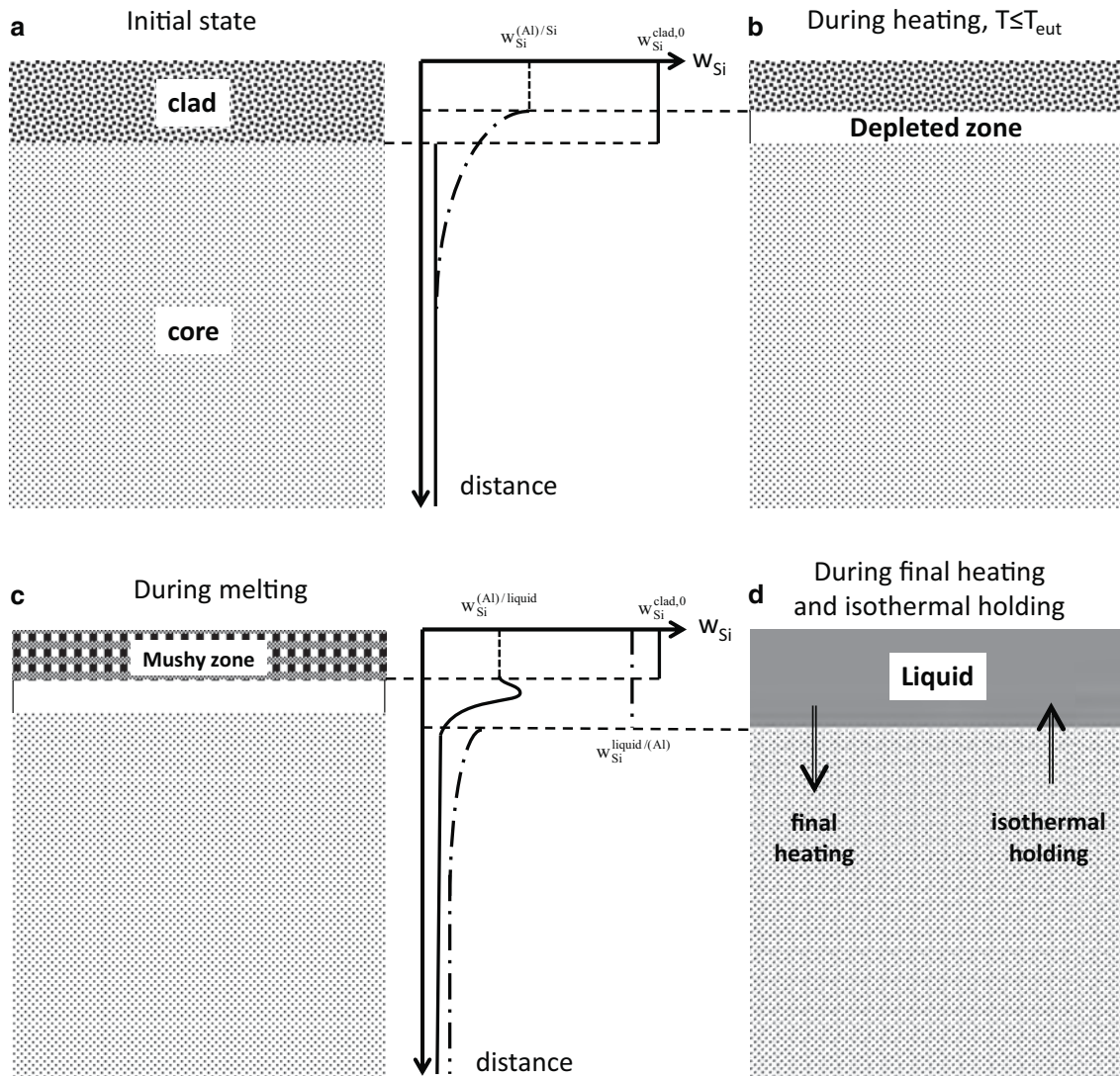
1. During heating of the assembly up to the melting temperature of the clad layer, solid-state diffusion of silicon from the clad to the core leads to progressive dissolution of silicon precipitates in the clad at the core/clad interface. The so-called depleted zone (Fig. 1b) thus develops, which was investigated a long time ago by Terrill [1].
2. When the Al–Si eutectic temperature  $T_{\text{eut}}$  is reached, the clad starts melting and this may be assumed to occur homogeneously within the thickness of the remaining clad layer. As the temperature of the assembly further increases, the silicon content of the (Al) matrix within the clad and the core at the core/clad interface decreases as it follows the solidus in the Al–Si phase diagram  $w_{\text{Si}}^{(\text{Al})/\text{liquid}}$  (Fig. 1c). The peak in silicon content which thus appears in the core is, however, quickly smoothed.

