JOURNAL OF MATERIALS SCIENCE 32 (1997) 719–725

Interface study for stainless steel fibre-reinforced aluminium matrix composite

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Regular interface zones with uniform thickness in AISI 304 stainless steel-reinforced aluminium-matrix composite have been obtained using a vacuum high-pressure diffusion-bonding technique. Extensive and intensive experiments were performed to examine the growth of interfacial compounds with the variation of hot-pressing time and temperature. In the initial stage, the overall growth rate of the interface was found to follow a parabolic law. After a certain diffusion time, the interface growth rate fell behind that predicted by the parabolic law. A modified parabolic law has been established to explain the deviation and proved to be a better model to fit the experimental data. An activation energy of 152 kJ mol^{-1} was found, which was somewhat lower than that obtained by previous work. The lower value of activation energy is attributed to the pressure (70 MPa) applied during hot pressing. Energy dispersive spectroscopic analysis and microhardness measurement indicated that the interface zone consists of a mixture of intermetallic compounds Fe(Cr, Ni)Al_2, Fe(Cr, Ni)Al_3 and (Fe, Cr, Ni)$_2Al_7$.

1. Introduction

The nature of the fibre/matrix interface is one of the key factors which determine the unique properties of the fibre-reinforced metal-matrix composites (MMCs). In general, owing to its brittle nature, the presence of an interfacial compound in MMCs degrades the composite properties. Interfacial reactions may take place if the composite is exposed to high temperature for some time. Most composite fabrication methods, such as liquid-metal infiltration [1], squeeze casting [2], investment casting [3], powder metallurgy [4], hot isostatic pressing [5] and diffusion bonding [6], involve a high-temperature process. The applications of MMCs in industry and defence sectors may also require their exposure to high-temperature environments [5]. Therefore, appropriate control of the reactions at the fibre/matrix interface and an understanding of the reaction mechanisms and growth kinetics have become very important in the fabrication of MMCs.

The examination of interface reactions in stainless-steel fibre(SSF)-reinforced aluminium composite (SSF/Al) has been carried out [7–12]. In 1974, Pattnaik and Lawley [7] published a paper on the manufacturing of 355 SSF/Al composite using a vacuum hot-pressing method. They found that the composite degradation after elevated-temperature exposure was due to the formation of irregular, ternary (Fe, Cr)Al intermetallic compound at the interface. In 1983, Pai *et al.* [8] fabricated 304 SSF/Al–10%Mg composite using the vacuum infiltration technique. They found that an irregular layer of interfacial compound was formed during heat treatment at 550 °C in the atmosphere for 24 h or longer. No growth kinetics and formation mechanisms for SSF/Al composites were presented until the publications of papers by Bhagat [9–12], in which 304 SSF/1100 Al composites with various volume fractions SSF were fabricated using P/M hot pressing, squeeze-casting and infiltration techniques. Bhagat [10–12] also observed that the interface layer developed on 304 SSF/Al was irregular. However, he was able to determine that the interface growth kinetics is diffusion controlled, and to calculate the diffusion-controlled parabolic rate constant [12]. The interface in the hot-pressed composites was characterized to contain FeAl, Fe_2Al_5 and $NiAl_3$ intermetallic compounds [12].

In this study we were able to produce a regular interface layer with uniform thickness in SSF/Al composite using a vacuum high-pressure diffusion-bonding technique. Extensive and intensive experiments were carried out to examine the growth of interfacial compound with the variation of hot-pressing time and temperature. Besides the determination of growth kinetics and parabolic rate constant, efforts were also made to calculate the activation energy of the

SSF/Al composite interface reaction, which was not previously determined. Process and heat-treatment parameters were varied to determine their effects on the growth rate and geometry of the interface. Energy dispersive spectroscopic (EDS) analysis and microhardness measurements were also performed to characterize the interfacial compounds.

2. Experimental procedure

Commercial 1100 aluminium foil with a thickness of 100 μm was used for the matrix. AISI 304 stainless steel fibre (SSF) with a diameter of 150 μm was employed as the reinforcement. The vacuum hot-pressing diffusion-bonding technique was carried out to fabricate the composite. Details of the manufacturing process were reported elsewhere [13]. Briefly, alternating layers of SSF and aluminium foils were stacked on a flat die equipped in a winding machine, followed by hot-pressing in a vacuum furnace under a pressure of 70 MPa at various elevated temperatures for various times. In order to avoid the touching and interference of interfacial compounds between neighbouring fibres during interface growth, the volume fraction of SSF was reduced from 44% [13] to about 15%.

In order to examine the growth rate of the SSF/Al interface, the hot-pressing temperature was kept at 600 °C and the hot-pressing time was varied from 15 min to 32 h. From the growth rate one may determine the growth kinetics of the interface. Different hot-pressing temperatures (550 and 575 °C) were also employed with variation of hot-pressing time from 1–16 h for the determination of the interface reaction activation energy. In order to investigate the effects of process parameters (time, temperature, pressure and atmosphere) on the morphology of the interface, some composites were hot-pressed at lower temperatures (500 and 525 °C), heat treated in air or vacuum, with or without the application of pressure, for various times.

