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Effect of ceramic particle size and applied pressure on time to complete infiltration of liquid aluminium into SiC powder compacts

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Monitoring of pressurized advance of liquid aluminium into SiC particle compacts of mean particle sizes 12.8, 22.8 and 36.7 μm was carried out under applied pressures of 400 to 900 kPa. Infiltration time of liquid aluminium in the compact was recorded by computer logging the output from pairs of open ended wires with a potential difference between them. Results showed that time necessary to complete infiltration in the compacts decreased with increasing particle size and increasing applied pressure. Experimental results are in reasonably good agreement with model predictions. *© 2000 Kluwer Academic Publishers*

1. Introduction

Among the liquid state routes to formation of Metal Matrix Composites, infiltration of ceramic preforms by liquid metal, especially by liquid aluminium, has attracted much attention [1–9]. However, the infiltration of a liquid metal into a porous preform is a complex physicochemical and hydrodynamic phenomenon that involves considerations of wettability, interfacial reactions and fluid flow through the porous medium. The non-wetting of ceramic reinforcements by liquid aluminium [10–15], at temperatures below 1100 K, has the consequence that the applied pressure P must exceed the threshold value P_0 required to overcome capillary back pressure before the melt will infiltrate the compact. The only additional force then to be overcome is viscous drag by the liquid metal. Following Semlak and Rhines [16], Delanney *et al.* [1] expressed the infiltration length L of a liquid through a capillary of circular cross section as:

$$L = \left[\frac{r^2 t}{4\mu}\left(P + \frac{2\gamma_{\mathrm{lv}}\cos\theta}{r}\right)\right]^{1/2} \qquad (1)$$

where r is the capillary radius, t is the time, μ is the viscosity of the fluid, γ_{lv} is the surface tension of the liquid and θ is the wetting angle where P is applied pressure.

Advance of liquid metal by infiltration into ceramic preforms has been investigated mainly for fibrous or whisker preforms with solidification occurring concurrently [17–21], with rather less work carried out on particle preforms with the preform temperature equal to that of the infiltrating liquid metal [22–24]. Martins *et al.* [22] developed a model based on a bundle of capillary tubes as an analogue for infiltration of SiC particle compacts by aluminium. Maxwell *et al.* [23] measured the infiltration distance versus time for aluminium into SiC particle compacts at 670°C and found that an incubation time had to be exceeded before infiltration could start. Alonso *et al.* [24] reported that infiltration distance of liquid aluminium into SiC or TiC particle compacts at 750°C was proportional to $t^{1/2}$ as predicted by Equation 1. We previously reported on the effects of particle size, impurities, melt superheat [25], and alloying additions [26, 27] to pure aluminium and on the role of surface tension in relation to contact angle for infiltration of SiC particle compacts [28]. The aim of the present work was to monitor isothermal advance of liquid aluminium into SiC particle compacts to explore further the applicability of Equation 1.

2. Experimental procedures

Aluminium with a purity of 99.95% was used as the matrix metal for infiltration. The preform material was abrasive grade green α-silicon carbide particulates with mean particle diameters of 12.8, 22.8 and 36.7 μm supplied by Washington Mills Ltd (UK). The particle size and chemical composition data given by the supplier for these particulates is given in Tables I and II respectively.

Details of the apparatus employed for the infiltration tests are given elsewhere [26]. Briefly, the apparatus consisted of four separate parts, a cylindrical vessel, lid assembly, pressure control unit and a cooling unit. Pairs of open ended wires were used with a 9 volt potential

TABLE I The average particle size and distribution range of green α-SiC particulate used (Supplier's data)

Identity	Mean diameter d_{50}, (μm)	Particle Size Distribution Range (μm)
A	12.8	5–25
B	22.8	12–40
C	36.7	22–59

TABLE II Typical composition (wt%) of SiC particles used (Supplier's data)