✉ J. Lacaze  
Jacques.lacaze@ensiacet.fr

<sup>1</sup> CIRIMAT, Université de Toulouse, 4 allée Monso, 31030 Toulouse, France

<sup>2</sup> Fives Cryo, 25 Bis Rue du Fort, B.P. 87 88194 Golbey Cedex, France

<sup>3</sup> Department of Materials Science and Engineering, KTH Royal Institute of Technology, Brinellvägen 23, 100 44 Stockholm, Sweden



**Fig. 1** **a** Initial state of the clad/core assembly; **b** formation of the depleted zone in between the clad and the core because of solid-state diffusion of silicon during heating below the Al–Si eutectic temperature  $T_{eut}$ ; **c** homogeneous melting of the clad at increasing temperature up to the clad liquidus temperature; **d** dissolution (downward arrow) of part of the core if the brazing temperature is above the liquidus temperature of the clad during final heating and then isothermal solidification (upward arrow) during holding

- If the temperature is increased above the liquidus temperature of the clad, the core starts dissolving so that the composition of the liquid in the clad follows the liquidus of the Al–Si phase diagram  $w_{Si}^{liquid/(Al)}$ . This dissolution first affects the depleted zone and then goes further into the core if the upper brazing temperature is high enough. This step is indicated with a downward arrow in Fig. 1d. Dissolution does not occur if the temperature remains lower than the clad liquidus.
- During holding at the brazing temperature, diffusion of silicon to the core leads to a decrease in the quantity of remaining liquid clad which is illustrated with the upward arrow in Fig. 1d. Unlike in the TLP bonding

process [2], isothermal solidification is generally not pursued to its end and the remaining liquid solidifies during cooling of the brazed assembly. This solidification leads to cells or dendrites developing from the core/clad interface towards the upper surface of the clad. In the intercellular areas and at the upper surface, solidification ends with precipitation of Al–Si eutectic. This step 4 applies to brazing at temperatures both higher and lower than the liquidus temperature of the clad.

In order to gain a better insight into the brazing process, a numerical analysis has been developed that allows accounting for changes with temperature of the diffusion coefficients and of the composition at various interfaces, which is not feasible

with an analytical method. This approach is here applied to literature information concerned with brazing at temperatures lower than the clad liquidus temperature.

## 2 Mathematical Model

### 2.1 Equations

Following the above description, diffusion will be considered only in the core and the problem is simplified by considering only Al–Si alloys. Diffusion is assumed to follow Fick's second law and will be described by assuming that diffusion coefficients do not depend on composition and that the molar volume is the same for all phases. For a 1D problem, one thus has:

$$\frac{\partial w_{\text{Si}}^{\text{core}}}{\partial t} = D_{\text{Si}}^{\text{core}} \cdot \frac{\partial^2 w_{\text{Si}}^{\text{core}}}{\partial z^2}, \quad (1)$$

where  $w_{\text{Si}}^{\text{core}}$  is the silicon content,  $t$  the time,  $z$  the space coordinate with  $z = 0$  being the centreline of the core, and  $D_{\text{Si}}^{\text{core}}$  the silicon diffusion coefficient given by [3]:

$$D_{\text{Si}}^{\text{core}} = 1.38 \cdot 10^{-5} \cdot \exp\left(-\frac{117,600}{R \cdot T_K}\right) \text{ m}^2 \cdot \text{s}^{-1}, \quad (2)$$

where  $T_K$  is the temperature in Kelvin.