Characterization of the SSF/Al interface was performed using optical microscopy (OM), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS) and Vickers' microhardness (HV) measurement.

3. Results and discussion

3.1. Determination of growth kinetics

A uniform and ringed interface was observed in composites hot-pressed at 600 °C under 70 MPa for various times; as shown in Fig. 1. The interface advanced both inwards and outwards with time. Inward growth dominated at the beginning of the reaction (Fig. 1a). After some time, outward growth became more pronounced (Fig. 1b–1d). To analyse the interface growth mechanism, the thickness, h, of interface was plotted against the square root of hot-pressing time, $t^{1/2}$, as shown in Fig. 2a. The plot starts with a straight line up to 4 h pressing time, followed by a curve representing a slower growth rate. The straight line indicates that the growth of the interface follows a parabolic law ($h^2 = kt$) which is a result of the diffusion-control mechanism [14]. k is temperature dependent and

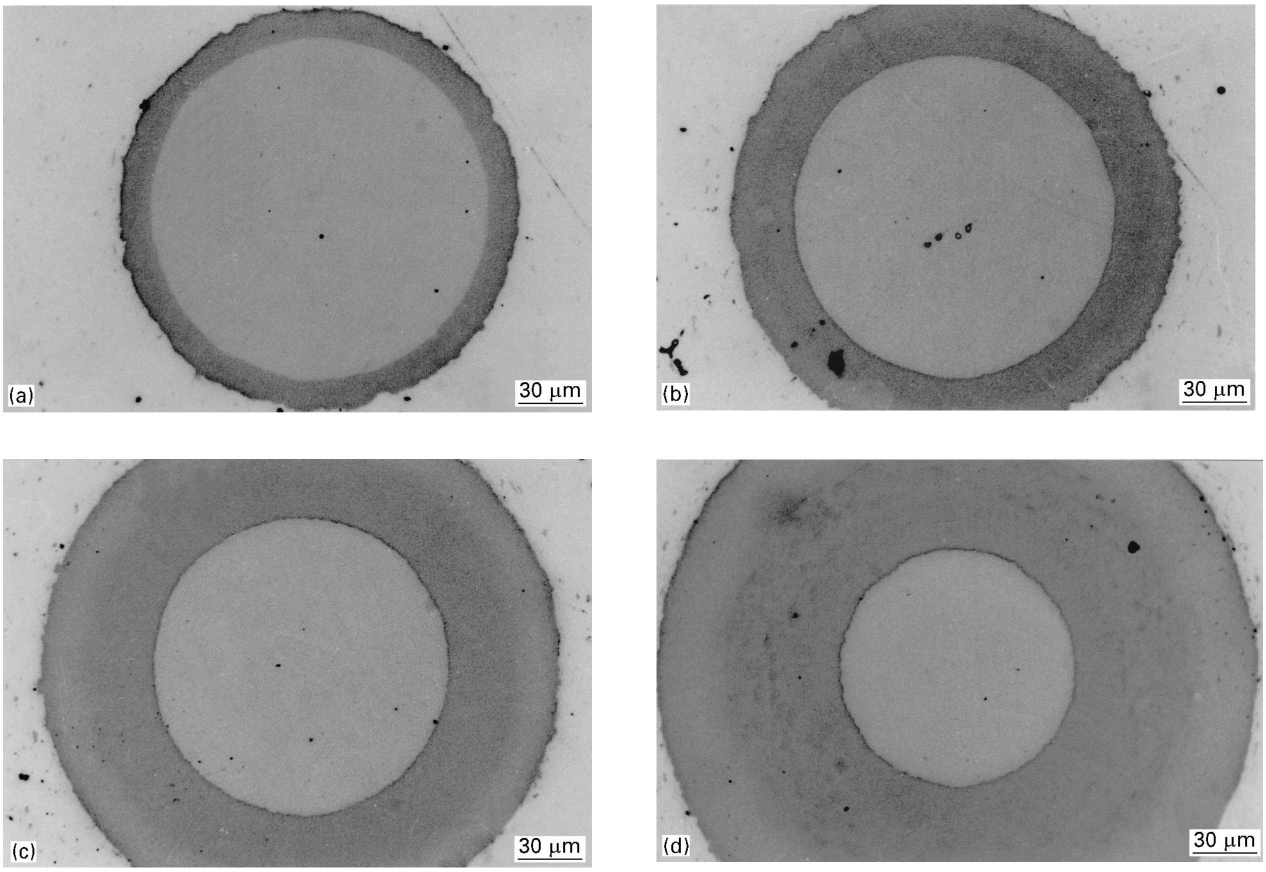

Figure 1 Optical micrographs for the composite hot-pressed at 600 °C, 70 MPa for (a) 15 min, (b) 1 h, (c) 4 h, and (d) 16 h.