Ceramic	SiC	SiO_2	Si	Fe	A1	Free C
Green SiC	99.7	0.15	0.02	0.02	0.02	0.15

difference between them to record infiltration advance. A fused silica tube of 15 mm O.D., 10.4 mm I.D. and 167 mm long was used as a sample holder. A schematic of the wired sample holder used for liquid infiltration advance measurements is shown in Fig. 1. The preparation procedure of the sample holder was as follows. Two copper wires approximately 600 mm long and 0.15 mm diameter were threaded into an elliptical section twin bore silica thermocouple insulator. After threading the wires through the insulator, the top ends with 40 mm of wire protruding were fixed with sellotape to prevent further movement of the wires. The lower protruding ends were cut 60 mm below the insulator to detect infiltration in the initial (lower) part of the sample. The same procedure was applied to the second insulator except that the wires were cut 35 mm below the insulator to detect infiltration to the middle part of the sample. The insulators were then joined to each other with ceramic cement to ensure that the ends of the insulators inside the tube would be at the same level. The lower end of the silica tube was plugged with a porous alumina plug and marked with a marking pen at distances 3, 28 and 53 mm upwards from the plug. This filter was 10 mm diam. and 5 mm long and had 85% porosity, offering negligible resistance to flow of the alloy melt into the SiC compact. Then the insulators were inserted with some ceramic cement into the silica tube ensuring that the ends of the wires matched the markings. After the ceramic cement had hardened, a third pair of wires were placed either side of the existing insulators to a distance of 53 mm from the alumina plug, and fixed with sellotape to prevent the wires from touching each other. Following that, 8 g of SiC particulate was poured into the silica tube and the samples were vibrated for about 15 seconds which gave a compact approximately 54 mm in height. After this vibratory compaction of the powder, a porous alumina block and an alumina rod as a support were inserted on top of the powder compact and fixed with ceramic cement in order to prevent disturbance of the compact during infiltration processing.

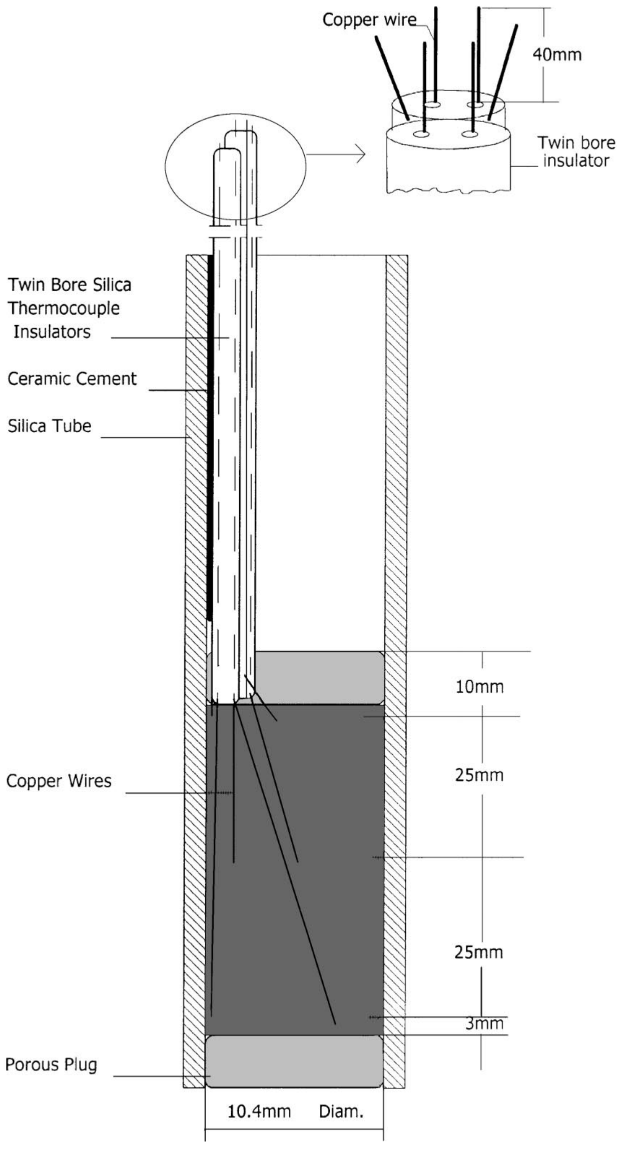

Figure 1 Schematic of the wired sample holder for liquid infiltration advance measurements in SiC powder compacts.

