New technique for pressureless infiltration of Al alloys into Al_2O_3 preforms

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Al alloys were infiltrated into alumina preforms without the aid of pressure in N_2 as well as in air at and above 750 °C. It was possible to eliminate termination of infiltration that was seen in open conditions (where N_2 was in continuous contact with the melt) by modifying the infiltration geometry. This configuration enables the infiltration to continue for longer periods of time, consequently producing greater thickness of composite. In air, Mg placed at the interface getters the in-coming oxygen until the alloy billet melts and seals off the front from the ingress of the furnace atmosphere thereby eliminating the need for prealloying Al with Mg and N_2 atmosphere. In addition, experiments in argon revealed that the infiltration requires some critical amount of N_2 in the atmosphere.

I. INTRODUCTION

High volume fraction metal-matrix composites (MMCs) are being considered for wear resistance, structural, and electronic packaging applications. Among all the techniques of processing MMCs, infiltration of molten metals into porous ceramic preforms is the only technique suitable for processing these composites. The infiltration can be classified into three categories based on the source of driving force: (i) external pressure assisted, (ii) vacuum driven, and (iii) capillarity driven or pressureless. The main drawbacks of pressure-assisted infiltration are expensive tooling and the difficulty involved in making dies for making complex-shaped components, although the process is quick. Pressureless infiltration is attractive due to its cost effectiveness and near-net-shape capability. In pressureless infiltration, Al-Mg-based alloys infiltrate spontaneously into ceramic preforms in nitrogen atmosphere above 700 °C. Although pressureless infiltration of Cu into Fe powder compacts was well established¹ way back in 1958, in the case of Al alloys it was first reported in 1989 by Aghajanian *et al.*² of the Lanxide Corporation. The essential requirements for the pressureless infiltration of Al alloys are presence of Mg in the alloy or in the infiltrating system and N_2 in the furnace atmosphere. Aghajanian *et al.*³ in their later study reported the effect of Mg content in the alloy, amount of N_2 in the atmosphere, particle size and temperature on the rate of infiltration, and the amount of nitride formed in the matrix. A critical amount of Mg is needed to initiate the infiltration process and it is dependent on the other process parameters, namely, particle

size, temperature, porosity, and partial pressure of N_2 . It has been suggested⁴ that Mg in the alloy evaporates and reacts with N_2 in the furnace atmosphere and forms a coating of Mg_3N_2 on the ceramic reinforcement, which on coming in contact with molten Al gets reduced to AlN thereby recycling the Mg. Hence, it is supposed that the reaction between Mg_3N_2 and Al induces wetting and allows the pressureless infiltration of Al alloys into ceramic preforms.

Srinivasa Rao and Jayaram,⁵ by studying the infiltration of Al-Mg-based alloys into Al_2O_3 preforms, reported that Mg played a role by reducing the native alumina layer on the alloy and by gettering the residual oxygen in the N_2 , thereby keeping the front free of any passivating alumina/spinel layer. In flowing nitrogen, infiltration tended to terminate after 1 to 2 mm of growth owing to surface depletion of Mg and the consequent inability to remove oxygen impurities in the in-coming gas. (See Fig. 1.) All methods to avoid termination involve the provision of Mg-rich getters that reduce the oxygen partial pressure sufficiently at the gas/melt interface. In the present work, the principle of gettering residual oxygen with a strong getter such as Mg has been combined with a modified infiltration geometry that makes it possible to infiltrate Al alloys without prealloyed Mg, even in air. Thus, the present article describes a technique of pressureless infiltration wherein both requirements of the conventional technique may be dispensed with, i.e., the infiltration can be carried out in air without prealloying Al with Mg. A related process⁶ utilizes the concept of self-generated vacuum in a porous body to enhance penetration of molten Mg into porous SiC. In addition, the present infiltration

FIG. 1. (a) Weight gain curve depicting various stages of the infiltration of Al-2% Mg alloy into a 53- μ m preform at 900 °C. Letter A indicates termination of infiltration as evidenced by the lack of continued weight gain. (b) Cross section of the infiltrated sample.

experiments under sealed conditions provide some clues regarding the role of atmosphere in pressureless infiltration of Al alloys into Al₂O₃ preforms.