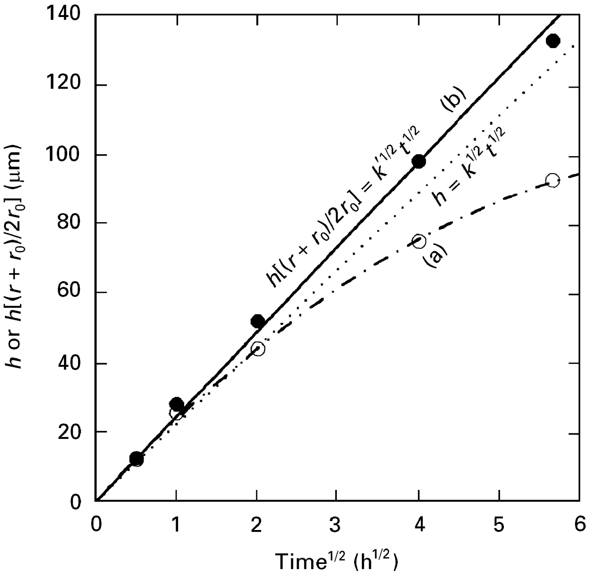

Figure 2 Variation of (a) (○) interface thickness, h, with $(\text{time})^{1/2}$, and (b) (●) the product of thickness, h, and $(r + r_0)/2r_0$ versus $(\text{time})^{1/2}$ for composites hot-pressed at 600 °C.

called the parabolic rate constant. The value of k can be determined from the slope of the straight line ($k^{1/2} = \text{slope}$) in Fig. 2a. It was found in this study from the straight-line portion of Fig. 2a that $k = 1.40 \times 10^{-13}\ \text{m}^2\text{s}^{-1}$ at 873 K (600 °C). The value of the parabolic rate constant k, for the SS/Al interface has been examined previously [12, 15]. Mannan *et al.* [15] investigated the diffusion zone growth in AISI 316 stainless steel–aluminium couples in the temperature range 763–913 K. They found that the growth of diffusion zone obeys a parabolic law. The parabolic rate constant, k, at 873 K was determined to be $0.7 \times 10^{-13}\ \text{m}^2\text{s}^{-1}$ which is only half of the value obtained in this study. It is worthwhile to note that Mannan *et al.* [15] carried out their study in vacuum without the application of pressure, while our research was performed in a vacuum with a pressure of 70 MPa. Bhagat [12] examined the interface growth in 304 SSF/Al composite fabricated by the P/M hot-pressing technique and found that the interface growth follows a parabolic law in Region II (see Bhagat [12]) at 800 K. The parabolic rate constant at 800 K in Region II was determined to be $3.6 \times 10^{-13}\ \text{m}^2\text{s}^{-1}$ which is more than twice the value obtained in this study. It should be pointed out that the interface growth examined by Bhagat [12] took place under much higher pressure (140 MPa). Comparison of our results with those of Mannan *et al.* [15] and Bhagat [12] clearly indicates that the pressure applied enhances the interface growth rate significantly.

As shown in Fig. 2a, the data points at $t > 4$ h fall below the extended straight line, indicating that the interface growth rate is slower than that predicted by the diffusion-control parabolic law. Bhagat [12] also found a sluggish interface growth after the interface grew to a thickness of about 20 μm and termed this Region III. However, the reason why the presence of a large thickness of interface would slow down the diffusion process has never been explained. In trying to understand the sluggish growth of interface in SSF/Al composite after the interface grows to some thickness, one should bear in mind that the parabolic growth rate ($h^2 = kt$) results from the assumption of planar diffusion of a single phase with a semi-infinite geometry [15] as shown in Fig. 3a. In contrast, radial diffusion takes place at the interface in SSF/Al composite, as shown in Fig. 3b. In planar diffusion, the length of the diffusion front is maintained constant (i.e. l, in Fig. 3a); however, in radial diffusion, the length of diffusion front (i.e. $2\pi r$ in Fig. 3b) continues to increase with diffusion time if the interface grows outwards. The variation of interface thickness in radial diffusion with respect to diffusion time can be obtained by the following simplified model.

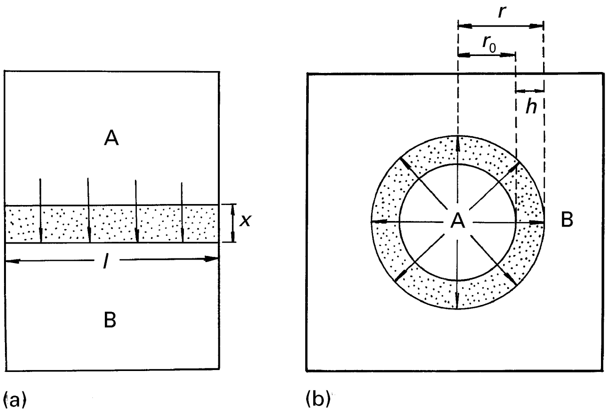

Figure 3 Schematic diagram for (a) planar diffusion and (b) radial diffusion.