The samples which had been specially prepared for infiltration advance measurements were fixed to the adapter (Fig. 2). The connection of the wired sample holder to the sample holder connection tube via the adapter and connection of the wires to the computer are shown in Figs 2 and 3 respectively. Approximately 360 g of aluminium alloy was placed in an alumina crucible which had been pre-positioned inside the vessel. The pressure vessel was then heated by means of an external induction coil. Normally the infiltration temperature was 750 ± 5°C and samples were held at this temperature for about 3 min. to achieve temperature uniformity between the liquid and the compact. Pressure was then applied to the vessel up to the predetermined pressure as rapidly as possible and held until a signal was received from the end position of the sample. Data were collected by the computer data logging system every 0.25 seconds during infiltration. The pressure was then released from the vessel and the lid assembly was detached from the vessel to admit air.

3. Results and discussion

Table III shows the experimental results for times t_1 and t_2 to complete 25 mm (L_1) and 50 mm (L_2) of infiltration of A, B and C SiC compacts by liquid aluminium at 750°C under applied pressures P between 400 and 850 kPa. Applied pressure was chosen in each

TABLE III Measured times t_1 and t_2 for Al at 750°C to isothermally infiltrate 25 mm (L_1) and 50 mm (L_2) of SiC particle compacts under various applied pressures P, compared with predicted values t_1' and t_2' using Equation 3 and $\mu = 1 \times 10^{-3}$ Pa s at 750°C

Particle Identity	d_{50} mm	r_h μm	P_0 kPa	P kPa	$P - P_0$ kPa	t_1 s	t_2 s	t_1' s	t_2' s	t_1/t_1'	t_2/t_2'
A	12.8	2.0	850	900	50	9,12,13 = 11.3 ± 1.5	16,17,19 = 17.3 ± 1.5	12.5	50.0	0.9	0.4
B	22.8	3.4	565	600	35	5,7,7.5,8,8,10,12 = 8.2 ± 2.2	12, 16, 16, 18, 20, 20, 23 = 17.8 ± 3.6	6.2	24.7	1.3	0.7
B	22.8	3.4	565	650	85	3,6 = 4.5 ± 2.1	11.5,13 = 12.3 ± 1.1	2.5	10.2	1.8	1.2
B	22.8	3.4	565	700	135	3, 5, 5, 6 = 4.8 ± 1.3	8.9, 10 = 9.0 ± 1.1	1.6	6.4	3.0	1.4
C	36.7	5.3	350	400	50	3	9, 12, 17 = 12.7 ± 4.0	1.8	7.1	1.7	1.8

Note: Effective channel radius r_h was estimated from d_{50} using Equation 4. Threshold pressures P_0 are measured values from ref [29].

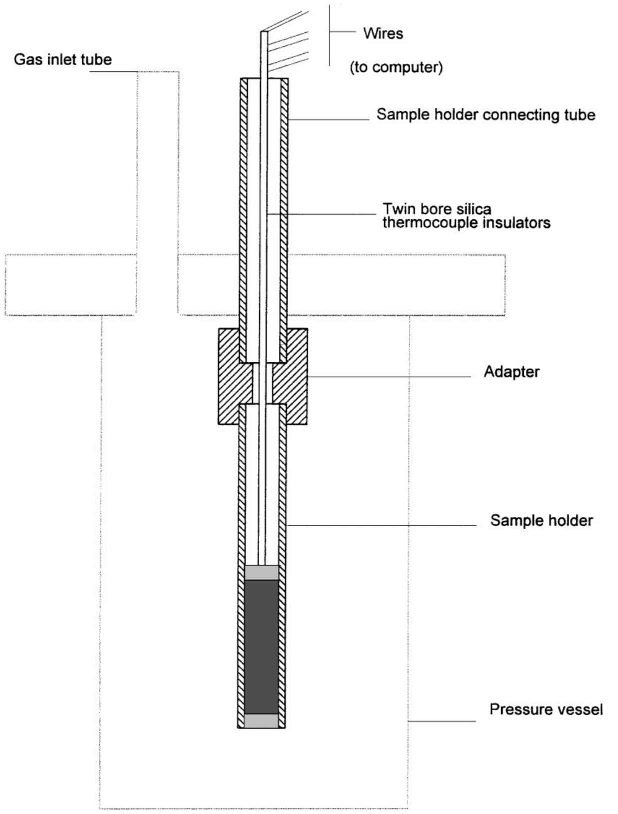

Figure 2 Schematic showing connection of the wired sample holder to the sample holder connection tube via the adapter.