The boundary conditions for (1) are as follows. Since  $z = 0$  constitutes an axis of symmetry, the silicon flux will be zero there; thus,

$$\frac{\partial w_{\text{Si}}^{\text{core}}}{\partial z} = 0 \quad \text{at } z = 0. \quad (3)$$

On the other hand, at  $z = z_{\text{int}}(t)$ , where  $z_{\text{int}}(t)$  is the location of the core/clad interface at time  $t$ , the silicon content in the core is taken to be equal to the equilibrium solvus of the (Al) solid solution,  $w_{\text{Si}}^{(\text{Al})/\text{Si}}$ , for temperatures lower than the eutectic Al–Si ( $T_{\text{eut}} = 577 \text{ }^\circ\text{C}$ ), and to the equilibrium solidus,  $w_{\text{Si}}^{(\text{Al})/\text{liquid}}$ , above; so,

$$w_{\text{Si}}^{\text{core}} = w_{\text{Si}}^{(\text{Al})/\text{Si}} \quad \text{or } w_{\text{Si}}^{\text{core}} = w_{\text{Si}}^{(\text{Al})/\text{liquid}} \quad (4)$$

at  $z = z_{\text{int}}(t)$  depending on temperature

with the following data from the Al–Si diagram:

$$w_{\text{Si}}^{(\text{Al})/\text{Si}} = 1.6034 \cdot 10^{-14} \cdot (T)^{5.0773},$$

wt.%, for  $300 \text{ }^\circ\text{C} < T < 500 \text{ }^\circ\text{C}$

$$w_{\text{Si}}^{(\text{Al})/\text{Si}} = 2.0749 \cdot 10^{-13} \cdot (T)^{4.6624},$$

wt.%, for  $500 \text{ }^\circ\text{C} < T < 577 \text{ }^\circ\text{C}$

$$w_{\text{Si}}^{(\text{Al})/\text{liquid}} = k_{\text{Si}} \cdot w_{\text{Si}}^{\text{liquid}/(\text{Al})}$$

$$= k_{\text{Si}} \cdot \{12.5 - 0.1501 \cdot (T - 577)\}$$

for  $T > 577 \text{ }^\circ\text{C}$

where  $k_{\text{Si}}$  is the silicon partition coefficient,  $T$  the temperature in Celsius, and  $w_{\text{Si}}^{(\text{Al})/\text{liquid}}$  the (Al) liquidus composition. Also, the silicon redistribution at the core/clad interface should obey the following general equation:

$$(w_{\text{Si}}^{\text{clad}} - w_{\text{Si}}^{\text{core}}) \cdot \frac{dz_{\text{int}}}{dt} = D_{\text{Si}}^{\text{core}} \cdot \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial z} - D_{\text{Si}}^{\text{clad}} \cdot \frac{\partial w_{\text{Si}}^{\text{clad}}}{\partial z} \quad (5)$$

at  $z = z_{\text{int}}(t)$ .

However, in what follows and in agreement with the description in the first section, we will assume very fast lateral homogenization and homogeneous evolution of the clad, which allows us to set the second term of the right-hand side of (5) to zero and to set the composition of the remaining clad  $w_{\text{Si}}^{\text{clad}}$  constant and equal to its initial value  $w_{\text{Si}}^{\text{clad},0}$ . Other initial conditions are that, the composition of the core is homogeneous, taking the value  $w_{\text{Si}}^{\text{core},0}$ , and that the initial interface position is  $z_0$ .

### 2.2 Transformed Model Equations

To solve the above set of equations numerically, it is convenient to employ boundary immobilization using a Landau transformation [4] by defining the variable

$$\tau = t, \quad \eta = \frac{z}{z_{\text{int}}(t)}. \quad (6)$$

Thus, on using the chain rule, we have

$$\frac{\partial w_{\text{Si}}^{\text{core}}}{\partial t} = \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \tau} - \frac{\eta \cdot \dot{z}_{\text{int}}(\tau)}{z_{\text{int}}(\tau)} \cdot \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \eta} \quad \text{and}$$

$$\frac{\partial^2 w_{\text{Si}}^{\text{core}}}{\partial z^2} = \frac{1}{z_{\text{int}}^2(\tau)} \cdot \frac{\partial^2 w_{\text{Si}}^{\text{core}}}{\partial \eta^2},$$

so that Eq. (1) becomes, for  $0 \leq \eta \leq 1$ ,

$$z_{\text{int}}^2(\tau) \cdot \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \tau} - \eta \cdot z_{\text{int}}(\tau) \cdot \dot{z}_{\text{int}}(\tau) \cdot \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \eta}$$