To simplify the approach, it is assumed that the diffusion is of single phase, unlimited diffusion source (element A) and unidirectional (a) or radially outward (b) from A to B as shown in Fig. 3. It is also assumed that at the beginning of diffusion ($t = 0$), the length of the diffusion front for planar diffusion, is equal to that of radial diffusion, that is $l = 2\pi r_0$. Because the length of the diffusion front in both cases is equal, the amount of atom A diffused through A/B interface should also be equal at any elapsed time. If the A/B interface reaction is the same for planar and radial geometry, the same amount of diffused atom will occupy the same amount of area. That is

$$lx = \pi(r^2 - r_0^2) \tag{1a}$$

where x is the interface thickness for planar diffusion (Fig. 3a), r is the outer radius of the interface for radial diffusion (Fig. 3b) and $r - r_0 = h$, where h is the interface thickness for radial diffusion. Because $l = 2\pi r_0$, Equation 1a becomes

$$2\pi r_0 x = \pi(r^2 - r_0^2) \tag{1b}$$

$$2r_0 x = (r - r_0)(r + r_0) = h(r + r_0) \tag{1c}$$

since $r - r_0 = h$, therefore

$$h(r + r_0) = 2r_0(kt)^{1/2} \tag{2a}$$

since $x^2 = kt$, therefore

$$h[(r + r_0)/2r_0] = k^{1/2}t^{1/2} \tag{2b}$$

or

$$[h(r + r_0)/2r_0]^2 = kt \tag{2c}$$

To check the validity of Equation 2b or c, $h[(r + r_0)/2r_0]$ was plotted against $t^{1/2}$ as shown in Fig. 2b. The well-fitted straight line from $t = 15$ min to $t = 16$ h indicates that Equation 2b or c is a much better model which describes the interface growth rate in radial diffusion for fibre-reinforced composites than conventional parabolic law $x^2 = kt$. The deviation of the data point at $t = 32$ h ($t^{1/2} = 5.66\ \mathrm{h}^{1/2}$) is expected, because after the long diffusion time in this study, region A (unreacted SS) becomes significantly smaller and the area of interfacial compound becomes very large; therefore, the assumption of unlimited diffusion source is no longer valid. In addition, the inward diffusion (from B to A) and multiple-phase reaction in this study also complicate the analysis. The parabolic rate constant (denoted by k'), obtained from the plot $h[(r + r_0)/2r_0]$ versus $t^{1/2}$ should be more accurate than the constant (denoted by k) obtained from the plot h versus $t^{1/2}$. The parabolic rate constant, k', obtained from Fig. 2b is $1.70 \times 10^{-13}\ \mathrm{m^2 s^{-1}}$.

3.2. Activation energy

In order to determine the activation energy of the interface reaction, vacuum hot-pressing was also carried out at 550 and 575 °C . Fig. 4 shows the interface growth at 550 and 575 °C under 70 MPa from 1–16 h. Although some sawtooth appearance occurred at the outer surface of the interface in the composite hot-pressed at 550 °C, the ringed interface is, in general, uniform and the thickness can be measured with sufficient accuracy. The plots of h versus $t^{1/2}$ and $h[(r + r_0)/2r_0]$ versus $t^{1/2}$ for the composite hot-pressed at 550 and 575 °C, respectively, are shown in Fig. 5. The parabolic rate constants obtained