II. EXPERIMENTAL PROCEDURE

A. Preparation of preforms, alloys, and crucibles

Alumina preforms with 40% porosity were prepared by cold pressing fused Al₂O₃ particles. The procedure for the preparation of preforms is as follows. The particles were poured into the die cavity and a few drops of polyvinyl alcohol were added to the powder. Approximately 100 MPa of pressure was applied to get the required density. These green preforms were heated slowly up to 400 °C at 5 °C/min so as to burn out the binder without changing the shape and dimensions of the preform. From 400 °C the samples were heated to higher temperatures (>800 °C) and held there for 1 h to get enough strength for handling in further processing operations. The sintering schedule used in preparing the preforms of different

particle sizes is shown in Table I. The diameter of the preforms used in this study is approximately 7 mm whereas the height ranges from 6 to 10 mm. The porosity of the preforms measured by coating the preforms with wax and using Archimedes' principle was found to be approximately 38%. Since there is a possibility of wax entering the pores of the preform, this method slightly underestimates the actual porosity of the preform. It should be noted that the preform porosity varied from 30% to 35% at the punch side to 55% to 60% at the pin side due to die-wall friction.

Commercial purity Al and Mg were used in preparing the Al-Mg alloys while LM6 (Al-12.5 wt% Si) master alloy was used in preparing Al-Si alloys. Commercial purity Al was melted in a clay-graphite crucible and Mg wrapped with Al foil was added to the melt. The melt was degassed before and after alloying and poured into cast iron finger molds of 15 mm diameter. In preparing Al-Mg and Al-Mg-Zn alloys, 10% excess of Mg was added to compensate for evaporation losses. Some of the Al-Mg and Al-Zn-Mg alloys were initially cast into 60-mm-diameter rods and after homogenization they were extruded to 10-mm- and 16-mm-diameter rods. The chemical composition of the alloys, as determined by either inductively coupled plasma spectroscopy or by atomic absorption spectroscopy, and homogenization temperatures used are given in Table II. Billets of 6.6 to 6.9 mm diameter and a height of 7 to 10 mm were machined from the cast and extruded rods for use in infiltration experiments.

Alumina crucibles for infiltration experiments were prepared by closing one end of an alumina tube of 7 mm inner diameter and 15 mm height with an alumina baseplate using alumina cement and firing at 1500 °C for 2 h.

B. Infiltration experiments under sealed conditions (air, N₂, O₂, and Ar)

In the present context, sealed conditions indicate that once the alloy billet melts and acts as a barrier for gas diffusion to the billet/preform interface, the infiltrating front is no longer in communication with the furnace atmosphere. The Al-alloy billet was kept on top of the preform with 4 to 30 mg of Mg turnings at the billet/preform interface in an alumina crucible and it was introduced into a furnace at 800 to 1000 °C in the case of air, whereas in other atmospheres the samples were heated at a rate of 20 to 50 °C/min in a thermogravimetric

TABLE I. Sintering schedule of the preforms.

Particle size (μ m)	Temperature (°C)	Time (h)
25	1500	1
53	1600	1
90	1600	1

TABLE II. Chemical composition of the alloys and homogenization temperatures.

Alloy	Mg wt%	Si wt%	Zn wt%	Fe wt%	Homogenization temperature and time
Al-2Mg	1.92	...	...	...	350 °C, 24 h
Al-5Mg	4.9	...	...	...	350 °C, 24 h
Al-12Si	...	12.6	...	...	...
Al-6Zn-0.3Mg	0.28	...	5.9	...	350 °C, 24 h

analyzer (TGA; Cahn, TG-171) and the gas was flowed at 100 ml/min. No Mg was kept at the interface in N₂ and N₂-2% H₂ unless specified otherwise. In atmospheres other than air, the TGA reactor tube, in which the sample was hung, was evacuated once to 0.07 mbar and back filled with N₂, or O₂ or Ar as the case may be. The samples were held at the set temperature for up to 3 h and then removed from the furnace and cooled in air, in the case of infiltration in air, whereas the samples were cooled at a rate of 10 °C/min within the TGA in other atmospheres. In the case of upward infiltration, the preform was kept on top of the billet in an inverted crucible such that the molten metal acted as a seal for one side of the preform, whereas the crucible base acted as a seal on the other side. Infiltration was carried out in both upward and downward modes. The schematic of the billet/preform configurations used for infiltration in sealed conditions is shown in Fig. 2. The metal did not flow down when the crucible was inverted, as shown in Fig. 2(b), in the upward infiltration configuration.