$$= D_{\text{Si}}^{\text{core}} \cdot \frac{\partial^2 w_{\text{Si}}^{\text{core}}}{\partial \eta^2}, \quad (7)$$

where the dot denotes differentiation with respect to  $\tau$ , subject to the boundary conditions

$$\frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \eta} = 0 \quad \text{at } \eta = 0 \quad (8)$$

$$(w_{\text{Si}}^{\text{clad}} - w_{\text{Si}}^{\text{core}}) \cdot z_{\text{int}}(\tau) \cdot \frac{dz_{\text{int}}}{d\tau} = D_{\text{Si}}^{\text{core}} \cdot \frac{\partial w_{\text{Si}}^{\text{core}}}{\partial \eta} \quad \text{at } \eta = 1 \quad (9)$$

$$w_{\text{Si}}^{\text{core}} = w_{\text{Si}}^{(\text{Al})/\text{Si}} \quad \text{or} \quad w_{\text{Si}}^{\text{core}} = w_{\text{Si}}^{(\text{Al})/\text{liquid}} \quad \text{at } \eta = 1, \quad (10)$$

depending on temperature

and the initial conditions at  $\tau = 0$

$$w_{\text{Si}}^{\text{core}} = w_{\text{Si}}^{\text{core},0}, \quad 0 \leq \eta \leq 1 \quad (11)$$

$$z_{\text{int}} = z_0. \quad (12)$$

### 2.3 Numerical Implementation

Equations (6)–(12) were solved using the finite element software COMSOL Multiphysics. The governing equations were discretized using Lagrangian quadratic elements, and the convergence criterion at each time step was taken as

$$\frac{1}{N_{\text{dof}}} \cdot \sum_{i=1}^{N_{\text{dof}}} \left( \frac{|E_i|}{A_i + \zeta \cdot |U_i|} \right)^2 < 1, \quad (13)$$

where  $N_{\text{dof}}$  is the number of degrees of freedom at each time step,  $(U_i)$  is the solution vector corresponding to the solution at a certain time step,  $E_i$  is the estimated error in the latest approximation to the  $i$ th component of the true solution vector,  $A_i$  is the absolute tolerance for the  $i$ th degree of freedom, and  $\zeta$  is the relative tolerance; for the computation,  $\zeta = 0.01$  and  $A_i = 0.001$  for  $i = 1, \dots, N_{\text{dof}}$ . Also, a uniform mesh was used and a mesh independence check was performed; for the results shown, a mesh consisting of 240 elements, corresponding to  $N_{\text{dof}} = 481$ , was employed.

## 3 Results

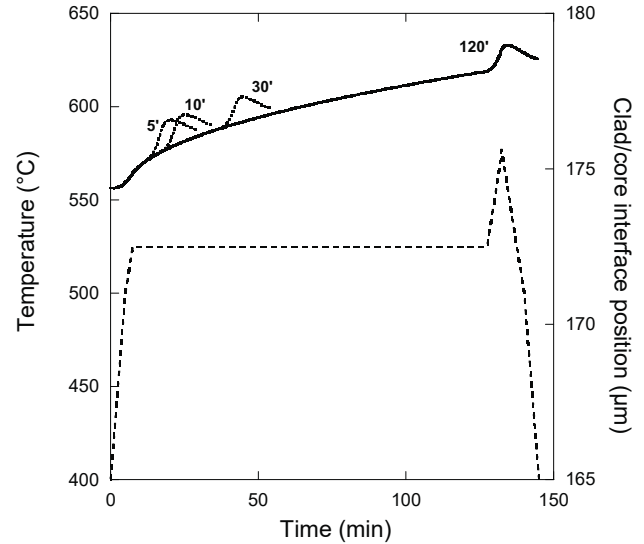
Terrill [1] and, more recently, Gao et al. [5] have studied quantitatively the solid-state development of the depleted zone during isothermal holding of brazing sheets at various temperatures below  $T_{\text{eut}}$ . Both works used ad hoc estimates of the silicon diffusion coefficient so as to obtain a good agreement between their analytical estimates and experimental values. The intention in the present study is rather to use the above-mentioned assessed value of the diffusion coefficient in order to be able to draw conclusions on the appropriateness of the assumptions made.