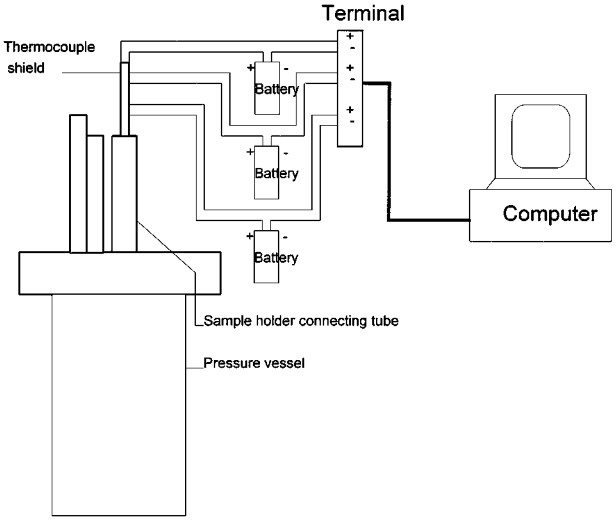

Figure 3 Schematic showing connection of the wires for infiltration advance measurements to the data logging computer system.

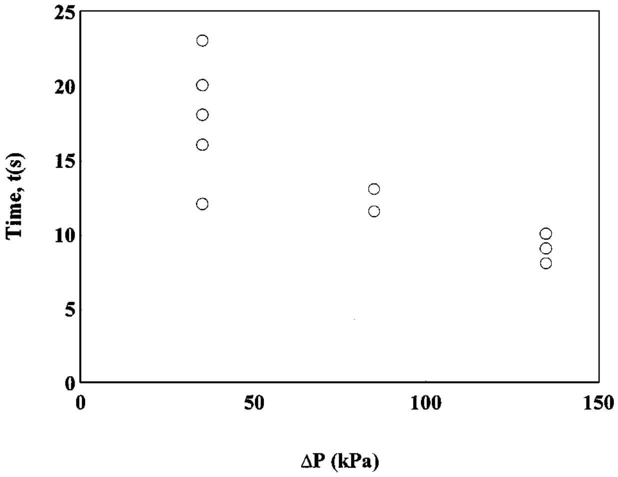

Figure 4 Infiltration time vs applied pressure differential to complete 50 mm infiltration length of liquid aluminium in particle B SiC compacts at 750°C.

case to exceed by 35 to 135 kPa the indicated threshold pressure P_0 to initiate infiltration which had been determined previously [29]. Typically three repeat experiments were carried out for each condition. Although the results show some scatter, t_{exp} at the two positions increases with increasing $\Delta P = P - P_0$ for particle B compacts (Fig. 4) and with decreasing SiC particle size at the same ΔP (Fig. 5). These results can be interpreted by reference to Equation 1 for advance of infiltrating liquid through a capillary of radius r under applied pressure P, which can be rewritten as:

$$t = \frac{4\mu\left(\frac{L}{r}\right)^2}{\left(P + \frac{2\gamma_{\text{lv}}\cos\theta}{r}\right)} \tag{2}$$

In the present case the capillary pressure $2\gamma_{\text{lv}}\cos\theta/r$ is negative reflecting nonwetting conditions ($\theta > 90°$), so that $2\gamma_{\text{lv}}\cos\theta/r$ can be identified with the measured threshold pressure for infiltration, P_0, and Equation 2 becomes

$$t = \frac{4\mu(L/r)^2}{\Delta P} \tag{3}$$

where $\Delta P = P - P_0$. Similarly, for particle compacts, capillary radius r in Equations 1 to 3 can be replaced