C. Characterization

After infiltration experiments, the samples were sectioned longitudinally with a high-speed diamond saw (Isomet 2000, Buehler Company, Lake Bluff, IL). The infiltration heights were measured using a traveling microscope with a least count of 10 µm. Composite samples were ground to a powder and characterized by x-ray diffraction (XRD) to identify the various phases present in the composite. In some cases, the solidified alloy remaining after infiltration was analyzed for its composition or was directly placed in the x-ray diffractometer for the purpose of identifying the compounds formed on the surface during infiltration. Infiltrated samples were sectioned with a high-speed diamond saw.

Sectioned samples were mounted and polished metallographically to 0.25-µm diamond finish. The polished samples were examined with an Olympus optical microscope to determine the distribution of various phases present in the composite.

III. RESULTS

The results of the infiltration experiments in N₂, air, O₂, and Ar atmospheres are reported in the Sects. III. A to III. D. The results presented in this section are of downward infiltration unless stated otherwise.

A. N₂-2% H₂/N₂ atmosphere

The results of the infiltration experiments carried out in N₂ and N₂-2% H₂ are given in Table III. In order to find out whether commercial purity Al without any Mg in the alloy form infiltrates alumina, infiltration was tried with Al by keeping 6 mg (15.6 mg/cm²) of Mg at the billet/preform interface. Al did infiltrate into the alumina. However, infiltration did not occur when the amount of Mg at the interface was reduced to 3 mg (7.8 mg/cm²) thereby demonstrating the fact that some critical amount of Mg is needed to infiltrate Al into alumina. It may be noted that in the absence of sealed conditions, i.e., under open conditions wherein flowing N₂ at 1 atmosphere is in continuous contact with the melt surface during infiltration, as shown in Fig. 2(c), infiltration ceases after approximately 2 mm for the case of Al-2Mg as reported elsewhere.⁵

B. Air atmosphere

The height of infiltration of Al and Al alloys into 53-µm particulate preforms at 900 °C with 10 mg of Mg powder (26 mg/cm²) at the billet/preform interface is given in Table IV. XRD of the powder collected from the

FIG. 2. Schematic of the billet/preform assembly used for infiltration under sealed conditions: (a) downward infiltration, (b) upward infiltration (no Mg turnings were used in N₂), and (c) schematic of the billet/preform assembly used for upward infiltration in nitrogen under open conditions. Gas was flown in the downward direction in all the cases. (In the case of air, the atmosphere was still.)

interface, where Mg powder was kept, of the sample held at 900 °C for 1 h and 15 min showed only Al and MgO peaks. Chemical analysis of the billet showed 1 wt% Mg due to the dissolution of Mg into the billet. For comparison, the amount of Mg introduced at the interface would, if completely dissolved in the alloy, lead to a concentration of 1.4 wt%. The amount of Mg dissolved into the billet came down to 0.4% when the weight of the billet was doubled. When Al–2% Mg and Al–8% Mg alloys were kept for 6 h 30 min at 950 °C without any Mg powder at the alloy/preform interface, no infiltration occurred thereby demonstrating that free Mg at the billet/preform interface is essential for effecting infiltration in air. Experiments with progressively decreasing amounts of Mg at the billet surface revealed that a negligible portion of the preform was infiltrated with 3 mg of Mg at 900 °C (2 h and 15 min) while a similar amount of Mg produced 1 mm of infiltration at 1000 °C (2 h). The results of the infiltration of Al into the 53- μ m preform as a function of time at 900 °C are presented in Table V. The interesting feature is that the rate of infiltration remained more or less constant over the time period investigated. However, the rate of infiltration is much faster when the original atmosphere is nitrogen (see Tables III and IV) than when it is air. In addition, infiltration did not even begin when the experiment was carried out under a vacuum of 0.03 mbar. This observation indicates that a critical amount of nitrogen in the atmosphere is required for effecting pressureless infiltration whether the balance is composed of argon or is a vacuum.