Quite recently, Benoit et al. [6] carried out a metallographic estimate of the thickness of residual clad after holding the brazing sheet at 525–550 °C for up to 120 min. At the end of the holding period, the samples were heated to 574 °C—which corresponds to their experimental onset temperature for melting of the clad—and then cooled to room temperature. The heating and cooling rates were 20 K/min below 500 °C and 10 K/min above. The clad alloy contained 9–11 wt.%Si (set to 10 wt.% for the

present calculations), and the core less than 0.3 wt.% Si (set to 0.3 wt.%). By metallographic inspection, the initial clad thickness was found to be 26.79  $\mu\text{m}$  for an overall sheet thickness of 201.17  $\mu\text{m}$ .

In Fig. 2 is shown with a dashed line the evolution of temperature during the thermal cycle for the case of holding at 525 °C for 120 min which includes heating before holding, holding, heating to 574 °C and final cooling. Calculations start from 400 °C as no effective diffusion is expected at lower temperatures. The solid line represents the change in the position of the core/clad interface during this cycle. It is seen that the interface does not move as long as the temperature is lower than about 450 °C during heating to the holding stage. Then, it starts moving at an increasing rate until the sharp difference in silicon content between the clad and the core is replaced by a smooth gradient zone. During further holding, the clad dissolves at a slowly decreasing rate. During the heating at the end of holding, the interface moves further quite rapidly, but then recedes during the final cooling stage nearly back to the position it had at the end of the holding. Fig. 2 also shows the calculations corresponding to shorter holding times as carried out by Benoit et al. [6], namely 5, 10 and 30 min long. The change in position at the end of holding shows the same characteristics as for the 120-min holding.

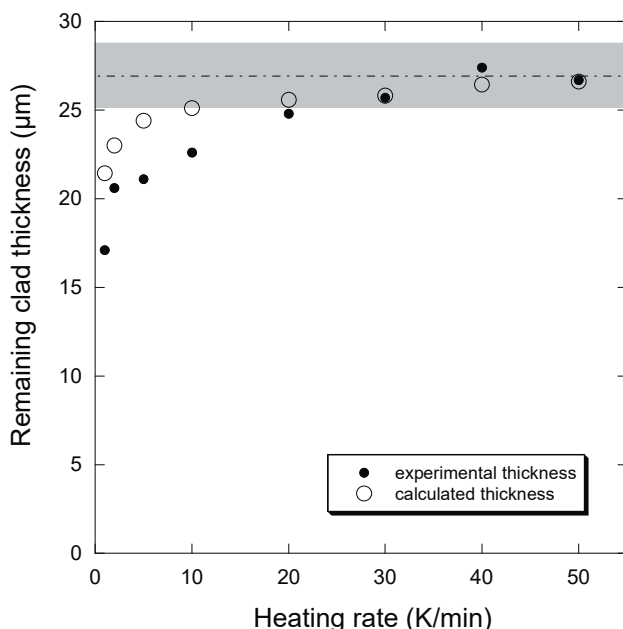
To complement their investigation on the effect of solid-state diffusion on the amount of available clad at the brazing temperature, Benoit et al. [6] also performed



**Fig. 2** Numerical simulation of experiments by Benoit et al. [6]: temperature–time cycle (dashed line) and calculated displacement of the core/clad interface position (solid line) during holding at 525 °C for 120 min. Dotted lines show the calculated evolution of the interface position for holding for 5, 10 and 30 min, as indicated on the curves

heating to 574 °C at rates from 1 K/min to 50 K/min. The remaining clad thickness was estimated by metallographic evaluation, and the results are reported in Fig. 3 with solid dots, while the values calculated with the present approach are reported with open circles. The predicted values are significantly larger than the experimental ones at low heating rates. However, the same conclusion as drawn by Benoit et al. [6] can be made, namely that a minimum heating rate of 10–20 K/min will be necessary to avoid consumption of the clad during the heating stage of the brazing process.