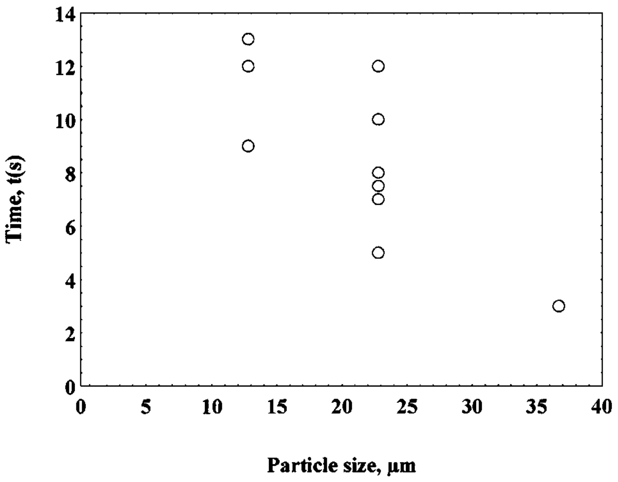

Figure 5 Infiltration time t to complete 25 mm infiltration of liquid aluminium as a function of SiC particle size under pressure differential of 35–50 kPa at 750°C.

by hydraulic radius r_h given by [23, 30]:

$$r_h = \frac{(1 - V_p)d}{6V_p} \tag{4}$$

where d is the mean particle size and V_p is the volume fraction of particles in the compact. The times t_{th} to infiltrate 25 and 50 mm of compact, calculated on this basis with $\mu = 10^{-3}$ Nsm^{-2} for aluminium at 750°C [31], are given in Table III for comparison with the measured values. Agreement is within a factor of 3 and typically within a factor of 2. The predicted relationship between L and t is also compared with the individual measurements in Figs. 6–10, while Fig. 11 shows a composite plot of $(r/L)^2t$ versus ΔP. Fig. 11 also shows the theoretical prediction based on $\mu = 10^{-3}$ Ns m^{-2} and the factor of 2 scatter band. The results showing most deviation were for the particle A compact at $\Delta P = 50$ kPa which completed 50 mm of infiltration in one-third of the predicted time and the particle B compact at $\Delta P = 135$ kPa which took three times as long as predicted to penetrate the first 25 mm (Table I). The latter discrepancy could possibly be attributed to sedimentation of small particles towards the

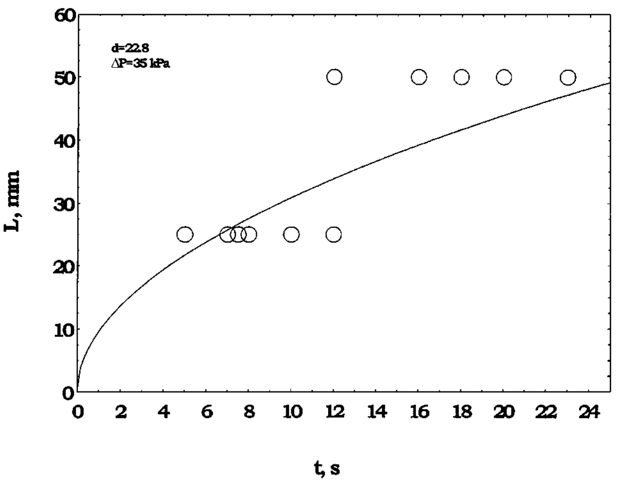

Figure 6 Showing experimental (as marked) and predicted (line) infiltration length vs itme for the infiltration of particle B compact at 600 kPa applied pressure at 750°C.