C. Oxygen atmosphere

The results of the infiltration experiments with Al–2 wt% Mg alloy, with Mg powder at the billet/preform interface and Al disk or billet on top of the alloy billet to reduce Mg evaporation, into the 53- μ m preforms in oxygen atmosphere at various temperatures are given in Table VI. Optical microscopy of the samples revealed that a few particles were seen to stick to the billets of the samples held at 800 °C with 4.5 mg (11.7 mg/cm²) and 5.2 mg (13.5 mg/cm²) of Mg at the interface. However, alumina particles sticking to the billet were engulfed in a layer of reaction product when the amount of Mg was

raised to 10 mg and the temperature to 900 °C thereby indicating that the amount of Mg was not sufficient to stop the oxidation of billet surface. When the amount of Mg increased to 30 mg (78 mg/cm²), the alloy started infiltrating into the preform. However, the alloy could not infiltrate the preform fully due to the formation of

TABLE IV. Height of infiltration (downward) of various Al alloys into 53- μ m Al₂O₃ preforms in air.

Alloy	Temperature (°C)	Time at set temperature (min)	Infiltration height (mm)
Al	800	180	6.0
Al	900	180	6.8 ^a
Al–2%Mg	900	135	5.0
Al–12%Si	750	150	1.0
Al–12%Si	900	120	6.8 ^a
Al–6%Zn–0.3%Mg	900	120	6.8 ^a

^aFull preform infiltrated.

TABLE V. Height of infiltration (downward) of Al into 53- μ m Al₂O₃ preforms as a function of time at 900 °C in air with 10 mg of Mg powder at the billet/preform interface.

Time (min)	Infiltration height (mm)
20	1.0
45	2.0
90	5.0

TABLE VI. Infiltration experiments with Al–2%Mg alloy with Mg powder and with 53- μ m Al₂O₃ preforms at different temperatures under sealed conditions in oxygen.

Temperature (°C)	Amount of Mg (mg)	Infiltration direction	Observation
800	...	Down	No infiltration
800	4.5	Down	No infiltration
800	5.2	Down	No infiltration
900	10	Down	No infiltration
900	30	Down	Started infiltrating but formed lot of MgAl ₂ O ₄ and stopped
800	24	Up	No infiltration; three-fourths of the preform turned black
900	5	Up	No infiltration; some portion of the preform turned black

TABLE III. Height of infiltration of Al–Mg alloys (without Mg turnings) in N₂ and N₂–2% H₂ at 800 °C.

Alloy	Particle size (μ m)	Atmosphere	Infiltration direction	Time at set temperature (min)	Infiltration height (mm)
Al–2%Mg	25	N ₂ –2%H ₂	Down	120	6.8 ^a
Al–2%Mg	53	N ₂ –2%H ₂	Down	120	6.8 ^a
Al–2%Mg	53	N ₂	Down	50	6.0
Al–5%Mg	53	N ₂	Up	30	7.0

^aFull infiltration.

reaction product at the infiltration front. The reaction product was identified by XRD as MgAl₂O₄ spinel. Therefore, it can be inferred that infiltration of Al alloys into Al₂O₃ preforms is not possible in oxygen under the conditions wherein infiltration occurred in air.

D. Argon atmosphere

The results of the infiltration experiments with Al-2 wt% Mg and Al with Mg powder at the billet/preform interface in argon atmosphere at various temperatures are given in Table VII. No infiltration occurred in the temperature range of 800 to 900 °C. In all cases, whether it is a preform containing 25-μm or 53-μm particulate, the preform turned black or dark gray depending on the extent of coating and reaction of Mg vapor. The greater the content of Mg in the alloy or at the interface, the more is the height and extent of preform discoloration. In addition, the extent of discoloration increased with temperature. Infiltration did not occur even when the entrapped argon gas pressure was reduced to 0.03 mbar by evacuating the system prior to heating. In this case, the whole preform turned black. (See Fig. 3.)

IV. DISCUSSION

From the infiltration experiments in various atmospheres, it is quite evident that atmosphere plays a crucial role in effecting pressureless infiltration. The physico-chemical phenomena taking place at the infiltrating metal/gas interface, which are responsible for the observed infiltration in nitrogen and air along with the reasons for no infiltration in argon, are discussed below. The results of the experiments in sealed conditions are complemented with the results of the experiments done in open conditions⁵ in understanding the role of atmosphere.