In addition to clad thickness measurements, comparison of calculated and experimental silicon profiles in the core could possibly give some support to the present numerical analysis. In the literature, two studies were found reporting silicon profiles after one thermal cycle below the eutectic temperature of the clad [5, 6]. It was decided to use the study by Gao et al. [5] who heated a coupon rapidly to 520 °C, held it for 5 min at this temperature and finally cooled it quickly to room temperature. Based on the experimental thermal records reported by Gao et al. [5], a heating rate of 50 K/min was considered. The composition of the clad alloy was given as 7.192 wt.% Si, and its thickness was evaluated to be 26 µm by means of the reported Mn profile. Finally, from the Si profile, the original Si content in the core was set to 0.07 wt.% and the core thickness at 200 µm, i.e., large enough so that its exact



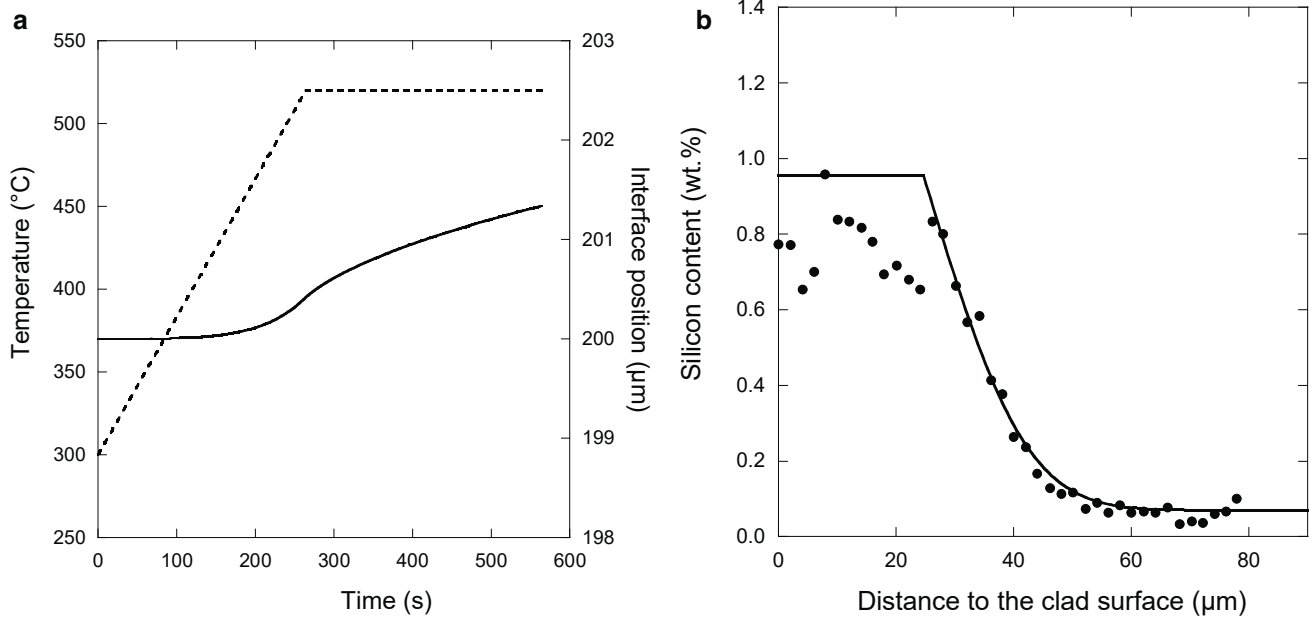
**Fig. 3** Comparison of experimental values from Benoit et al. [6] and calculated values of the remaining clad thickness after heating to 574 °C at various rates, from 1 to 50 K/min. The dot-dashed line is the average original thickness, and the grey area shows the possible deviation around this value as indicated by Benoit et al. [6]

value does not have importance in the present case. Figure 4a shows the simulated temperature profile and the calculated change in the core/clad interface position with time. It is seen that the interface starts moving above 400 °C at a rate that increases with temperature and then decreases during holding, as already seen in Fig. 2. The whole change in position of the interface is small at 1.4 µm.