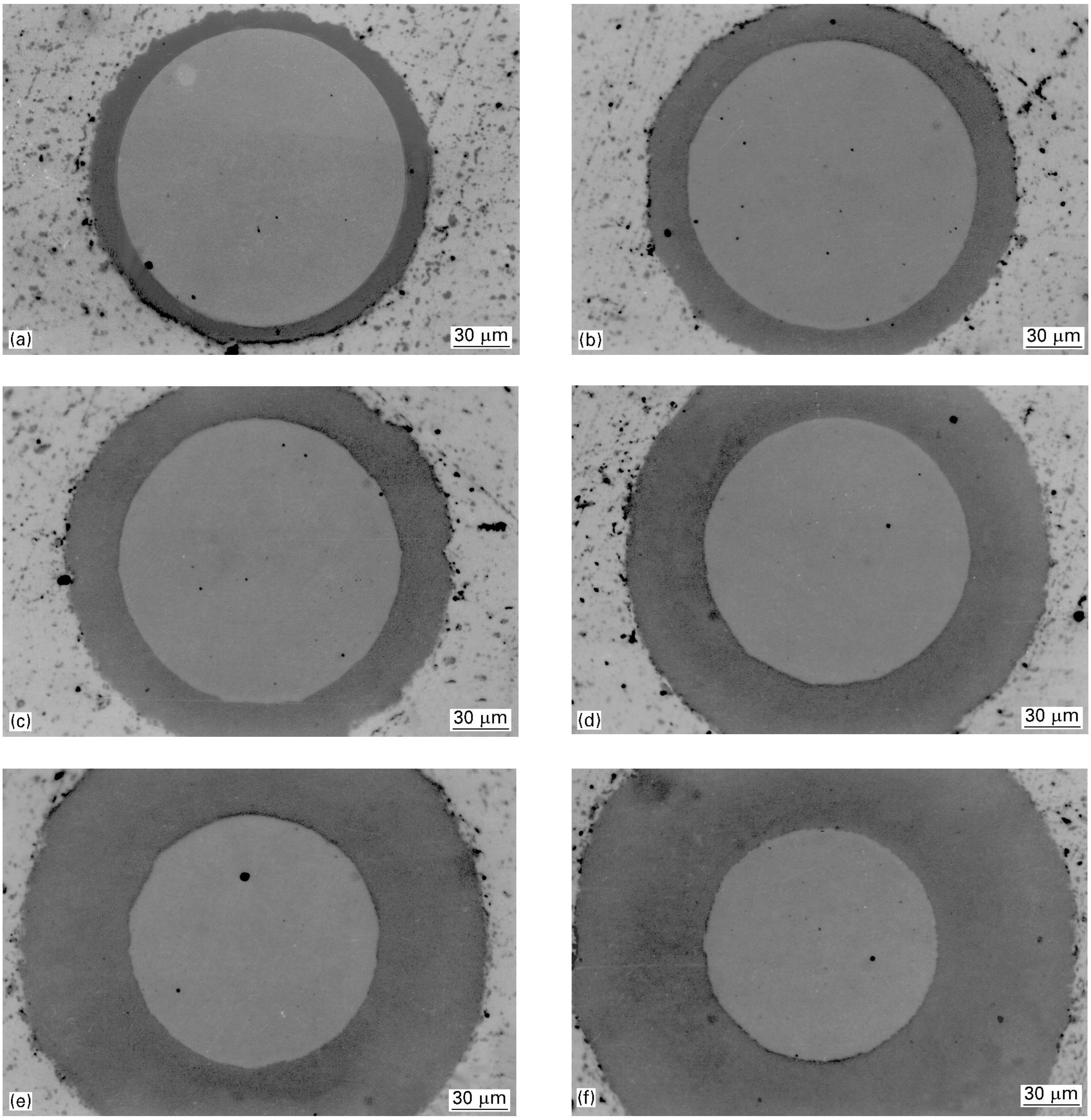

Figure 4 Optical micrographs of the composite hot-pressed under 70 MPa for (a, b) 1, (c, d) 4, and (e, f) 16 h at (a, c, e) 550 °C and (b, d, f) 575 °C.

from Fig. 5 are $k = 0.40 \times 10^{-13}\,\mathrm{m^2 s^{-1}}$ and $k' = 0.51 \times 10^{-13}\,\mathrm{m^2 s^{-1}}$ for the composite hot-pressed at 550 °C, and $k = 0.90 \times 10^{-13}\,\mathrm{m^2 s^{-1}}$ and $k' = 1.19 \times 10^{-13}\,\mathrm{m^2 s^{-1}}$ for the composite hot-pressed at 575 °C. It is assumed that the temperature dependence of the reaction parameter k (and also k') follows an Arrhenius-type behaviour, that is

$$k = k_0 \exp(-Q/RT) \tag{3a}$$

or

$$\ln k = \ln k_0 - Q/RT \tag{3b}$$

where Q is the activation energy of the reaction, R is the gas constant (8.314 J mol^{-1} K^{-1}) and T is the hot-pressing temperature (K). The activation energy, Q, can be determined from the plot of $\ln k$ (or $\ln k'$) versus $1/T$ as shown in Fig. 6. The plot of $\ln k$ versus $1/T$ and $\ln k'$ versus $1/T$ can be approximated as straight lines and the activation energy obtained from the slope of the straight line is $Q = 152\,\mathrm{kJ\,mol^{-1}}$. The constants k_0 and k'_0 determined by inserting the value of Q into Equation 3a are $k_0 = 2.1 \times 10^{-4}\,\mathrm{m^2 s^{-1}}$ and $k'_0 = 2.8 \times 10^{-4}\,\mathrm{m^2 s^{-1}}$. The values of Q and k_0 obtained by Mannan *et al.* [15] in their study of the interdiffusion between AISI 316 stainless steel and aluminium are 166 kJ mol^{-1} and $7 \times 10^{-4}\,\mathrm{m^2 s^{-1}}$, respectively. The values of Q and k_0 obtained in this study are of the same order of magnitude as those calculated by Mannan *et al.* The value of Q obtained in this study is about 8% lower than Mannan's Q value. The lower Q value can be attributed to the pressure (70 MPa) applied during hot-pressing in this study.