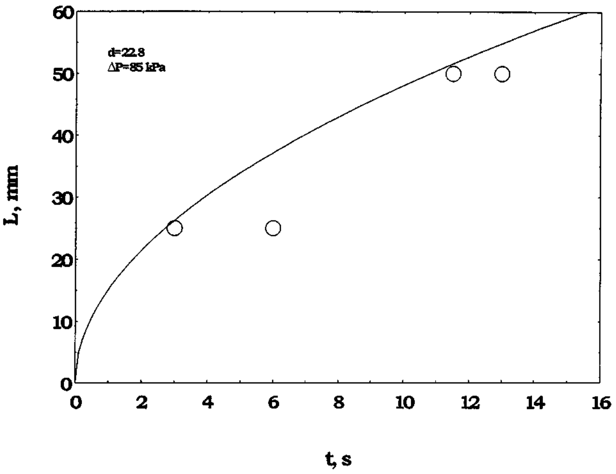

Figure 7 Showing experimental (as marked) and predicted (line) infiltration length vs time for the infiltration of particle B compact at 650 kPa applied pressure at 750°C.

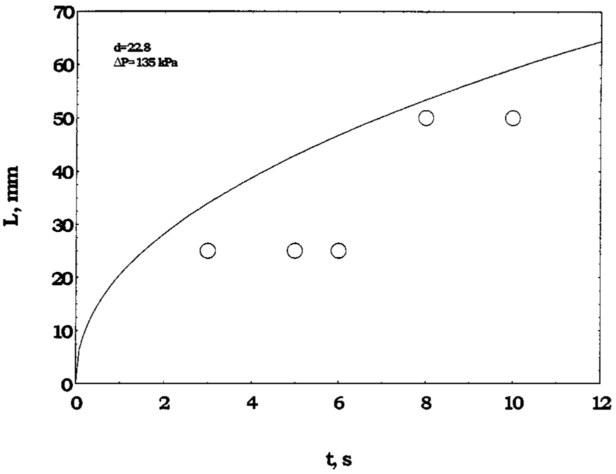

Figure 8 Showing experimental (as marked) and predicted (line) infiltration length vs time for the infiltration of particle B compact at 700 kPa applied pressure at 750°C.

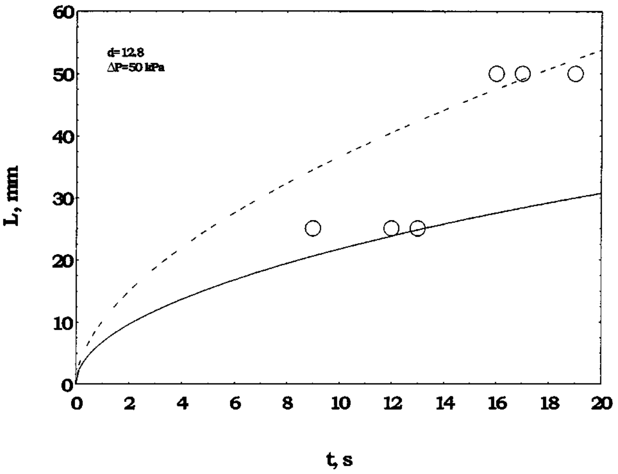

Figure 9 Showing infiltration length vs experimental (as marked) and predicted (line) time for the infiltration of particle A compact at 850 kPa applied pressure at 750°C. Dashed line corresponds to a particle diameter d of 19 μm under $\Delta P = 50$ kPa pressure.

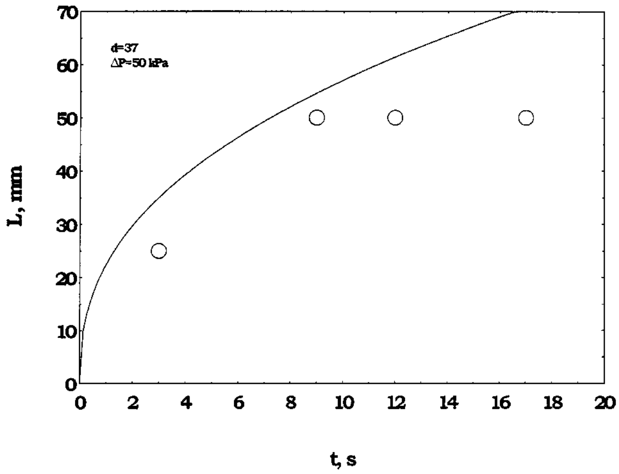

Figure 10 Showing experimental (as marked) and predicted (line) infiltration length vs time for the infiltration of particle C compact at 400 kPA applied pressure at 750°C.