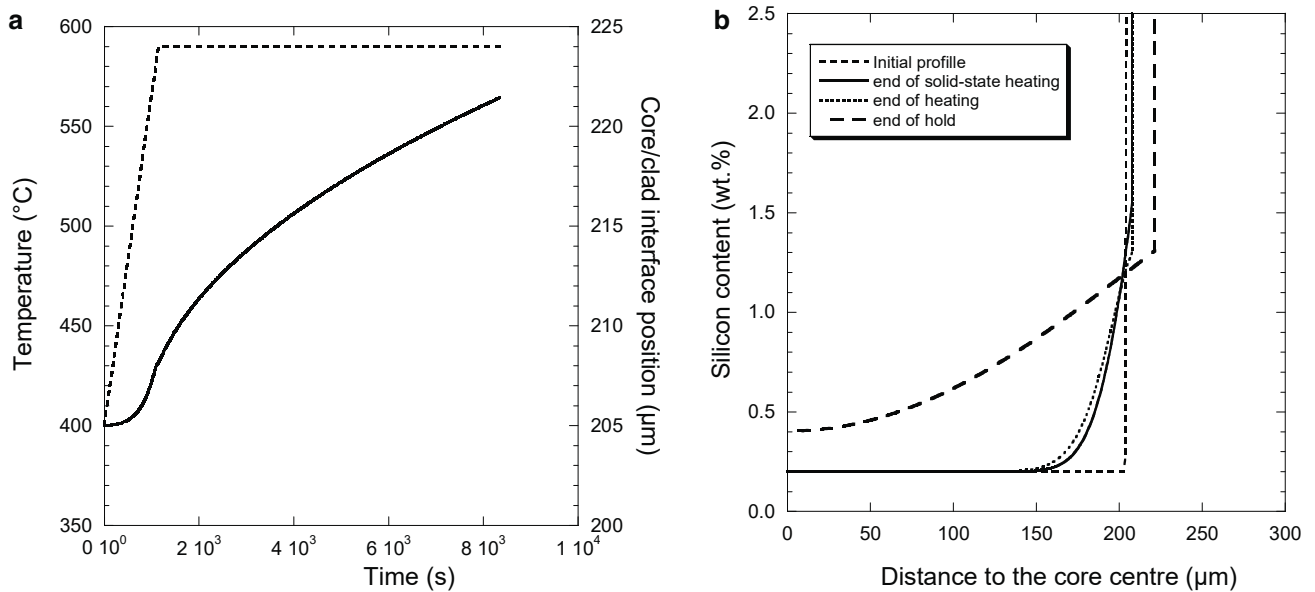
In Fig. 4b, the calculated silicon profile at the end of holding is compared with the experimental one, whose values have been picked up from the figure in the referred work [5]. As the measurements are taken avoiding silicon precipitates, the calculated value reported in the clad is the solubility value of silicon  $w_{\text{Si}}^{(\text{Al})/\text{Si}}$  in (Al) at 520 °C, which appears as the horizontal line. That the experimental values in the clad are lower than those indicated by this line is certainly due to precipitation of silicon onto the coarse pre-existing silicon precipitates during final cooling. It is observed that the profile in the core can be perfectly reproduced and that the far-field silicon value is the original core silicon content. Indeed, the thickness of the diffusion profile is limited to about 35 µm, i.e., far below the 200 µm core thickness considered for calculations. The excellent agreement that is obtained suggests that, in the core, precipitation of silicon during cooling to room temperature occurs as fine precipitates within (Al) and is thus taken into account during microanalysis.

The final step in the present work is to consider the evolution of a brazing sheet during holding at a temperature within the melting interval of the clad. Both Turriff et al. [7] and Yiyu Tu et al. [8] used DSC experiments to evaluate the decrease in liquid clad available during a brazing process at temperatures above the Al–Si eutectic, but below the clad liquidus. Both studies have arrived at the same conclusion which has been illustrated with the results from Turriff et al. [7]. The brazing sheet is 510 µm in thickness with a clad of 51.4 µm on each side. The silicon content is 0.2 wt.% in the core and 7.36 wt.% in the clad. The brazing cycle consists of heating at 10 K/min, then holding at 590 °C for up to 120 min and finally cooling at 10 K/min to room temperature.

Figure 5a shows the time evolution of the temperature (dotted line) and of the core/clad interface position (solid line) until the end of the hold, while Fig. 5b presents the silicon profile at various times: (1) before the experiment (initial profile); (2) at the end of solid-state heating (577 °C); (3) at the end of heating (590 °C); and (4) at the end of the hold. In Fig. 5a, it is seen that the clad layer thickness decreases steadily during holding, which is due to the large thickness of the core that maintains a finite silicon gradient. This is illustrated in Fig. 5b, where it is seen that, even after 120-min holding, the change in silicon content at the core centre is still quite limited.