It is useful to review briefly the infiltration behavior of an alloy such as Al-2 Mg in open conditions at temperatures approximately 800 to 900 °C. Initial, rapid infiltration to a thickness of about 2 mm is followed by a sudden termination whose cause was identified to be the localized depletion of Mg at the advancing interface. It was shown that this depletion could arise out of the inability

FIG. 3. Cross section of Al with the preform held in argon at 900 °C with 9.6 mg of Mg at the interface and under a vacuum of 0.03 mbar.

of diffusion in the alloy channels to keep up with evaporative loss and reaction in the vapor phase to form MgO. This led to the likelihood of melt passivation owing to the formation of alumina or spinel. Thus, the primary role of Mg appeared to lie in its ability to getter oxygen. In the present experiments, further confirmation of this important role is evident from the observation that most of the Mg kept at the interface turned to MgO with only 0.4 wt% of Mg dissolved into the melt. Thus, in air, after the Mg at the billet/preform interface getters the oxygen coming to the interface until the billet melts and seals off the interface from the ingress of air, further gettering is no longer needed. Any air entrapped within the preform reacts with Mg vapor and forms minute amounts of MgO and Mg₃N₂. Thus, the termination problem is completely eliminated unlike the case of infiltration under open conditions.⁵

When the sealed and open conditions are compared, it is found that the rates of infiltration are comparable, despite the substantially different partial pressures in the two cases. In addition, it may be readily established that

TABLE VII. Infiltration experiments with Al-2%Mg and Al with Mg powder and with 25-μm and 53-μm alumina preforms under sealed conditions in ultrahigh purity argon at different temperatures.

Temperature (°C)	Alloy	Amount of Mg (mg)	Observation
800	Al-2% Mg	...	No infiltration
900	Al + Mg	8.9	No infiltration; three-fourths of the preform turned black
900	Al + Mg	8.0	No infiltration; a portion of the preform turned black
900	Al + Mg	10.5	No infiltration; a portion of the preform turned black
900 ^a	Al + Mg	9.6	No infiltration; whole preform turned black

^aVacuum of 0.03 mbar.

the rates are orders of magnitude slower than would be expected by simple conditions of laminar flow in a capillary pressure gradient. (See Appendix.)

Thus, in both open and sealed conditions, it appears that chemical reactions at the surface of the infiltration front play an important rate limiting role. Whereas under open conditions it is suspected that layers of Mg₃N₂ or AlN may form periodically and need to be disrupted, a similar sequence of events is more difficult to envisage under sealed conditions in which the number of available gas molecules from the outer atmosphere is fixed and negligible, after the metal seals the preform. The possible surface films that can exist on the melt surface under various conditions are now examined.

In N₂, under open conditions, Mg evaporated from the melt surface getters the residual oxygen in the vicinity of the infiltration front. In commercial purity nitrogen, the amount of residual oxygen was around 5000 ppm, which corresponds to a partial pressure of 500 Pa. At this partial pressure of oxygen, the number of oxygen molecules in a region of 10 μm thick and 0.1539 cm² (effective cross-sectional area of the preform) at 800 °C is around 10¹³. The rate of evaporation of Mg from an infiltration front area of 0.1539 cm² for an Al–8% Mg alloy was around 9.2×10^{19} atoms/s, assuming that the whole weight gain was due to the formation of MgO.⁵ With such a high flux of Mg compared to the oxygen, the partial pressure of oxygen in the 10-μm-thick region will be reduced to a very low value. Because Mg is the most dominant vapor species at 800 °C (vapor pressure of Al is very low; $P_{\text{Al}}/P_{\text{Mg}} = 10^{-7}$), Mg reacts with O₂ and N₂ thereby forming MgO and Mg₃N₂ according to the following reactions:

In static N₂ atmosphere, Mg can reduce the partial pressure of residual O₂ to less than 10^{−40} bar at 800 °C by Reaction (1). For example, the number of oxygen atoms in a region of 10 μm thickness and a cross-sectional area of 0.1539 cm² at an oxygen partial pressure of 10^{−10} bar is around one. Therefore, it would not be possible for one oxygen atom to cover the entire surface of 0.1539 cm² area, not to mention an oxygen partial pressure of 10^{−40} bar. Therefore, near the surface of the Al–Mg alloy melt, the O₂ partial pressure is so low that the melt will react with N₂ and form a nitride rather than an oxide of the Al–Mg–O system. At 800 °C, with respect to MgO, the formation of Mg₃N₂ is thermodynamically favored at an oxygen partial pressure of 10^{−39} bar or less whereas, with respect to Al₂O₃, AlN formation is favorable even at a relatively higher oxygen partial pressure of approximately 10^{−30} bar. Thermodynamically,

the formation of AlN is more favorable than Mg₃N₂. Schweighofer and Kudela⁷ have studied the high-pressure (1–4 MPa) nitridation of Al–Mg alloys. As a function of N₂ pressure and Mg content of the alloy, they observed two regimes of nitridation: (i) surface nitridation and (ii) volume nitridation. In surface nitridation, AlN formed on the surface of the melt and the reaction of Al with N₂ was confined to the surface and did not extend into the bulk of the melt. However, with increasing pressure of N₂ and Mg content, the nitridation reaction at the surface changed to the bulk. They did not notice any Mg₃N₂ formation in the melt. Therefore, the above arguments support the hypothesis that the surface of the melt must be covered with a thin layer of AlN during infiltration under open conditions in N₂.

In sealed conditions, there is the possibility that under the conditions of extremely low oxygen and nitrogen partial pressures the Al₂O₃ reinforcement may react with Al melt to form aluminium suboxides such as Al₂O, AlO and AlO₂, which will be in the gaseous state at 800 °C. Among the three suboxides of Al, Al₂O formation is favored thermodynamically, and it may increase the partial pressure of oxygen inside the preform by being a source of oxygen. It has been reported in the literature on the measurement of contact angle of Al on ceramic substrates that the alumina layer on the molten Al droplet is disrupted by the formation of Al₂O due to the reaction of the molten Al with the overlying alumina layer under high vacuum conditions and at temperatures above 1000 °C.⁸ Thus, under the high vacuum conditions prevailing inside the preform, Al₂O may react with the Mg and form MgO on the melt surface in sealed conditions. The above explanation at this stage is speculative because direct evidence for the existence of decomposition or reaction products is not available. However, it is also true that despite the low gas pressures infiltration does not display the speed that would be expected of a truly nonreactive combination of liquid and particulate as seen, for example, in the case of water into sand or Cu into Fe powder compacts.

Similar arguments apply to the argon atmosphere as far as the evaporation of Mg and gettering of residual oxygen is concerned. However, because argon does not react with Al–Mg alloy melt the melt surface must have been free of any oxide layer. Aghajanian *et al.*³ reported that infiltration occurred up to a nitrogen content of 10%, whereas no infiltration occurred below 10% of N₂ when a mixture of argon and N₂ was used as the atmosphere. Clark *et al.*⁹ reported that no infiltration took place in argon atmosphere. They attributed it to the difference in the reaction products on the surface of the melt in N₂ and argon. In the present case, discoloration of the preforms due to the coating of Mg evaporated from the melt surface shows that gettering of residual oxygen in argon was effective and the reason for the absence of infiltration

cannot be attributed to the formation of an impervious alumina layer. Indeed, the discoloration of the alumina particles is consistent with their reduction by Mg to form elemental Al.

Thus, there is no consistent difference in the likely reaction products formed on the melt surface in different atmospheres that can explain the role of atmosphere in pressureless infiltration. As a next step in understanding the role of atmosphere, the physical phenomena happening at the melt/preform/gas interfaces are analyzed. In fact, whatever chemical reactions occur at the melt/preform/gas interface will ultimately exhibit their effect through the physical properties such as surface tension (γ_{LV}), solid-liquid (γ_{SL}), and solid-vapor (γ_{SV}) interfacial energies and viscosity. The wetting of a ceramic by a molten metal is assessed based on the contact angle between the ceramic and the metal. According to the Young-Dupre equation, under the conditions of mechanical equilibrium, for a liquid on a solid substrate,