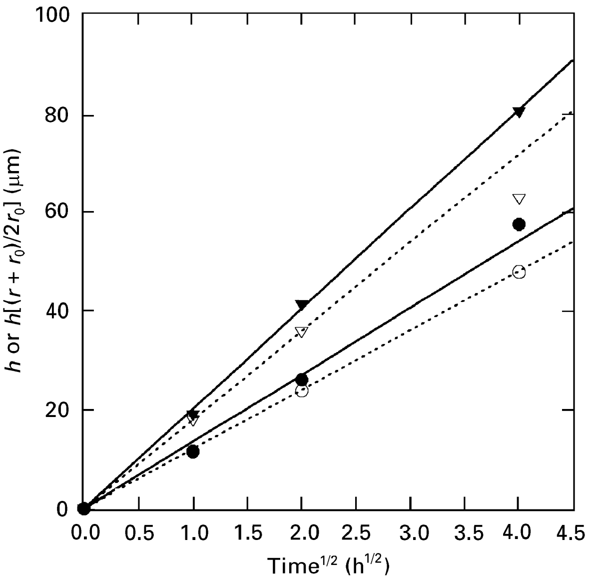

Figure 5 Variation of (○, ▽) interface thickness, h, and (●, ▼) its product with $(r + r_0)/2r_0$ versus (hot-pressing time)$^{1/2}$ for composites hot-pressed (○, ●) 550 °C and (▽, ▼) 575 °C.

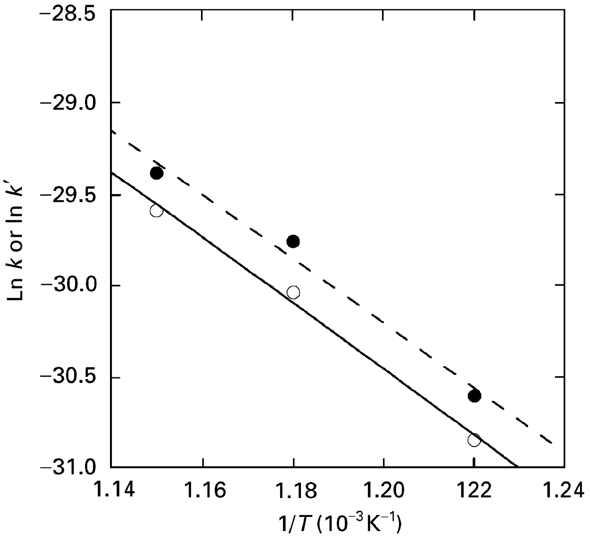

Figure 6 Temperature dependence of the parabolic rate constants (○) k and (●) k' for the SSF/Al interface reaction.

3.3. EDS and microhardness analysis of the interface

SSF/Al composite hot-pressed at 600 °C, 70 MPa for 32 h was selected for the EDS analysis and microhardness measurement on the interface and its vicinity, because the interface was sufficiently thick and very uniform (Fig. 7) such that precise analysis and measurement are possible. Three zones of different appearance were observed on the interface. As shown in Fig. 7, the inner zone consists of radial marks, the centre zone is mottled, and the outer zone is homogeneous. The variation of microhardness across the interface can be clearly seen from the indents in Fig. 7, resulting from the microhardness test. The change in hardness from the centre of the SSF through the interface to the aluminium matrix is shown in Fig. 8. The hardness at the interface is much higher than that in the SSF and aluminium matrix. The fact that hardness drops continuously from the inner zone to the outer zone of the interface indicates that multiple phases exist in the interface and the percentage of various phases changes gradually from the inner to the outer zone of the interface.

EDS analysis was performed at various points across the interface and the results are given in Table I. At a point very close to the remaining

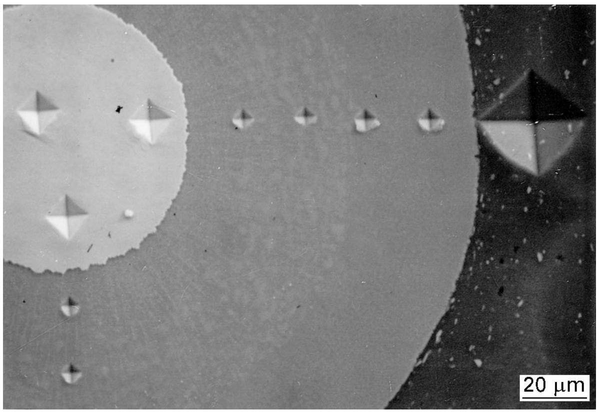

Figure 7 Optical micrograph (with microhardness indents) of the interface and its vicinity for the composite hot-pressed at 600 °C, 70 MPa for 32 h.

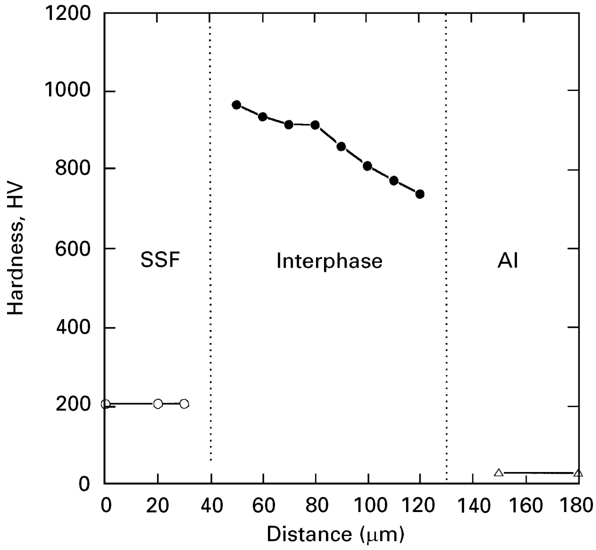

Figure 8 Variation of microhardness from the centre of the SSF to the aluminium matrix.