**Fig. 4** Simulation of the evolution of a core/clad assembly during heating to 520 °C and holding at that temperature for 5 min. **a** Temperature record (dashed line) and change in position of the core/clad interface (solid line); **b** calculated silicon profile (solid line) plotted from the clad surface with dots representing the experimental values reported by Gao et al. [5]

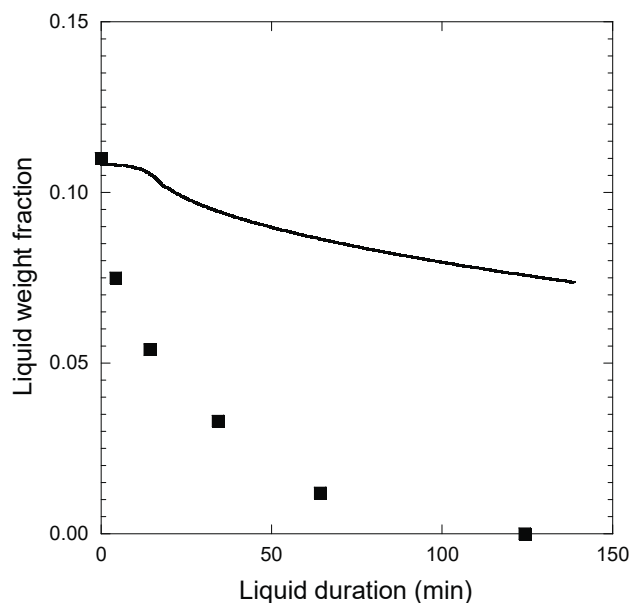


**Fig. 5** Temperature–time profile and time evolution of the core/clad interface position (a); silicon profiles at various times during the brazing process, see text (b). Note that the silicon content in the clad is at 7.361 wt.%

During cooling at the end of the hold, re-solidification of (Al) proceeds immediately from the core/clad interface, leading to the formation of cells or dendrites and to the development of microsegregation. When the Al–Si eutectic temperature is reached, the remaining liquid solidifies quite rapidly, as seen on the experimental DSC records [7, 8].

The amount of eutectic may be evaluated applying Scheil’s law for the liquid present at the brazing temperature. Using the phase diagram data listed above, the lever rule predicts a liquid fraction at 590 °C of 0.655, from which the Scheil model leads to a eutectic fraction of 0.54 at the eutectic temperature. Following the calculation made by Turriff





**Fig. 6** Comparison of calculated (solid line) and experimental [7] amounts of liquid available after various holding times at brazing temperature

et al. [7], the amount of liquid after holding is expressed as a fraction of the brazing sheet thickness, i.e., it is given by  $0.54 \cdot (z_{\text{tot}} - z_{\text{int}}) / z_{\text{tot}}$ , where  $z_{\text{tot}}$  is the overall brazing sheet thickness. The values thus calculated are compared in Fig. 6 with the evaluation made by Turriff et al. [7] from their DSC experiments.

It is seen that there is a huge discrepancy between prediction and experiments, which is thought to be due to two reasons: Firstly, DSC underestimates the remaining clad thickness, as can be seen from the micrographs in the original papers. At 120 min, when DSC gives no remaining liquid clad, the microstructure still presents some eutectic silicon [7]. In agreement with Kuntz [9], Benoit et al. [10] have noticed that DSC results are not quantitatively reliable, although they may show the right trends. Secondly, similarly using DSC, Tu et al. [8] have shown convincingly

that the grain size in the core may have a dramatic effect on the decrease in residual clad thickness when the latter is liquid. A possible mechanism for this effect is preferential diffusion of silicon and liquid penetration along the grain boundaries in the core, which may greatly enhance the decrease in available liquid [8] as it happens in TLP bonding [2].

## 4 Conclusion

A simple 1D numerical model for brazing was applied to the CAB process of aluminium alloys. It was found that solid-state diffusion of silicon in the core might be accurately described. On the other hand, the strong interaction between the liquid clad and the solid part of the brazing sheet needed further consideration for a proper prediction of the remaining clad at brazing temperature.

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