$$\gamma_{SV} - \gamma_{SL} = \gamma_{LV} \cos\theta$$

The difference ($\gamma_{SV} - \gamma_{SL}$) is the driving force for the spreading of a molten metal into a ceramic porous preform. The contact angle of Al on Al₂O₃ was reported^{8,10-12} to range from 90° to 170° at 900 °C. This large scatter in contact angle has been attributed to the surface oxide layer formed on the molten Al, which inhibits the spreading of the molten Al droplet.¹³⁻¹⁶ It has been shown^{5,17} that addition of Mg to Al makes the overlying oxide layer nonprotective thereby bringing about a true contact between Al and Al₂O₃. It was reported¹⁸ that the surface tension value of aluminium saturated with oxygen is lower than that of the unsaturated aluminium. So, it can be expected that the contact angle is more in argon compared to N₂ or air because the surface tension of Al is more in argon because of the fact that the melt is free of reaction products. However, no information is available on the solid-liquid interfacial energy of Al/Al₂O₃ in N₂ or argon. In the Cu/Al₂O₃ system, it was reported¹⁹ that Cu wets the Al₂O₃ in the presence of dissolved O₂ and the marked improvement in wetting was attributed to the decrease in solid-liquid interfacial energy with dissolved O₂. So far, there are no reports on the effect of N₂ on the solid-liquid interfacial energy of Al/Al₂O₃. Therefore, it is difficult to say whether it has any effect on the contact angle. The wetting behavior can also be assessed by inferring the contact angle or solid-liquid interfacial energy from the equilibrium height of infiltration. The contact angle can be calculated from the equilibrium height using the following equation:

$$\rho g H_e = (2 \gamma_{LV} \cos\theta)/r$$

where ρ is density of the liquid metal and H_e is equilibrium height of infiltration. It was not possible to measure the contact angle of Al-Mg alloys on alumina from the

equilibrium height measurements because the equilibrium height for Al-2% Mg, which contained the lowest amount of Mg of all the alloys used, is higher than the maximum height of the preform (i.e., 10 mm) that can be handled in the experimental apparatus used in the present work.

From the foregoing analysis, it can be concluded that the difference in the reaction products on the surface of the melt in N₂, air, and argon cannot explain the marked difference in the infiltration phenomenon in these atmospheres. However, it indicates that a critical amount of N₂ is necessary for the pressureless infiltration to occur, even though that amount is unlikely to persist after the preform is isolated and infiltration begins.

V. CONCLUSIONS

It has been demonstrated that commercial purity Al can be infiltrated into Al₂O₃ preforms in air at 800 °C, without the application of pressure, by placing a small amount of Mg at the billet/preform interface. By using this new technique a number of Al alloys such as Al-Si, Al-Zn, Al-Mg, and combinations thereof have been infiltrated into the Al₂O₃. Infiltration studies in different atmospheres indicate that a critical amount of N₂ in the original atmosphere is necessary for the pressureless infiltration of Al alloys. The rate of infiltration increases with nitrogen partial pressure.

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APPENDIX

Calculation of infiltration height using the Washburn equation

Height infiltrated by a liquid into a capillary (horizontal or early stages of ascent, i.e., $h \ll h_{\max}$) is given by the following equation, which is from Washburn:²⁰

$$(h^2/t) = (r\gamma_{LV} \cos \theta)/2\mu \quad (A1)$$

where r is capillary or pore radius, γ_{LV} is surface tension, θ is contact angle, μ is viscosity of the liquid, and t is the time taken to infiltrate a height h .

The height infiltrated by Al–5% Mg alloy into a 53- μm alumina preform is calculated using the above equation. Surface tension data were taken from the work of Goicoechea *et al.*,²¹ viscosity data were taken from the work of Gebhardt *et al.*,²² and contact angle data were taken from the work of Weirauch.¹³ Pore size of the preform was estimated using image analysis: $\gamma_{LV} = 0.782 \text{ N/m}$, $\mu = 1.03 \times 10^{-3} \text{ N s/m}^2$, $\theta = 85^\circ$, $r = 12 \mu\text{m}$, and $t = 30 \text{ min}$.

By inserting the above data in Eq. (A1), $h = 84.5 \text{ cm}$ whereas experimentally determined infiltration height is 0.43 cm. The two values differ by two orders of magnitude. In order to reconcile the discrepancy the contact angle would have to be increased to 89.99985° .