TABLE I EDS analysis (at %) across the interface of the composite hot-pressed at 600 °C, 70 MPa for 32 h. The first column gives the distance from the surface of the residual SSF

Position (μm)	Al	Fe	Cr	Ni
5	67.68	23.84	6.56	1.93
20	69.21	22.34	6.78	1.67
40	69.71	22.36	6.88	1.04
60	74.75	18.56	5.32	1.37
80	76.77	16.19	4.96	2.08
90	77.13	15.96	4.60	2.31

stainless steel fibre (5 μm away from the fibre), the aluminium content is 67.68 at %, indicating that the interfacial compound is mainly $\mathrm{Fe(Cr, Ni)Al_2}$. At a point very close to the aluminium matrix (90 μm away from the fibre), the aluminium content is 77.13 at %, indicating that the interfacial compound is mainly $\mathrm{(Fe, Cr, Ni)_2Al_7}$. The aluminium content increased gradually from 67.68 at % to 77.13 at % and, at the same time, the iron content decreased from 23.84 at % to 15.96 at % from the innermost zone to outermost zone of the interface. Multiple phases, including $\mathrm{Fe(Cr, Ni)Al_2}$, $\mathrm{Fe(Cr, Ni)Al_3}$ and $\mathrm{(Fe, Cr, Ni)_2Al_7}$, exist concurrently in the interface, and their percentage changes gradually from the inner to outer zone of the interface. These results are generally in agreement with those obtained by Mannan *et al.* [15] and Bhagat [12]. Because the aluminium and iron contents varied continuously with the position of the interface, various mixtures of different interfacial compounds, including $\mathrm{Al_6(Fe, Cr)}$, $\mathrm{Al_9(Fe, Cr)_2}$ predicted by Mannan *et al.* [15], and FeAl, $\mathrm{Fe_2Al_5}$, $\mathrm{NiAl_3}$ predicted by Bhagat [12], could not be excluded. Further study is required to determine the percentage of the individual intermetallic compounds at various positions of the interface.

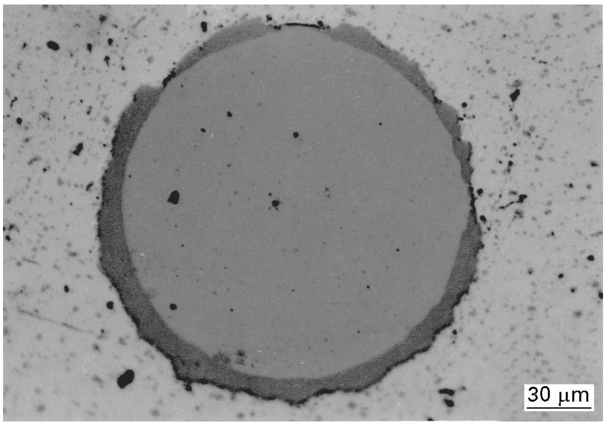

Figure 9 Optical micrograph for the composite vacuum hot-pressed at 525 °C, 70 MPa for 4 h.

3.4. Effects of process parameters on the morphology of the interface

The uniform formation of the interface makes accurate analysis of the reaction kinetics possible. Appropriate control of process parameters is essential to obtain a uniform and regular interface in fibre-reinforced MMCs. For SSF/Al composite, previous works [7–12] were not able to obtain an interface as uniform and regular as that obtained in this study, simply because the process parameters used were different from this study. In order to obtain detailed understanding of the effects of process parameters on the morphology of the interface, the following experiments were also carried out.

SSF/Al composite was vacuum hot-pressed at 525 °C, 70 MPa for 4 h. A clear interface was observed, as shown in Fig. 9; however, the ringed interface was not uniform and its thickness varied from position to position. Comparison of Figs 1, 4 and 9 indicates that higher hot-pressing temperature and longer hot-pressing time will lead to the formation of a more uniform interface. The most uniform interface obtained in this study was a composite vacuum hot-pressed at 600 °C, 70 MPa for 32 h (Fig. 7).

No interface was observed in SSF/Al composite vacuum hot-pressed at 500 °C, 70 MPa for 30 min as shown in Fig. 2b of [13]. This composite was further heat treated in air and vacuum, respectively, at 600 °C for 5 h. The heat treatment was performed without application of pressure. Fig. 10 shows the resulting optical micrographs. The resulting interfaces were irregular in both cases; however, the composite heat treated in a vacuum possessed a much more regular interface than the composite heat treated in air. Comparison of Figs 1 and 10 clearly indicates that the application of pressure (70 MPa in this case) and vacuum heat treatment promote the formation of a uniform interface. The application of pressure could enhance the bonding and contact between SSF and the aluminium matrix which would, in turn, make the interdiffusion more uniform across the SSF/Al interface; therefore, a uniform interfacial compound is formed. Heat treatment in vacuum avoids the diffusion of oxygen into the interface which could complicate the interdiffusion of SS and the aluminium matrix.