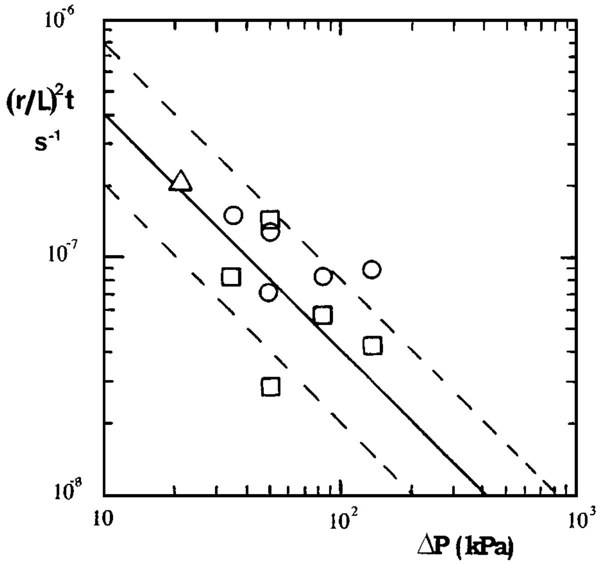

Figure 11 Composite plot of $(r/L)^2t$ versus ΔP for isothermal infiltration of aluminium into SiC powder compacts at 750°C. Key: ○: $L = 25$ mm; □: $L = 50$ mm. Full line indicates predictions of Equation 3, and dashed lines define the factor of 2 scatterband. Δ is from ref. [23].

bottom of the compact during vibratory compacting, reducing the effective initial value of r_h and increasing resistance to penetration (decreasing t to penetrate a given distance L). The former is more difficult to explain. One possibility is a short-circuiting of the pair of sensor wires involved as a result of liquid running ahead of the main advancing front due to longitudinal cracks through the upper part of compact or a gap between the upper part of the periphery and its quartz containment, even though every care was taken to avoid such eventualities.

Alonso *et al.* [24] reported penetration L proportional to $t^{1/2}$ for infiltration of 99.98 pct aluminium into SiC and TiC particulates compact at 750°C with $\Delta P = 70$ and 40 kPa respectively. For SiC the resulting $(r/L)^2t$ value is 1×10^{-6} s which is more than one order of magnitude larger than the value for $\Delta P = 70$ kPa in Fig. 11. These results, however, were obtained by measuring the distance infiltrated during a total period of sustain of the applied pressure. There was no internal sensor to record the instant during sustain at which melt first entered the compact, so the actual duration of infiltration could have been much shorter. The results of Maxwell *et al.* [23] displayed an incubation time t_0 as long as 25 s for infiltration of Al, Al-1Si and Al-1Mg into SiC compacts at 670°C at applied pressures at or just above the threshold pressure (defined by them as the minimum applied pressure required to initiate infiltration within 30 minutes of its application). Their result for pure aluminium at 670°C with $\Delta P = 21$ kPa gives $(r/L)^2(t - t_0) = 2.5 \times 10^{-7}$s in excellent agreement with the predictions in Fig. 11.

4. Conclusions

1. Time to isothermally infiltrate molten aluminium 25 and 50 mm into SiC particle compacts decreased with increasing applied pressure P in excess of the threshold pressure P_0 and with increasing SiC particle size.

2. Measured times of infiltration at 750°C are in good agreeement with the predicted relationship $t = 4\mu(L/r)^2/(P - P_0)$ where L is distance infiltrated, r is effective channel radius in the preform and μ is the viscosity of the infiltrating molten aluminium.

3. The previous result of Maxwell *et al.* [23] for the same system at 670°C and $P - P_0$ of 21 kPa is also in good agreement with this equation but the infiltratiom times reported by Alonso *et al.* [24] for 750°C and 70 kPa are more than an order of magnitude larger, possibly reflecting an initial delay at the outset of the pressure sustain, before infiltration started, as was found earlier by Maxwell *et al.*

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