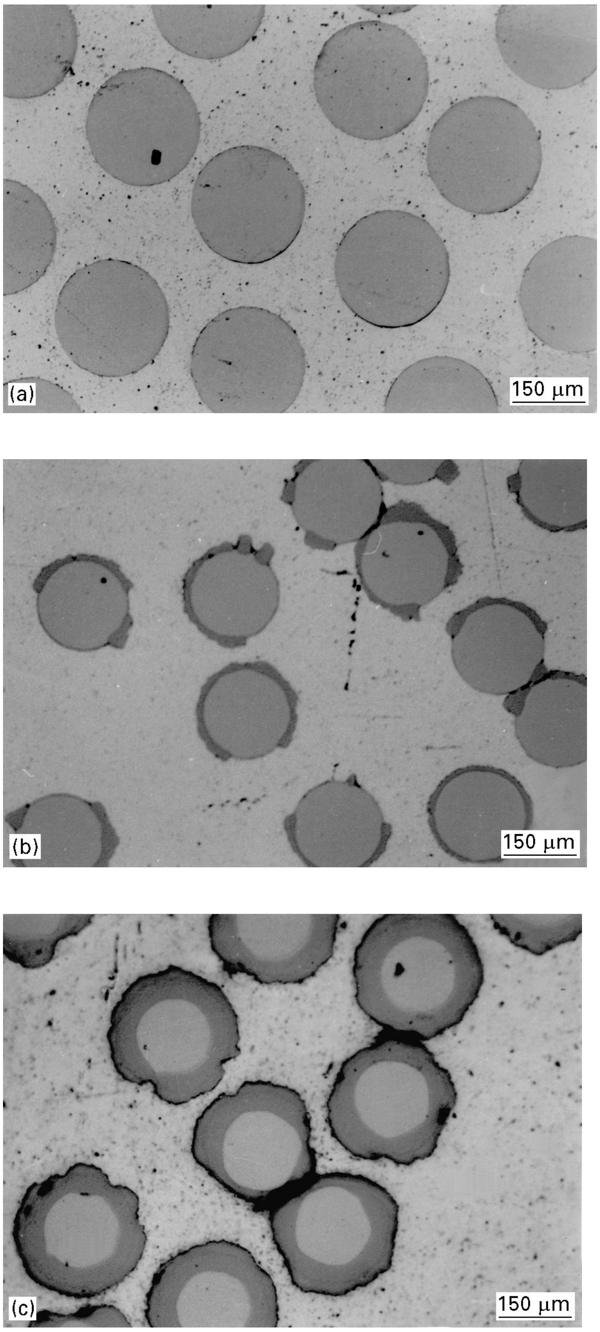

Figure 10 Optical micrographs for the composite vacuum hot-pressed at 500 °C, 70 MPa for 30 min (a) followed by heat treatment at 600 °C for 5 h in (b) air and (c) vacuum.

4. Conclusions

Based upon the experimental results and discussions provided above, the following conclusions can be drawn.

1. A uniform and ringed interface has been obtained in AISI 304 SSF/Al composite which enables us to determine the growth kinetics of the fibre/matrix interface in a more accurate way than has ever been done before.

2. It has been determined that the growth of the SSF/Al interface follows the conventional diffusion-controlled parabolic law ($h^2 = kt$) in the initial stage of interface formation. After some time, the experimentally obtained interface growth rate fell behind that predicted by the parabolic law. To explain the deviation, a modified parabolic law ($[h(r + r_0)/2r_0]^2 = kt$) has been proposed and proved to be a better model to fit experimental data.

3. It has been determined that the activation energy of the interface reaction is 152 kJ mol^{-1} which is about 8% lower than obtained by Mannan *et al.* [15]. The lower Q value is attributed to the pressure (70 MPa) applied during hot-pressing in this study.

4. An EDS analysis and microhardness measurement indicates that the SSF/Al interface consists of a mixture of intermetallic compounds Fe(Cr, Ni)Al_2, Fe(Cr, Ni)Al_3 and (Fe, Cr, Ni)$_2Al_7$. Further investigation is required to determine the exact percentages of various compounds.

5. It has been determined that the morphology of the interface is rather sensitive to the process parameters. The parameters which promote the formation of a uniform interface have been found to be: hot-pressing at higher temperature (600 °C); application of pressure during heating (70 MPa); hot-pressing in vacuum; and hot-pressing for a longer time (32 h).

Acknowledgement

We acknowledge the financial support for this research by the National Science Council, Taiwan, under Grant NSC-83-NSPO-B-RDD-007-05.

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Received 10 August 1995
and accepted 2 August 1996